DEVELOPMENT OF A NOVEL COUPLED-MODE FLUTTERING ENERGY HARVESTER THROUGH ELECTROMAGNETIC COUPLING AND DIELECTRIC ELASTOMER GENERATORS

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a Giulia e Iris
Thesis abstract

The topic of the research is the development of a novel energy harvester exploiting fluids in motion. FLEHAP device (Fluttering Energy Harvester for Autonomous Powering, patent pending), conceived and developed at DIFI of University of Genova, is an aeroelastic flutter-based system of an airfoil exploiting an electromagnetic coupling (EMc) and smart materials to extract electrical energy from the wind. It is performed the experimental study of the aeroelastic system through different prototypes, highlighting the main operating parameters and their correlations, analyzing the kinematics and the fluid dynamics aspects. The design of Dielectric Elastomer Generator materials (DEGs), their realization, characterization and application in the FLEHAP device are carried out. Electrical conversion and storage solutions are investigated for both the EMc and DEGs. The topics discussed are developed in terms of Research and direct technological application.
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Introduction

Context

Transportation, manufacturing, health care, environmental monitoring, safety and security applications, for which the wiring and its maintenance are too expensive or technically not feasible, find the solution in the Wireless Sensors Networks (WSN) or Internet of Things (IoT)\textsuperscript{1,2}, distributed sensor nodes that need an autonomous powering. Energy Harvesting (EH), also known as power harvesting or energy scavenging, is the process by which energy is derived from external sources (e.g. solar power, thermal energy, wind energy, salinity gradients, and kinetic energy, also known as ambient energy), typically directly in the place where the energy is needed. EH finds an interesting application in the stand-alone applications, as life-time and costs of batteries still require further technological development\textsuperscript{3,4}.

On the other hand, the global efficiency of the EH devices is strictly connected to the management of both mechanical and the derived electrical energy. In fact, the excitation times and conditions, and the amount of energy stored and requested for a certain technological application, impose the use of specific algorithms to optimize the final duty cycle of the devices.

In the last decades significant efforts have been devoted in the develop of EH devices\textsuperscript{5}. The reliability, the efficiency and the costs of the solutions proposed, often suffer a lack of practical and technological applicability, while where these conditions are satisfied, socio-economical and political aspects have been limited their development on the market (see traffic in the city, monitoring of buildings, sources and services, health control, fire emergencies, etc.). Because of its wide availability, fluids in motion represent interesting sources for the scavenging purposes.

Our life is surrounded by fluids in motion: the mean of the wind speed revealed in cities is typically between 3 and 6 [m/s], the air conditioned, largely diffused in the indoor spaces, are typically between 1 and 4 [m/s], then the gas and the oil pipelines, currents of the sea in offshore (on the surface or on the sea bed) and during tides, high-altitude wind, and last but not least, every vehicle which is moving produces a relative velocity available for inducing a self-sustained excitation, and being exploited for harvest energy.

FLEHAP device (Fluttering Energy Harvester for Autonomous Powering, patent pending), conceived and developed at DIFI of University of Genova, is a novel aeroelastic flutter-based system of an airfoil exploiting hybrid solutions for extract electrical energy from the wind. The solutions proposed consist in an electromagnetic coupling (EMc) composed by moving coils and permanent magnets, and soft smart materials (Dielectric Elastomer Generators, DEGs) used for generate electrical energy during their periodical deformation in the aeroelastic system.
Objectives and structure

The current Ph.D. thesis is a continuation of the seminal investigation of my Master Degree work, and it aims to develop a novel aeroelastic system for extract electrical energy from fluids in motion. The Research project involved multidisciplinary activities: the optimization of the mechanical energy absorption, through the aeroelastic system, the optimization of the electrical energy conversion, through the two main strategies explored (EMc and DEGs), the investigation of electrical storage solutions, and the prototyping.

The objectives of this thesis are:

• To introduce the basic concepts of the Fluttering, a particular Aerelasticity phenomena.
• To present the aeroelastic system proposed, providing an overview of the different types of response observed in the experiments, introducing some predictive models, and posing some considerations for the maximization of the performance.
• To optimize the system exploiting an Electro-Magnetic coupling for extract electrical energy from the mechanical one, from the energy extraction to the energy storage.
• To perform the study, the realization and the characterization and of Dielectric Elastomer Generators (DEGs), in order to directly convert the mechanical energy provided by self-sustained oscillation experienced by the aeroelastic system in electrical one, focusing on the performance needed for the application in study.
• To present and characterize a system exploiting DEGs and EMc, providing a Proof of Concept of the hybrid device.
• To illustrate the possible future studies related to the system.

In order to fulfil these objectives, the Ph.D. thesis is organized as follows. Chapter 1 provides some preliminary concepts useful to get familiar with the aerelasticity problems involved in the Research. Chapter 2 presents the experimental setup used for the aerelastic analysis. It follows Chapter 3, in which the device in study is illustrated; it includes some models developed for describe and predict the behaviour of the aeroelastic system, a multi parametric experimental analysis and a summary of the not usual evidences observed during the self-sustained oscillations. Chapter 4 illustrated the studies of the systems equipped with EMc, the study of the effect of electrical extraction on the mechanics of aeroelastic system, the comparison between the use of commercial integrated circuits and the design of a specific electronic, and the placing of the device in study in the framework of the international performances of similar devices. Chapter 5 deals with the study of DEGs, from their operational conditions, to the synthesis and characterization of soft elastomeric composite, to the design of the stretchable electrodes and a simple and reliable procedure for their realization, to the exploration of dedicated electronics. To end, the conclusions summarize the main findings of this thesis.
In the thesis the variables are shown in italics (e.g. mass is $m$), the units of measurements are included in square brackets (e.g. gram is [g]), the acronyms are capitalized (e.g. Energy Harvesting is EH), and the references are enumerated, placed in apex, and collected at the end of the thesis in the References part.
1 Aeroelasticity

Aeroelasticity is the branch of physics and engineering that studies the interactions between the inertial, elastic, and aerodynamic forces that occur when an elastic body is exposed to a fluid flow. Although historical studies have been focused on aeronautical applications, and general failure prevention, recent research has found applications in fields such as energy harvesting and physiological aspects. The study of aeroelasticity may be broadly classified into two fields: static aeroelasticity, which deals with the static or steady response of an elastic body to a fluid flow, and dynamic aeroelasticity, which deals with the body’s dynamic, typically vibrational, response. The term aeroelasticity was coined in the early 1930’s by A. Pugsley and H. Roxbee Cox, two engineers at the British Royal Aircraft Establishment (RAE) whose efforts considerably advanced the knowledge of aeroelasticity, its consequences and its cures. In 1947, A. R. Collar defined aeroelasticity as "the study of the mutual interaction that takes place within the triangle of the inertial, elastic, and aerodynamic forces acting on structural members exposed to an airstream, and the influence of this study on design". Aeroelasticity therefore draws on the study of fluid mechanics, solid mechanics, structural dynamics and dynamical systems. The interaction of these interdisciplinary activities can be summarized by a “three-ring” Venn diagram, shown in Figure 1.

![Aeroelasticity Venn three-rings diagram.](image)

In the development of aeronautical engineering at Caltech, the pioneer Theodore von Kármán started a course "Elasticity applied to Aeronautics". E.E. Sechler then developed aeroelasticity in the same course, and in publication of textbooks on the subject.

Aeroelasticity is still an open issue in many sectors of unsolicited vibrations, such as aerospace, biomimicry, design of buildings, and many other complex systems.
1.1 Fluttering

This paragraph aims at introducing the fluttering effect, followed by a description of classical flutter, stall flutter and limit cycle oscillations (LCO), in the field of Dynamic Aeroelasticity, controversy topic in Physics and Engineering. The reader is referred to Dowell et al. \(^8\) for a complete, comprehensive discussion on flutter. Nevertheless, a brief introduction, reported from Ref.\(^9\), will try to discriminate the similar, but different, dynamic aeroelasticity conditions.

A typical Fluttering problem consists in the dynamic instabilities occurring on an aircraft wing exposed to a uniform flow during the flight. Despite the multiplicity modes with which the solid could deform, typically in concomitance, such as bending, twist, etc., a common outline of the physical problem is drafted in the Figure 2.

Here, a section of wing of Chord \(C\) (length between the Leading Edge \(LE\) and the trailing Edge \(TE\)) exposed to a flow of speed \(U\) is represented with its two main degrees of freedom (DOF), named in this case \(\alpha\) and \(y\): \(\alpha\) is the angular displacement, and corresponds to the so called pitch mode, while \(y\) is the vertical displacement, called plunge mode. The system is schematically represented with two springs, \(k_y\) and \(k_{\alpha}\), related to the two DOF, and a damper \(D\). Similarly to the aerodynamic static case, the resulting forces act from a specific location of the wing, called aero-dynamical centre, which is typically around \(C/4\); however, this value could change with shape of wing or flow conditions \(^11\). The two primary forces in aerodynamics are Lift and Drag. Lift refers to (usually upward) forces perpendicular to the direction of motion of an object travelling through the air, while Drag forces are parallel and opposite to the object's direction of motion and are caused largely by friction.

Defining the free-stream dynamic pressure as \(\frac{1}{2} \rho U^2\) and \(A = C S\), the characteristic body area, where \(S\) is the span of the wing, the lift force depends on the attitude and the shape of the body to deform the fluid, called Lift coefficient \(C_{L}\); then,
This is the fundamental Lift formula for usual aircraft flight. It is very important to realize that the Lift coefficient $C_L$ is a number dependent upon the so called Reynolds number, surface roughness, air turbulence, attitude, and body shape. It is not by any means a constant. $C_L$ is generally found by wind tunnel or flight experiments by measuring Lift and the free-stream conditions varying the pitch angle and having a knowledge of the body dimensions. This is typically led in static cases. Reynolds number is the dimensionless quantity associated with the fluid viscosity and it is defined as $Re = \rho U C / \mu$, where $\rho$ and $\mu$ represents the density and the kinematic viscosity of the fluid. One obtains analogous equations to previous of Lift for the Drag:

$$D = \frac{1}{2} \rho A U^2 C_D$$

(2)

The moment acting on a body is a measure of the body's tendency to turn about its centre of gravity. This moment represents the resultant aerodynamic force times a moment distance. Let it be stated that a similar derivation may be applied to the moment equation as used for Lift and Drag, such that

$$M = \frac{1}{2} \rho A l U^2 C_M$$

(3)

$C_M$ is the coefficient of moment and an additional characteristic length $l$ is necessary for it to be dimensionally correct. To reiterate, $C_L$, $C_D$, and $C_M$ are dependent on the Reynolds number, surface roughness, air turbulence, attitude, and body shape.

Such forces and moments are the causes and the effects of the coupling with the fluid. The modification of the upward free stream by the motion of the body consists in the formation of pressure fluctuation, which results in a leading edge vortex when the pitch angle grows. Its detachment from the body (called stall) depends on the geometry and operational conditions, and during the fluttering it typically occurs at higher pitch angles (more than 40 [°]) with respect the static cases (around 12-15 [°] for symmetric NACA airfoils).

1.2 **Classical Flutter**

Classical flutter is a type of dynamic instability in which the amplitude of motion may drastically grow following an external perturbation of some kind. In the case of a streamlined body, such as a wing, this external perturbation could be any symmetry-breaking mechanism: turbulence, atmospheric wind gust, detection of a control surface, etc. The loss of stability is traditionally explained with the concept of negative aerodynamic damping. As the free stream velocity ($U$) of the flow to which the airfoil is exposed increases, the aerodynamic damping first increases. In other words, the motion induced by an external perturbation is damped (i.e., the kinetic and the potential energy is transferred from the structure to the flow). Further increasing the free stream velocity ($U$) may result in a rapid decrease of the aerodynamic damping. At a
critical flutter velocity \( (U_{cr}) \), the aerodynamic damping becomes null, and any further increase of the free stream velocity \( (U) \) leads to a negative aerodynamic damping. As a result, when this critical velocity is passed and the airfoil is disturbed from its equilibrium position through a perturbation of any amplitude, the aerodynamic forces are such that the motion is not damped, and the flow provides energy to the structure, resulting in some possibly violent, exponentially-growing oscillations. Structural failure is very often the outcome to such an event, which strongly suggests that the typical objective of an aeroelastic study is to avoid such detrimental instabilities.

The use of the term classical flutter is normally restricted to an oscillatory instability observed in an incompressible potential flow (this assumes the flow to be irrotational and thus inviscid). This implies that nonlinearities are absent from the flow (i.e., neither separations of flow nor shocks are present) or, at least, are not involved in the physical mechanisms through which the instability occurs. Nevertheless, few real cases admit this assumption in air.

Although one-degree-of-freedom classical flutter has been observed, and the linear dynamic instability is generally the result of a well-defined coupling between the various degrees of freedom of the structure, with the phase shift between motions being a key parameter.

1.3 Stall flutter

Stall flutter, as explicitly revealed by its name, involves separations of the flow from the streamlined body undergoing flutter. These separations of the flow may either be complete or partial, and, unlike classical flutter, stall flutter may reveal not to be catastrophic. In fact, due to the presence of aerodynamic nonlinearities, the outcome is very often a cyclic motion of limited amplitude (see Arena et al. \(^1\), Dimitriadis and Li \(^2\) and Razak et al. \(^3\)). This phenomenon is known as limit-cycle oscillations (LCO), and this will be the subject of the next sub-section. Even if the amplitude of motion usually stabilizes itself before structural failure occurs, the large-amplitude, cyclic motion may sometimes result in a significant impairment of the service life of the device due to fatigue (see Arena et al. \(^1\)).

For this reason, it is generally better to avoid stall flutter over the complete operational range of a device, or, at least, to drastically limit the amplitude of motion resulting from it.

Figure 3 - Illustration of Stall, phenomenon in which a separation of the flow from the streamlined body occurs with the formation of vortex. From \(^4\).
The physical mechanisms involved in stall flutter greatly differ from those observed in classical flutter. This time, the transfer of energy from the flow to the structure is not the result of elastic or an aerodynamic coupling between the degrees-of-freedom involved, and the phase lag between the motions is not a parameter of importance. It must be understood that some level of coupling between the various degrees-of-freedom, along with a lag between the motions, may be present and alter the dynamics, but they are not essential features of stall flutter. In fact, stall flutter is possible as a result of the nonlinear behaviour of the aerodynamic forces on the body, and these nonlinearities are the result of separations occurring in the flow\textsuperscript{13-15}.

It is well known that separations occur when an airfoil is undergoing a large-amplitude, periodic motion in a free stream flow. When this happens, there is a time lag in the aerodynamic forces and moments exerted by the flow on the body, along with a hysteresis in the aerodynamic forces. These two phenomena combined, in turns, may give rise to a negative aerodynamic damping, thus implying that the flow is doing some work on the structure. As a result of this positive transfer of energy from the flow to the airfoil, flutter may occur if the structural damping is insufficient. The occurrence of stall flutter is very often dependent upon the amplitude of the initial external perturbation, especially if the airfoil is linearly dynamically stable. This is in contrast with classical flutter, where an infinitesimal perturbation is enough to induce flutter. For stall flutter, if the initial external perturbation is large enough, the dynamic instability may even be observed at flow velocities well below the critical flutter velocity as predicted through a linear theory\textsuperscript{13}, such as the one presented in the previous section.

Further, stall flutter implies that separation occurs on a cyclic basis. Indeed, separation must be followed by a reattachment of the flow to the airfoil\textsuperscript{12,14,15}. For instance, in the galloping of a bluff body the flow is always separated, and there is no reattachment of the flow\textsuperscript{14,15}. Galloping is a very different from stall flutter, as the motion of the body consists only in plunge; if also the torsion mode is exhibited, then a similar vortex shedding phenomena is observed.

Dynamic stall, the mechanism that accounts for the instability, is the abrupt loss of Lift and/or pitching moment observed as a result of a flow separation over an airfoil undergoing motions of large amplitude\textsuperscript{14,16}, and it is well documented in the literature. This unsteady, nonlinear aerodynamic phenomenon may be further subdivided based on the level of flow separation, in light stall and deep stall (see McCroskey 1981\textsuperscript{12}, 1982\textsuperscript{17}). The airfoil is said to be undergoing deep dynamic stall when the main feature of the flow is the shedding of a large vortex at the leading edge. In such situation, the viscous layer can be as thick as the airfoil's chord length. For deep dynamic stall to occur, a motion that induces pitch angles much larger than the pitch angle needed to reach static stall is required. This large vortex traveling close to the airfoil, at an approximate velocity of $U/2$, can generate boosted Lift and aerodynamic moments that are far in excess from their static counterparts. Conversely, light dynamic stall is more similar to static stall. Trailing-edge separation is encountered, and the thickness of the viscous layer is of the order of...
the airfoil's thickness. The Lift and the moments remain closer to the typical values observed during static stall. Typically, light dynamic stall is observed when the maximum pitch angle is only a few degrees greater than the pitch angle at static stall. It must be mentioned that stall flutter may occur under both types of dynamic stall.

As previously mentioned, negative aerodynamic damping arises as a result of the phase lag between the motion and the aerodynamic forces, combined with a hysteresis in the aerodynamic forces. The large hysteresis, shown with the gray area in Figure 4a and 4b and the phase lag are associated to the cyclic detachment and reattachment of the flow involved in dynamic stall. In the light dynamic stall regime, the lag and the hysteresis are moderate, which implies a potential for some negative aerodynamic damping. For the deep dynamic stall regime, there is a very large hysteresis, which may lead to a considerably more negative aerodynamic damping, thus implying a larger potential for stall flutter.

Returning to deep dynamic stall, the effect of the shed vortex is particularly evident when considering the behaviour of the pitching moment, which exhibits an abrupt and strong change of amplitude.

Figure 4 - Typical Lift coefficient $C_L$ (a) and aerodynamic moment coefficient $C_M$ (b) for a static airfoil (dashed red line, maximum at 12-15°) and for an airfoil undergoing deep dynamic stall (solid line). In c) hysteresis in the free response of the airfoil increasing and decreasing the wind speed $U$. From 9,12.
The solid curve of Figure 4 shows the typical behaviour of the Lift coefficient \((C_L)\) and the aerodynamic moment coefficient \((C_M)\) as the pitch angle is varied, while the dashed curve shows the trends for static angles. Large differences between the static and the dynamic trends, along with a hysteresis for both \(C_L\) and \(C_M\) (gray areas), are observed. The dots identified with numbers correspond to the typical qualitative sequence of events described in Table 1. As it may be seen, the shedding of the vortex plays a crucial role in the aerodynamics. In fact, stall flutter is sometimes seen as the result of a coupling between the structure and the flow achieved through vortex shedding. This is because the low pressure in the core of the shed eddy has the potential of generating fairly large forces on the surface of the airfoil as it travels downstream.

Another typical nonlinear behaviour of stall flutter is the hysteresis in the response of the airfoil (not to be confused with the hysteresis of the aerodynamic forces previously discussed). A qualitative example is provided in Figure 4c. When a critical parameter is varied, here the free stream velocity \((U)\), a sudden appearance of flutter with a brutal jump to large amplitudes of motion is observed at a critical velocity, say \(U_{cr}\). Then, if the velocity is reduced, flutter may persist down to a value well below \(U_{cr}\). The amplitude of motion at a specific velocity may even differ depending on the fact that the velocity is being increased or it is being decreased. Memory effects are therefore inherent to the stall flutter phenomenon.

Recalling that a potential aerodynamic theory has been used in the discussion on classical flutter, one could seek a similar method in order to study stall flutter. In light of all the nonlinear characteristics involved within stall flutter, this appears to be unrealistic. Indeed, potential theories do not aim at predicting the aerodynamic forces exerted on a body when separations are encountered, and, as a result, they fail at predicting stall flutter. To illustrate this statement, stall flutter has been observed at flow velocities well below the critical flutter velocity \((U_{cr})\) predicted with the linear theories. In order to study the stability of an airfoil when compared to periodic stalling, semi-empirical stall models have to be used, such as the well-known Leishman-Beddoes (see Leishman and Beddoes 1989) and the Onera (see Tran and Petot 1981) models.

<table>
<thead>
<tr>
<th>Point</th>
<th>Main flow feature</th>
<th>Forces and Moments</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Thin and attached boundary layer</td>
<td>Linea regime</td>
</tr>
<tr>
<td>2</td>
<td>Flow reversal within the boundary layer</td>
<td>Linear and maximum static Lift exceeded</td>
</tr>
<tr>
<td>3</td>
<td>Vortex shed and moves over the airfoil</td>
<td>Pitching moment diverges and Lift due to the vortex is present</td>
</tr>
<tr>
<td>4</td>
<td>Vortex continues downstream at approximately (U_{cr}/2)</td>
<td>Maximum Lift and moment followed by a rapid decay</td>
</tr>
<tr>
<td>5</td>
<td>Secondary vortex forms</td>
<td>Secondary peaks on both Lift and moment</td>
</tr>
<tr>
<td>6</td>
<td>Reattachment of the flow</td>
<td>Return to linear regime</td>
</tr>
</tbody>
</table>

Table 1 - *Typical qualitative sequence of events during the deep dynamic stall of an airfoil, from* 9.
As pointed out by Larsen et al.\textsuperscript{19} the objective of these models is essentially to capture the main characteristics of the aerodynamics in a fast and efficient way. That being said, this may prove to be sufficient to conduct the stability analysis of a device, or to roughly estimate the amount of energy harvested from a flow when the motion of the airfoil is enforced, such as in the work of Bryant et al.\textsuperscript{20}. However, it may fail at predicting accurately the motion of a fully-passive device. Indeed, in the fully-passive case, the motion depends upon the aerodynamics, and the aerodynamics depends upon the motion. This interdependence may lead to an inevitable amplification of the discrepancies inherent to such a model, the behaviour of which is dependent upon an adequate calibration that is case dependent.

![Typical qualitative response over time of a dynamically unstable structure that involves nonlinearities leading to limit-cycle oscillations (LCO) of constant amplitude. From\textsuperscript{9}.](image)

**Figure 5 -**

1.4 **Limit-cycle oscillations**

Limit-cycle oscillations (LCO) are self-excited, cyclic oscillations of limited and fairly constant amplitude\textsuperscript{14}. For this type of motion, which could easily be characterized as being well-behaved, the amplitude of motion happens to be limited by some nonlinearities present in the dynamics of the system\textsuperscript{13}. The typical qualitative response of a dynamically unstable structure that involves nonlinearities leading to LCO is shown in Figure 5. Although the representation may suggest that only one degree-of-freedom is involved, LCO may in fact consist in the simultaneous excitation of several modes. In the first stage shown in Figure 5, the structure is in static equilibrium and free from any significant motion other than vibrations of very small amplitude (not shown). Then, following an external perturbation that can take the form of any symmetry breaking mechanism, the structure becomes unstable and is subject to an oscillatory motion of growing amplitude throughout the transition regime. As the amplitude grows, the effect of the
nonlinearities becomes greater (e.g. flow separations may appear). This turns out to limit the growth of the amplitude of motion until the instability finally saturates, and a permanent LCO regime of constant amplitude is reached. As previously discussed, the nonlinearities involved in LCO may be due to stall flutter. In this specific case, they are caused by large separations of the flow from the airfoil. However, LCO may emerge from several other sources of nonlinearities. Indeed, the nonlinear behaviour introduced by large shocks in a flow or by structural (e.g., free play in control surfaces), material (e.g., large deformations) or inertial (e.g., concentrated masses) nonlinearities may be sufficient to limit the exponential growth of a fluttering airfoil's amplitude of motion \(^{13,15,21}\). Also, as pointed out by Dowell et al. \(^{21}\), LCO is not the only possible outcome to flutter when nonlinearities are involved. Other types of response, such as beating and period doubling, may sometimes occur. These behaviour are also interesting for a fully-passive fluttering-airfoil the energy harvesting purposes application, different to what is reported in Ref.\(^9\).

As pointed out in the previous section of stall flutter, a linear stability analysis does not permit to predict the onset of stall flutter due to its nonlinear behaviour. As it might be expected, the same is true for most types of LCO. In fact, a linear analysis generally predicts fairly well the frequency of oscillation, but it fails at predicting the onset of the instability or its terminal amplitude of motion. Since one involved in such work generally seeks a flutter boundary in order to avoid the instability, this type of study is not worth the trouble. Instead, some semi-empirical or empirical models can be developed to help study a specific type of system. Nowadays, computational fluid dynamics (CFD) is also used to study LCO. From CFD and experimental work, the general trend of the amplitude of an LCO is generally to increase as the flow velocity increases, and hysteretic responses, such as the one depicted on Fig. 4c, are very often observed.

Several key parameters affect the location of the flutter boundary, such as the distribution of mass, the location of the elastic axis, the location of the aerodynamic center, the natural frequencies and the elastic configuration. The dynamic instability of stall flutter differs greatly from classical flutter in the physical mechanisms involved. During classical flutter, the negative aerodynamic damping arises from the elastic and/or aerodynamic coupling between the modes. For stall flutter, the negative aerodynamic damping is due to the hysteresis in the aerodynamic forces and the phase lag between the motion and the forces. These two phenomena are the result of the highly unsteady flow and the dynamic stall encountered. Very often, the instability saturates through the nonlinearities involved in the massive separations present. This limits the growth of the amplitudes of motion, thus leading to a well-behaved, cyclic motion called limit-cycle oscillations (LCO). Although stall flutter often results in LCO, this type of response may also be due to other physical mechanisms. For the purposes of energy harvesting, LCO are the typical outcome of interest due to its fairly constant amplitude of motion.
2 Experimental setup for the aeroelastic investigation

In order to carry out the studies on aeroelasticity problems a common experimental setup is typically used. It includes a flow-controlled wind (or water) tunnel, force sensors and accelerometers, ultra fast camera acquisition system, and eventually flow visualization apparatus, such as PIV (Particle Image Velocimetry) or smoke. This chapter describes the apparatus utilized for understand, characterize and optimize the aeroelastic system explored for harvest energy from the airflow.

2.1 Wind tunnel

An aspirating subsonic open circuit wind tunnel, designed and assembled in Lab.206c of DIFI during my Master degree thesis, was used to characterize the devices from the aeroelastical point of view. It is composed by a three-phases motor, placed in the end part of the tunnel, named diffuser (on the left side of Fig. 6), a test chamber of dimensions of 70×40×40 [cm$^3$], in the center of which the devices are placed, and a frontal inlet, properly designed for compressing the flow in a quasi-laminar condition, named Nozzle. The latter is equipped by a honeycomb grid able to break the inlet turbulences. A free-stream turbulence level around 0.6% was detected using a Cobra probe. With the same instrument also a calibration of the wind speed inside the test chamber, relatively to the voltage applied to the motor, was led and confirmed by a video acquisition of the flow visualization. The conversion obtained by such measurements was $c^*=1.62 \ [m \ s^{-1} \ V^{-1}]$.

The wind tunnel used allows obtaining laminar flow in a wide range of speed, from 1.6 to 16.0 [m/s], while the maximal dimensions of the devices characterized in the test chamber are 25×20×15 [cm$^3$]. The latter are positioned on the middle of the test chamber with the use of sharp and rigid supports.

Figure 6 - Subsonic aspirating wind tunnel in DIFI utilized for the aeroelastic analysis.
2.2 **High speed camera**

The high speed camera Fastec IL3100-SM4, showed in Fig. 7a, was placed externally to the test chamber and orthogonal to its side-part. It is characterised by a maximal resolution of 1280x1024 [pixel], maximal frame rate of 510 [Hz], and a digital shutter parameters between 2 [μs] and 41.7 [ms]. These latter are very important parameters for a precise acquisition of the motion parameters.

2.3 **Three axis balance**

For a direct acquisition of the forces acting on the device during the fluid-solid interaction, a 3-axis balance 3A6010N-D11, produced by Interface Advanced Force Measurement, and showed in Fig. 7b, was used. This instrument is able to provide at the same time the values of the forces in the main three directions, with a maximal sample frequency of 205 [Hz], a maximal capacity of 10 [N] and sensitivity of $10^{-4}$ [N]. The devices are typically connected by two screws to the load cell transducer (in the center of the balance, reported in Fig. 7b), while the load cell is fixed to a support of dimensions that permits the positioning of the device in the center of the test chamber, avoid the induced vibrations of the support, and at same time avoid a relevant shape impact of the flow. In fact, for these measurements, a $15 \times 8 \times 3$ [cm$^3$] wood column was used as support: the frontal surface with respect the incoming flow consisted in $3 \times 15$ [cm$^3$], compared to $40 \times 40$ [cm$^3$] section of the test chamber.

A typical measurement of forces on a fluttering device consisted in two steps: first, a measurement of the contribution of the frame was performed by increasing the wind speed on the device without the wing, fixed to the load cell and to the main support in the test chamber. This step is crucial also to evaluate eventual induced Lift or Drag caused by the supports. The values related to 0 [m/s] were subtracted to all the relative wind speed, while all measurements had normalized to the zero of the balance. Second, the measurements of forces when the wing is free to interact with the fluid were carried out following the procedure of the first step, and then the frame contribution was subtracted to them. The forces acting on the system can be thus evaluated.

![Figure 7 - High speed camera used for the video acquisition of motions (a), and 3D load cell, supplied by Interface AFM, used to measure the forces acting on the systems (b).](image)
2.4 **Smoke generator apparatus**

Inside the test chamber flow visualization was possible by the use of a smoke generator, modified for our purposes. A peristaltic pomp inserts, drop by drop, medical oil in the upper-front part of the test chamber, where a metallic wire is placed orthogonal to the flow, in tension thanks to a bottom spring, and heated by Joule effect due to the dissipation of electrical current applied. A white led light is placed on the top of the test chamber and was used for light the flow vanes. The frequency of the oil drop delivery is regulated by the change in voltage of a first voltage generator to the pomp; the drops fall down for the gravity. An electrical dissipation of the wire is controlled by another voltage generator, which is responsible of the evaporation of the oil on the wire, but also of the falling speed of drops. In fact, if on one hand the electrical energy dissipation heats the wire, permitting the evaporation of the oil, the flowing fluid (air in motion) cools it, reducing the intensity of the visualization, composed by white ‘smoke vanes’ on a dark background: as higher the wind speed is, more electrical dissipation on the wire is necessary to permit a relevant evaporation of the oil, but at the same time a decrease of the viscosity of the oil by the temperature of the wire reduces the time of the visualization. This is because each single oil drop leaves an amount of little drops on the wire proportional to its falling time, depending to the oil viscosity. For example, if one increases the dissipation on the wire, the ‘smoke vanes’ will be thicker but also less in amount, as the main drops will evaporate in the first part of the wire, and so, the portion of flow visualized will be less. This problem is theoretically solvable by the use of a metallic wire with larger diameter, but this will increase the induced turbulences due to a too big Reynolds number, causing a non-2D disturbed flow. Consequently, a difficult trade-off must be found out to obtain a good visualization of the air flow for an extended speed range.

![Figure 8 - Vortex shedding created by cylinders ('bluff bodies') of different diameter: 1, 2, and 4 [mm].](image-url)
In the experiments conducted, typically in the range of 2-6 [m/s] for the vortex visualizations, 10-40 [W] of electrical power was dissipated on a wire of 0.2 [mm] of diameter with a drop frequency of 0.5-3 [Hz], recorded by camera at 510 [Hz]. The internal sidewall of the test chamber in the background was equipped by a black textured carpet, while a white led light of 2500 [lumen] and colour temperature of 4000 [°K] was located on the top of the test chamber in downstream position and behind the device, seeking the best contrast for the visualization of the smoke wires. Figure 8 reports the visualization of the vortex shedding occurring on cylinders of different dimensions by the use of the described apparatus. A lateral view of the stream, flowing from the right to the left, permits to capture the vertical plane of smoke wires, displaying the mid-cross section of the rods. The periodicity of the wakes is very clear, and appears larger, larger is the characteristic length, in this case the diameters of the cylinders. In the figure, the down pressure vortexes are also painted: they define the so called von Karman vortex street.

In Figure 9, the Alejandro Selkirk Island (before named Isla Más Afuera, the Robinson Crusoe island), off the Chilean coast, is acting like the cylinders previous reported. A vortex street is shown disrupting a layer of stratocumulus clouds which are low enough to be affected by the island, which is nearly 1.3 km above sea level and 1.5 km in length. When the wind-driven clouds run into the obstacle of the island, they flow around it clockwise and anticlockwise to form the beautiful spinning eddies, advecting hundreds of kilometres downwind to make a street 10,000 times longer than those made in the laboratory.

Study of these vortices is very important in the understanding of laminar and turbulent fluid flow; these control a wide variety of real-world phenomena, like the lift under an aircraft wing and Earth's weather. A vortex street can be observed only over a given range of Reynolds numbers \((Re)\), normally above about 90. In fact, as reported in sect.1.1, Reynolds number is a measure of the ratio of inertial to viscous forces in the flow of a fluid; therefore almost all fluid dynamic aspects (and effects) depend on its value.

Figure 9 - Satellite photography of Von Karman vortex street in Isla Más Afuera (Chile).
3 FLEHAP device

The technologically well-developed windmills and micro turbines suffer very low efficiencies at small speeds due to laminar separation effects that subsist at low Reynolds number.

An alternative method to harvest energy from a wind is based on the fluttering instability effect, in which moving bodies are typically connected to elastic materials, for example bluff bodies \(^{23-25}\) and T-shaped cantilever \(^{26}\), or in which the body can be directly deformed by the incoming fluid, for example elastomeric belts \(^{27,28}\) and piezoelectric flags \(^{29}\).

Airfoils, deeply investigated since the early XX\(^{th}\) century in order to understand and optimize the flight, are able to exhibit self-sustained motion when properly coupled to an elastic systems. After the seminal work of McKinney and De Laurier \(^{30}\) in recent years many systems adopting this effect applied to airfoils have been proposed.

A few examples are reported in Bryant et al. \(^{31}\), Zhu et al. \(^{32}\), Fei et al. \(^{33}\), Abdelkefi \(^{34}\), Nabavi and Zhang \(^{35}\), and McCharty et al. \(^{36}\), in which different possibilities for the system activation are studied and discussed. There are basically three kind of activation: (i) the fully-driven devices, in which the motion is totally prescribed \(^{30,37}\), (ii) semi-passive devices, where typically only the pitching mode is driven \(^{38-40}\) and (iii) fully-passive devices, where the motion is totally governed by the resulting dynamics. \(^{38,41-43}\).

These latter are interesting by virtue of their constructive simplicity, since no control mechanisms and actuators are employed, but pose specific challenges regarding the physical comprehension and modelling \(^{44}\).

The device in study, named FLEHAP (Fluttering Energy Harvester for Autonomous Powering, patent pending), conceived and developed in DIFI of University of Genova, is characterized by a fully passive ‘pitch and plunge’ fluttering of an airfoil with any torsional spring. The mechanical system is simply composed by a semi-rigid wing (typically an acetate airfoil of thickness of 0.2 [mm] and density of 1.4 [g/cm\(^3\)]), free to rotate around an axis, intimately connected to one or more elastic elements (e.g. see Fig. 10).

When a fluid interacts with the structure, if the parameters are correctly set, a self-sustained oscillation characterized by a vertical harmonic motion of the axis (Pivot Point PP), and consequently, by a periodical lemniscate motion of the trailing edge (TE) of the wing, is achieved by the system in a wide range of flow speed.

Important characteristics of our system are the elastic arrangements chosen (shown in details in the next section, the global dimensions (few centimetres) and the weight (wing around 1 [g]), which allows us to reach high frequencies during the self-sustained oscillations.
The main mechanical input parameters of the system consist in dimensions \((C, S)\), mass \(m_{\text{wing}}\) and distribution of mass of the wing, mass of translational part \(m_{\text{plunge}}\), effective elastic constant in the direction of the oscillation \(K_{y}^{\text{eff}}\), described in the next sections), mechanical damping \(D\) and wind speed \(U\), while the output can be summarised in PP amplitude \(PPy\), frequency of oscillation \(v_0 \neq v_n = \frac{1}{2\pi} \sqrt{\frac{K_{y}^{\text{eff}}/(m_w + m_p)}{}}\), and phase lag between the rotation (pitch) and the translational (plunge) modes of the wing \(\phi\). The main electrical extraction strategy proposed in this work exploits an electromagnetic coupling (EMc) obtained by placing a pair of coils at the ends of the pivot axis of the wing and two series of permanent magnets in alternated polarity configuration in front of their predicted trajectories (see Fig. 10). Complementary strategy is the use of Dielectric Elastomer Generators (DEGs, or electro-active polymers EAP), in replace of traditional elastomers or springs, exploiting their periodical deformation for directly amplifying the voltage from EMc.

3.1 Configurations

Different mechanical configurations have been explored during the experimental part of this work in order to maximize frequency and amplitude of motion in a wider wind speed range, and better understand the complexities of the system. The system demonstrated its applicability also in a water tunnel, meaning that it could works in any kind of fluid, with opportune configuration and proper parameters. In all the configurations explored the wing was totally free to rotate (with very low friction) around a rigid axis, connected to a frame elastically.
The first configuration (Fig. 10 and 11a) consists in two (or four) elastomers connected to the rotational axis, parallel to the flow and to the floor. It possesses two degree of freedom: a passive rotational motion of the wing, named pitch, and a vertical harmonic motion of the Pivot Point, called plunge. The second configuration (Fig 11b) uses two elastomers orthogonal to the flow and parallel to the floor: this configuration, with three degree of freedom, is mechanically more complex in terms of kinematics and vibrational modes, but helps the comprehension of the aeroelastic peculiarities of the system. It is also the only configuration that doesn’t exploits an EMc for the electrical extraction, because of the more complex trajectory of the PP: DEGs can be supplied by another external source, for instance piezoelectric elements, placed between the frame and the elastic connections. The possibility to exploit a piezo-electric element on the wing found technical complications because its electrical connection, as the wing in our system must be totally free to rotate. The third configuration (Fig. 11c) represents a prototype realized for overcoming the procedure of elastic calibration involved in the other configurations, designed for high speed and turbulent flow applications: the Pivot Point of the wing is constraints to exhibit an arc of circumference path, according to which the magnets are arranged in alternated polarity.

In all the configuration explored piezo-electric elements could be applied on membranes constituting roof and floor of the devices, exploiting the vortexes detachments and their pressure fluctuations involved during the self-sustained oscillations.

### 3.2 Kinematics

The dynamics, obtained through the experimental setup reported in Chapter 2, revealed a rich and complex framework, characterized by sundry motion regimes including stable, periodic and aperiodic (or semi-chaotic) states, depending on the main system parameters. These include geometrical and mechanical properties.
Considering only the aeroelastic aspects, the behaviour of the system is governed by many parameters, e.g.: fluid properties, type and length of the elastomers, wing geometry, and Pivot Point position, global mass and mass distribution. To give a qualitative representation, Fig. 12 shows the resulting motions for a centimetre-size device where the elastomers are orthogonal to the wind (second configuration, Fig. 11b), while varying the incoming flow velocity and the elastic equivalent stiffness $K_y^{eff}$ (that will be introduced in next sections). Looking at the reported trajectories, several observations can be made:

1. The aeroelastic instability exists only if the flow velocity is above a critical value (which we will denote as $U_{cr}$), otherwise the wing aligns with the flow in a stable condition. This onset speed clearly appears to depend on the value of the elastic constant: as an example, at $2.9 \text{ m/s}$, for $K_y^{eff} \leq 1.05 \text{ [N/m]}$ flutter is triggered (where the asymmetric trajectories are due to the relative importance of gravity at low wind speeds), while for larger values we are still below the threshold.

2. If one increases the flow velocity beyond $U_{cr}$, for all the reported cases the wing enters a regular flapping regime characterised by limit cycle oscillations (LCO). The trailing edge trajectory rapidly changes its shape and the amplitude increases with $U$ up to a value which is comparable to the chord size. We highlight how the flow velocity corresponding to this maximum in amplitude varies with $K_y^{eff}$. On the other hand, it can also be noted how the flapping amplitude does not vary significantly on $K_y^{eff}$. 

Figure 12 - Lateral view of trajectories of a centimetre-sized wing in the second configuration (see Fig. 10b) changing the vertical elastic constant $K_y^{eff}$ and increasing the wind speed $U$ (from right in Fig.). From \cite{22}. 

![Lateral view of trajectories of a centimetre-sized wing in the second configuration](image.png)
3. Although not always so visible, the Pivot Point trajectory is also reported in the Figure 11. For certain ranges, the amplitude of this latter is larger than the one of the trailing edge, while for \( U > 3.5\div4.2 \) [m/s] (depending on the stiffness value) the situation gets opposite. When the Pivot Point displacement is maximized, a relative maximum is typically found also for the fluttering frequency: even if this aspect is still under investigation, this condition appears as the most efficient for energy harvesting. Therefore, such ‘transition’ identifies different energy states with which the system manages the incoming flow, qualitatively discriminated by the competition between the maximization of the Pivot Point and the trailing edge absolute amplitudes.

4. When increasing further \( U \), the motion regime may vary significantly. At the highest velocity tested \( (U = 8 \) [m/s]), two particular behaviour is observed: for the smallest stiffness the system goes back to a stable condition, while for larger values the motion becomes aperiodic. It is highlighted that, although the second configuration has a further degree of freedom compared to the first one (the horizontal motion, see Fig. 11b), the trajectories are similar, and carved by the same peculiarities. In fact, after a first initial evolution which differs between the two configurations, the system seems to limiting the degree of freedom in the \( x \) direction (direction parallel to the flow), likewise to what happens in the first configuration when it is mechanically limited, showing a vertical harmonic motion of the Pivot Point. Conversely, no aperiodic regimes are observed in the first and third configurations, for which at high flow speed the system decreases its oscillation amplitude up to stable conditions, characterized by the wing parallel to the flow. Therefore it could be asserted that when the elastic constant in second configuration is enough stiff, it shows same behaviour of configuration one, within the operating conditions of the device. This is also because part of the Drag of the system is transformed in Lift in particular conditions \(^{22}\), such as when the degree of freedom related to the displacement along the fluid is partially or completely limited.

3.3 *Fluttering conditions*

The identify of fluttering states is based on a simple resonance condition between the characteristic (elastic) time of the system and the flow time-scale. This part has been published in \(^{45}\). Similar arguments have been successful in other fields of fluid dynamics and fluid-structure interactions including turbulent flows of dilute polymer solutions and interactions between the wake originated by bluff bodies and elastic structures. The predictions of the geometrical and or physical properties associated to critical conditions (i.e. those separating stable stages from fluttering regimes) have been compared for the first mechanical configuration against the results of experiments, numerical simulations and a phenomenological model based on a set of ordinary differential equations. Here a part of the contents is reported.
In the system reported in Fig. 13, \( x \) and \( y \) are defined as the horizontal and vertical displacement of the Pivot Point (assumed to be small), respectively; \( L \) is the stretched length, \( L_s \) the initial length and \( L_0 \) is the rest-length of the elastomer; hence, \( L_s = L_0 + \epsilon = L_0 \lambda_s \) where \( \epsilon \) is the initially given pre-stretching. Additionally, let \( G \) be the shear modulus of the elastomer material and \( A_0 \) the initial cross-sectional area of the elastomer. The elastic force modulus, considering the first term of elasticity, is assumed to be:

\[
F_{el} = GA_0 \left[ \frac{L}{L_0} - \frac{\left( \frac{L_0}{L} \right)^2}{L_s} \right] \tag{4}
\]

The total elastic force in the equilibrium position along the \( y \)-direction can be expressed as

\[
F_y = 4GA_0 \left[ \frac{\sqrt{L_s^2 + y^2}}{L_0} - \left( \frac{L_0}{L_s} \right)^2 \frac{y}{L_s} \right] \tag{5}
\]

that in the limit of small oscillations (i.e. \( y \ll L_0 < L_s \)) yields

\[
F_y = 4GA_0 \left[ \frac{L_s}{L_0} - \left( \frac{L_0}{L_s} \right)^2 \frac{y}{L_s} \right] \tag{6}
\]

which can be intended as a linear function of \( y \) so that it can also be written as

\[
F_y = K_y^{\text{eff}} y \tag{7}
\]

where we have introduced the equivalent or effective stiffness, with respect to the vertical direction:

\[
K_y^{\text{eff}} = \frac{4GA_0}{L_s} \left[ \frac{L_s}{L_0} - \left( \frac{L_0}{L_s} \right)^2 \right] \tag{8}
\]

Additionally, for the sake of simplicity we can consider the pre-stretching to be small compared to the rest-length \( L_0 \), so that \( \epsilon \ll L_0 \). In this case, \( K_y^{\text{eff}} \) reduces to:

\[
K_y^{\text{eff}} = 12GA_0 \frac{\epsilon}{L_0^2} \tag{9}
\]

The natural frequency associated with the vertical oscillations can thus be expressed as:
Figure 14 - Elastic force along the length of the elastomer (a) and along vertical shear deformation direction (b), varying the pre-stretching value $\lambda_s$, imposing the same absolute deformation.

$$ v_n = \frac{1}{2\pi} \sqrt{\frac{K_{y}^{\text{eff}}}{m}} $$

where $m$ is the mass of the device. Similar considerations can be made for the horizontal displacement. The total elastic force can be expressed now as

$$ F_x = 2GA_0 \left[ \frac{L_s - x}{L_0} - \left( \frac{L_0}{L_s - x} \right)^2 \right] - 2GA_0 \left[ \frac{L_s + x}{L_0} - \left( \frac{L_0}{L_s + x} \right)^2 \right] $$

(10)

that for $x \ll L_0$ yields:

$$ K_x^{\text{eff}} = \frac{4GA_0}{(10L_s^3)(L_s^3 + 2L_0^3)} \varepsilon \ll L_0 \quad \Rightarrow \quad K_y^{\text{eff}} \ll 1 $$

(11)

Under these assumptions, it is easily seen that:

$$ \frac{K_y^{\text{eff}}}{K_x^{\text{eff}}} = \frac{\varepsilon}{L_0} \ll 1 $$

(12)

which means that the natural frequency associated with the horizontal oscillations is much higher than the one related to the vertical oscillations. For this reason, it is assumed the latter to be the first frequency to couple with the hydrodynamic one.

Figure 14 shows the effect of pre-stretching on the effective vertical elastic constant $K_y^{\text{eff}}$, and how its non-linearity increases at low pre-stretching. Our system typically uses values between 1.1 and 1.6.

After the characterization of the natural frequencies, the next step is to identify the proper one concerning the fluid dynamical side. For this purpose, it is considered the situation in which the wing is hinged at the Pivot Point with only the pitching motion to be allowed. Note that this corresponds to the limit of infinitely large spring stiffness that was already defined as the wind vane situation in Ref. 47. In this case, it is sufficient to refer to the moment equation only, which yields:

$$ I_{pp} \ddot{\theta} = M_{pp}^{\text{drag}} $$

(13)
and assuming the rotation angle to be small (with the wing almost aligned with the unperturbed flow) and the Lift force to be applied at a distance $C/4$ from the Pivot Point, the aerodynamic moment can be expressed as $M_{pp}^{lifo} = \frac{\pi}{4} \rho_f CA \theta U^2$, where $A$ is a reference area (usually $A = C \times S$) and $\rho_f$ is the fluid density. If we search for harmonic solutions, the following frequency can be derived:

$$v_p = \frac{\rho_f CA}{16\pi I_{pp}} U$$

which we denote as the wind vane frequency.

Let us now assert that the critical condition for the emergence of fluttering is related to a resonance condition between the natural frequency and the aerodynamic frequency. Arguments of this type have been successful, e.g., to explain symmetry breaking mechanism in fluid-structure interaction as well as the emergence of elastic instabilities and macroscopic spatial scales at which microscopic polymers cause viscoelastic behaviour. Following this line of reasoning, when these two frequencies are comparable, i.e. $v_p \approx v_n$, one can easily derive an expression for the critical velocity for the onset of sustained fluttering.

If we assume that the natural frequency to be considered is the one associated with vertical oscillations ($y$ direction). By equating Eq. (15) with Eq. (10) we obtain:

$$U_{cr} \approx \frac{2}{\sqrt{\pi}} \frac{l_{pp} K_y^{eff}}{\rho_f C^2 S m}$$

It should be said that the moment of inertia $I_{pp}$ is a function of the wing mass distribution and geometry. Nevertheless, this form for the expression is useful for our scope, as it will be show in the experimental part.

### 3.4 Experimental results

The use of an high definition camera coupled to an appropriate illumination and video processing system, allowed to obtain a precise acquisition of the wing motion: a light scatterer, placed on the rotational axis of the wing, was illuminated by a line beam laser, while a numerical elaboration of the movie, conducted by Matlab®, returned the main quantities in time such as the Pivot Point position and the pitching angle.

The results reported in this section are related to the first aeroelastic configuration (Fig 11a) exploiting two elastomers parallel to the flow, and a rod to which an airfoil is fixed or it is free to rotate. No electrical extraction is adopted for these measurements. The centimetre-sized systems are characterized by a wing typically very light, in the order of few grams, while the design of the mechanical connections follows the experimental and modelling requirements.
3.4.1 **Onset speed**

A hysteresis phenomenon has been observed in what it concerns the evolution of motion whether increasing or decreasing the wind speed. This is found to be crucial also for the fluttering threshold determination. As also observed in similar systems\(^{16,51}\) this opens the problem to define a convention to evaluate the critical flow velocity \(U_{cr}\): we will assume this to be the lower value of the range, i.e. the value that, if further decreased, leads the system to stop the existing self-sustained excitation. We mention however that we typically find this lower value to be between 0.8 and 0.9 times the upper one (which is measured while increasing the wind speed), similarly to \(^{51}\). In order to verify the resonance condition for the emergence of fluttering previously proposed, two studies have been performed. For all measurements, the type of elastomers is fixed such that \(G A_0 = 0.604 \text{ [N]}\) while the wing geometry is set to be: \(C = 60 \text{ [mm]}, S = 70 \text{ [mm]}\). In the first study, we investigate the dependence of the critical velocity \(U_{cr}\) on the elastomer rest-length \(L_0\) (while a constant pre-stretching is retained) i.e. varying the effective stiffness. Results are shown in Fig. 15 together with the prediction expressed by Eq. (16) where we make use of the simplified expression for \(K_y^{eff}\) Eq. (9), so that \(U_{cr} \propto L_0^{-1}\). The measurements were found to be in good agreement with the expected scaling law. For the lowest tested value of \(L_0 = 25 \text{ [mm]}\) the critical velocity was around 8.5 \([\text{m/s}]\) while for the highest one \((L_0 = 120 \text{ [mm]})\) it dropped to approximately 3 \([\text{m/s}]\). In Fig.15a reduced error bars related to \(U_{cr}\) increasing \(L_0\) can be observed: this is linked to the higher hysteresis encountered in the estimation of \(U_{cr}\) by increasing and decreasing \(U\) in the cases when the rest length of the elastomers is small (low \(K_y^{eff}\)).

The plot shows that, for the given configuration, \(U_{cr}\) may be reduced of around 33% by placing a 4.5 \([\text{g}]\) additional mass. Similarly to what we have found in the ideal framework, the wind tunnel study appeared to verify the simple resonance condition idea considering the more

![Figure 15](image)

*Figure 15 - Experimental measurements of the critical velocity for the onset of sustained flapping (a) as a function of the elastomer rest-length \(L_0\) (here, \(m = 1.63[\text{g}]\)) and (b) as a function of the wing mass \(m\) (with \(L_0 = 58[\text{mm}], L = 66[\text{mm}]\)).*
realistic model. We underline the importance of this result in view of the practical application, supplying useful insights. This clearly showed how the cut-in speed of the system can be governed by such a simple parameter. Subsequently, we focused on the dependence of $U_{cr}$ upon the mass of the system $m$. In this experiment, the mass was varied by placing additional weights on the wing rotational axis so that the moment of inertia $I_{pp}$ remained constant. Hence, a scaling according to $U_{cr} \propto \left(\sqrt{m}\right)^{-1}$ was observed. These important considerations helped the design of the energy harvesting systems.

### 3.4.2 LCO Analysis

In order to guarantee a good electrical power extraction, the amplitude of the Pivot Point motion and the frequency of oscillation must be maximized. Currently, there is a lack of analytical models able to describe accurately the system in the nonlinear regime with large excursions we want to exploit. Furthermore, the correlations between the numerous governing parameters involve a complex framework: e.g., adding mass on the axis increases the amplitude but, at the same time, decreases the oscillation frequency, while the increase of the elastic tension to recover a higher frequency leads to a higher onset speed $U_{cr}$, and therefore it modifies the operational flow speed range. Consequently, similarly to other fluid-solid interaction systems, predictive scaling laws are still unknown.

In this sub-section, few measurement campaigns realized to investigate the sensitivity of the parameters on the evolution of the LCOs are reported, for giving an idea of the complexity of such aeroelastic system, and at the same time for showing some unusual effects, not reported in literature.

#### 3.4.2.1 Effect of moving the centre of mass

One way to easily change the aeroelastic condition of the system is to change the centre of mass of the wings: this give the advantage of keeping fixed the elastic parameters and masses, which heavily modifies the response of the system in terms of on-speed and oscillation frequencies, as it will be clear during the analysis in this sub-section. The main mechanical parameters are collected in Tab.2: the first row is referred to the elastic properties of the polyisoprene cylindrical elastomers used, consisting in two pre-stretched portion of initial length $L_0$, initial cross-section $A_0$ and pre-stretching length $L_s$. These lead to an effective vertical elastic constant $K_y^{eff}$, which brings a natural frequency $\nu_n$ when the global mass $m=m_{wing}+m_{plunge}$ is hanged to the middle parts of the elastomers, as portrayed in Fig. 10a.
Table 2 - Details of the measurements of the analysis of LCOs varying CM.

<table>
<thead>
<tr>
<th>CM</th>
<th>( x_{CM/C} )</th>
<th>Span [m]</th>
<th>Chord [m]</th>
<th>PP</th>
<th>( m_{CM0} ) [g]</th>
<th>( m_{CM1-4} ) [g]</th>
<th>( K_c^{off} ) [N/m]</th>
</tr>
</thead>
<tbody>
<tr>
<td>CM0</td>
<td>0.19</td>
<td>0.07</td>
<td>0.02</td>
<td>0.05</td>
<td>0.63</td>
<td>0.85</td>
<td>5.4</td>
</tr>
<tr>
<td>CM1</td>
<td>0.21</td>
<td></td>
<td>0.05</td>
<td>0.05</td>
<td>0.63</td>
<td>0.85</td>
<td></td>
</tr>
<tr>
<td>CM2</td>
<td>0.24</td>
<td></td>
<td>0.05</td>
<td>0.05</td>
<td>0.63</td>
<td>0.85</td>
<td></td>
</tr>
<tr>
<td>CM3</td>
<td>0.3</td>
<td></td>
<td>0.05</td>
<td>0.05</td>
<td>0.63</td>
<td>0.85</td>
<td></td>
</tr>
<tr>
<td>CM4</td>
<td>0.35</td>
<td></td>
<td>0.05</td>
<td>0.05</td>
<td>0.63</td>
<td>0.85</td>
<td></td>
</tr>
</tbody>
</table>

The 6% of the mass of the wing \( m_{wing} \) was moved along the chord length (of 20 [mm]), positioning two strings of tape of 5x70x0.2 [mm³] on both sides of the acetate airfoil at 4, 8, 12 and 16 [mm] from the leading edge \( LE \), which for this case coincided with the Pivot Point \( PP \).

The analysis was conducted also with a wing which does not possess any strings, thus resulting lighter (CM0), in order to show the effect of a slight change in mass which was positioned almost at the point of attack of the elastomers, thus without altering the moment of the wing. The evolution of the LCOs was evaluated increasing the wind speed \( U \) by the experimental acquisition of the \( PP \) and pitch angle \( \theta \) displacements, by which the position of the trailing edge of the wing \( (TE) \) was calculated. The lateral trajectories of the LCO were thus obtained, and reported in the Figure below. The LCOs appeared almost the same of those observed in the section 3.2 in the second configuration (see Fig. 11b and Fig. 12), confirming the attitude of the system, for these mechanical configurations, to limit its degree of freedom in rotational and vertical displacements.

The effect of the change of \( CM \) is clear when the pitch angle is examined: higher values of \( CM \) determine more asymmetric LCOs, as the gravity for this system plays a relevant role. In addition, by increasing \( CM \), higher \( U_{cr} \) were observed, and the transitions (change of dynamics on LCOs) endured a delay in \( U \), if all \( CM \) were compared.

Figure 16 - Lateral trajectories of LCOs (PP and TE) varying CM and U. Wind flows from right to left.
The latter appeared to be related to different hydrodynamic characteristic lengths, involving vortex creation and vortex shedding conditions, as the elastic constant was fixed. This phenomenon is expressed in Eq. (16) in sec. 3.3 by the term $I_{pp}$, which depends on $CM$. The evolution of frequency and amplitude of oscillation of the $PP$ are reported in the above Figures. As it can be noted, the maximum values corresponded to the same LCOs. This particular LCO in this work is named pre-transition, and it corresponds to the best condition for the electrical conversion, as it was stated in the introduction of this sub-section. Only after this condition both the parameters showed a decrease followed to a slightly slower increase along the wind speed. Looking at the Figure 18a, it can be also observed that when $PP$ amplitude decreases $TE$ amplitude increases: this behaviour clearly distinct the pre-transition and transition phenomena in interest.

Another relevant parameter measured during the LCOs is the phase $\phi$ between the two main DOF (Fig. 18b); it seems to exceed 90 [°] only after the transition. This sounds surprising, as the maximum work of a two-modes body oscillation theoretically is expected to be observed when their delay phase is around 90 [°], implying an always positive work during the cycles.
In our system $\Phi$ remains lower than 90 [°] in the LCOs before transition, typically between 10 and 40 [°] for light system and up to 80 [°] for heavier ones, and it jumps to values around or over 90 [°] in the transition.

Lissajous curves, introduced in 1857 and deeply used to represent systems of parametric equations describing complex harmonic motions, are thus used to display the LCOs of the system, and reported in Figure 19. When $\phi$ is about 90 [°] the curves tends to became a circle, while it can be observed a different inclination of the curves before and after the transition: this could be addressed to a difference in the delay detachment of the LEV.

![Lissajous Curves of CM1 varying U.](image)

Figure 19 - *Lissajous Curves of CM1 varying U.*

![Maximal and minimal $\theta$ during the LCOs (a) and $\theta$ amplitude (b) varying CM and U.](image)

Figure 20 - *Maximal and minimal $\theta$ during the LCOs (a) and $\theta$ amplitude (b) varying CM and U.*
It is highlighted that symmetric LCOs before the transition are observed in many cases (included in CM0 and CM1), and this indicates that the gravity is not the responsible of this unusual effect. A rotation of the system in order to have the axis of the wing parallel to the gravity also confirmed it.

In fact the circumference could be wrongly attributed to the condition of which negative pitch angle reaches the values of the positive ones. In Figure 19 the Lissajous curves at 3.5 and 4 [m/s] appear symmetric both in pitch and plunge, but with a relative phase of 20-30 [°]. The trends of maximum, minimum and averaged $\phi$ are plotted in Figure 20. The results are coherent with the previous reported parameter trends: the pitch angle $\theta$ increases with $U$ until the transition occurs, similarly to $PP$ amplitude and frequency. It can be also noted that a clear difference in behaviour persists for $CM$ higher and lower 0.25 of chord ($C/4$). Despite $C/4$ represents the aerodynamic centre in symmetric airfoil in steady flow; this is not true for our cases, characterized by highly separated unsteady flow. No interpretation has been found for this behaviour.

### 3.4.2.2 Effect of the airfoil dimensions

Another way to easily change the aerodynamic condition of the system is varying the chord length of the wing. Because of the light weight of the airfoils, composed by a foil of acetate glued to a 3d-printed tubular part, the varying of chord consist in a minimal change of global mass.

Anyway, differences are observed firstly in natural frequencies, then in oscillation frequencies increasing the wind speed, and also in the evolution of LCOs. The latter, reported in Figure 21, exhibit a delay also in what it concerns the transitions: for instance for C15 it is observed between 3.5 and 4.0 [m/s] and in C20 between 4.0 and 4.5 [m/s]. The vertical harmonic motion of the $PP$ is brake out for C30, where the Drag force acting on the wing increases at the minimum and maximum pitch angle, implying a curvature of the $PP$ trajectory (Fig. 21, C30).

<table>
<thead>
<tr>
<th>$K_{eff}$</th>
<th>Span</th>
<th>Chord</th>
<th>$PA$</th>
</tr>
</thead>
<tbody>
<tr>
<td>5.34 $10^5$</td>
<td>6.00 $10^{-4}$</td>
<td>1.13 $10^{-6}$</td>
<td>4.36 $10^{-2}$</td>
</tr>
</tbody>
</table>

Table 3 - Details of the measurements of the analysis of LCOs varying C. C20d corresponds to C20 decreasing $U$ (in all other experiments $U$ is always increased), presented for showing the little, but not negligible, hysteresis along the parameters in interest.

$G$  | $r$  | $A_0$  | $L_0$  | $L_*$  | $\lambda_*$ |
<table>
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<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>[Pa]</td>
<td>[m]</td>
<td>[m²]</td>
<td>[m]</td>
<td>[m]</td>
<td>[°]</td>
</tr>
<tr>
<td>5.34 $10^5$</td>
<td>6.00 $10^{-4}$</td>
<td>1.13 $10^{-6}$</td>
<td>4.36 $10^{-2}$</td>
<td>4.93 $10^{-2}$</td>
<td>1.1</td>
</tr>
</tbody>
</table>

$K_{eff}^*$  | Span | Chord | $PA$ |
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>[N/m]</td>
<td>[m]</td>
<td>[m]</td>
<td>[°]</td>
</tr>
<tr>
<td>13.9</td>
<td>0.07</td>
<td>0.02</td>
<td>0.05</td>
</tr>
</tbody>
</table>

$u_{CM/C}$  | $m_{win}\theta$  | $m_{tot}$  | $\nu_h$ |
| [°]    | [g]  | [g]    | [Hz]   |
| 0.19   | 0.14 | 1.10   | 17.9   |

$U$ (in all other experiments $U$ is always increased), presented for showing the little, but not negligible, hysteresis along the parameters in interest.
Concomitances of LCO are also observed. These include LCOs during the hysteresis in $U$ led for $C=20$ [mm] (C20d), but not only (see C15, $U=4.0$ [m/s] and C20 at $U=3.5$ and 4.0 [m/s]): this phenomenon will be analyzed in next sections. The oscillation frequency increases with $U$ similarly to the previous case (sub-section 3.4.2.1), but showing a slightly lower slope. This is attributed to the higher pre-stretching: $K_{yy}^{eff}$ is now 13.9 [N/m] with a pre-stretching of 1.1, while in the previous case $K_{yy}^{eff}$ was 5.4 [N/m] and $\lambda$ was 1.04. Interestingly, for the first range of wind speed (from 3 to 4 [m/s]) the oscillation frequencies appear very similar for all the chord lengths used, if compared to their different natural frequencies (see Fig. 22b).

Figure 21 - Lateral trajectories of the wing (PP and TE) increasing $C$ and $U$. An example of the hysteresis in LCO for chord=20 [mm] (C20d) is reported, obtained by decreasing $U$. Wind from right to left.

Figure 22 - Chord-normalized PP amplitude (a) and Frequency oscillation (b) varying $C$ and $U$. 
The TE amplitude and the phase between pitch and plunge \( \phi \), reported in Fig. 23, show similar behaviour by varying the chord length and increasing the wind speed. The curves related to C30 appears smoothed and monotonic, meanwhile the curvature of PP trajectory seems to delay the condition of \( \phi=90 \) [°], despite the system showed a post-transition motion. Also the maximal and minimal \( \theta \), reported in Fig. 24, appear very similar for C15 and C20, while C30 shows a smoother trend. The reduced frequency of oscillations, defined also as the ratio of between the maximum velocity of \( PP \), \( PP_{y_{max}} = 2\pi v_0PP_y/2 \) and the wind speed \( U \), is also reported in Figure 24b. It can be noted that, fixing \( k_y^{eff} \), lower is the chord length (and thus the mass), lower is the reduced frequency, however less than one for all cases.

The concomitance of two LCO can be also evaluated from the Lissajous curves, reported in Figure 25. At 3.5 and 4.0 [m/s] two alternated curves can be clearly distinguished. The two LCOs in each case seem to correspond to lower and higher energy conditions with respect the previous and next single LCOs.
Figure 25 - Lissajous Curves of C=20 [mm] varying U. Period doubling at 3.5 and 4.0 [m/s].

3.4.2.3 Effect of the mass on Plunge

The effect on the dynamics of the wing adding of mass exclusively on the PP position (contributing only to the plunge term) provides important information, because, as already introduced in Chapter 2, during the electrical extraction through EMc, the coils are placed in such position. For this reason, the mass was varied by gradually adding identical cylindrical discs of Nd$_2$Fe$_{14}$B permanent magnets of diameter 7 [mm] and height 1 [mm] on the extremes of the pivot axis. To fix the magnets, two thin laminated iron discs were glued to 3D-printed supports, linked to the pivot axis. The magnets were then added to each side, from 1 to 4. The increase of mass was followed by the increase of the magnetic energy density, and thus of the EM coupling, while the natural frequency decreased.

The analysis of the dynamics reported in this sub-section does not include any electrical extraction. This case is reported in next chapter (sub-section 4.3.2), where two coils were fixed in front of the trajectory of magnets (opposite respect the traditional configuration studied, see Fig. 10), in order to provide an example of the effect of the electrical extraction on the dynamics of the system. In fact, as the energy absorption through EMc acts from the plunge term, the LCOs and their characteristics are definitively modified. The absence of electrical extraction coincides with the case in which no electrical current is request from the coils that means in open circuit configuration.
The configuration adopting 2 magnets per side revealed mechanical instabilities during fluttering: an additional mode has been emerged, characterized by an out of phase of $\pi$ between the deformation of the elastomers, while pulsations and combinations of other modes were shown increasing the wind speed. In addition to the difficulty in the acquisition motion, for the EMc configuration the coils are connected in series, the vertical motion of the pivot extremes must be in phase, in order to avoid the destructivity effect of the electrical signals from the coils. Therefore, the case M2 is not reported.

From Figure 27 it is evident how adding of the mass of plunge, reducing the natural frequencies of the system, brings to higher amplitude LCO. Not only: all the transient phenomena and the asymmetry braking of the pitch angle. Again, some double LCOs are observed before the PP amplitude transition.

As expected, oscillation amplitude increases with the increasing of the mass on plunge, as the mass of the wing is fixed (see Fig. 27); in fact, this involves that a constant moment of inertia is experienced by the wing, and the only difference between the systems consists in the plunge contribute of the coupled-mode, which in turns only decrease the natural and oscillation frequencies, as stated before.

![Lateral trajectories of the wing increasing C and U varying the m_plunge and U.](image)
As already observed in the two previous studies, if the pre-stretching is relevant the increase of oscillation frequencies appear reduced. In this case $K_{Y}^{eff}$ is 47.5 [N/m] and $\lambda_s$ is 1.5.

These high values are chosen in order to maintain the natural frequencies relatively high (more than 10 [Hz], as it will be explain in Chapter 4) also with the heavier masses, as Fig. 27b shows. Oscillation frequencies remain very close to the natural ones. A moderate increasing is measured after the transition (M3 and M4), while during the LCO that corresponds to the transition (M3 and M4 at 4 [m/s]) a drop, instead of the classical maximum of frequency, is detected. This could be attributed to two contributory cases: the very heavy mass (4-5 [g] for M3 and M4) and the already cited high pre-stretching ($K_{Y}^{eff}$ = 47.5 [N/m] with $\lambda_s$=1.5). The presence of a double period LCO is excluded as a possible reason.

A sharp increase in oscillation frequency could be probably observed at higher $U$ for the cases M0 and M1, for which the transition is still absent in the considered wind speed range.
The pitch angles (and therefore $TE$ amplitude) and phase between pitch and plunge are also coherent with the LCOs evolution and with previous measurements: $\phi$ exceeds $90^\circ$ only after the transition (see Fig. 28b), averaged $\theta$ tends to zero if transition occurs (see Fig. 26), and because of larger $PP$ excursion, also minimum and maximum $\theta$ increase with the amount of translational mass (see Fig. 29).

### 3.5 Dynamics

The system has 3 DOFs: $x$, $y$ and $\theta$. However, in order to evaluate the aerodynamic forces acting on the wing, in a first approximation the time behaviour of $x$ is neglected, since in the considered configuration it is always much smaller than $y$. Then we assume $x = 0$ for any $t$.

In this way, our system can be described by the usual equations of a pitch and plunge system. Adopting the reference system depicted in Fig. 2, the equation of motion of the system can be derived from the Lagrange’s equations respect to the variables $y$, $\theta$ defined as the position $r = (0, y)$ of the Pivot Point $PP$ and $\theta$ the angle of the wing (positive nose up). In the present experiment $PP$ is placed at the Leading Edge $LE$.

Following Veilleux and Dumas the equations read:

$$m\ddot{y} + S_i(\dot{\theta}^2 \sin(\theta) - \dot{\theta} \cos(\theta)) + K^\text{eff}_y y + D_y \dot{y} + m \, g = L$$

$$I_{PP} \ddot{\theta} - S_i(y \cos(\theta) + g \cos(\theta)) + K_\theta \theta + D_\theta \dot{\theta} = M$$

where $m = m_a + m_w$ is the total mass of the system, $S_i = m_w x_\theta$ is the static imbalance, $x_\theta$ represents the distance between the Pivot Point $PP$ and the centre of mass $CM$, $K^\text{eff}_y$ is the effective elastic constant along $y$, $K_\theta$ is the torsional elastic constant, $D_y$ and $D_\theta$ are the structural damping related to $y$ and $\theta$ respectively, $g$ is the gravity acceleration, $I_{PP}$ is the inertia moment, $L$ is the Lift force and $M$ the aerodynamics moment at $PP$. 

---

**Figure 29** - Maximal and minimal (a) and averaged (b) pitch angle varying $m_{\text{plunge}}$ and $U$.

---
In our case $K_\theta$ is equal to zero, as any torsional spring is adopted in the system, and this, united to the peculiar elastic configuration, corresponds to the main difference between the classical pitch and plunge systems investigated in literature.\(^4\)

The mechanical damping in the vertical direction $D_y$ can be easily evaluated by measuring the Pivot Point displacement versus time after an external perturbation of the wing from its equilibrium position, which corresponds to the position due to the gravity, as the elastic elements are placed orthogonal to the latter. This experimental condition coincides with that used to measure the natural frequency of the system (Eq. 7) when the wing lies in the gravity direction, coherently to the results obtained by a Fourier analysis on the displacement.

On the other hand, the estimation of $D_\theta$ results very complex, since the wing rotates with a variable high angular speed during the different LCOs (20-100 [rad/s]), and moreover, also the flow direction changes significantly along the dynamics.

One of the methods investigated for evaluating $D_\theta$ is measuring the kinematics of the wing suddenly eliminating the wind to the structure when it is experiencing a LCO. A typical device is thus fixed on a table and subjected to the flow provided by a fan equipped by a conic-shaped conveyor ($U$ about 4 [m/s]): after waiting that the system reached a stable LCO, a wood panel was slantwise introduced between the fan and the system to deviate the flow, and therefore providing a sudden stop to the forcing source of the self-sustained oscillation.

This permits to evaluate the first periods of oscillations at large angles, that means meanwhile fluttering, and the effect of the turns of the wing on the damping of the PP. From the Figure 30b it can be observed how the wing, after the stoppage of the flow, firstly decreases both angle and amplitude with the same trend (see the negative part for more clarity), and then exhibits a rotation that exceed 360 [°], followed by a complex stabilization, led by the PP (Fig. 30a) with a quite low perturbed exponential decay. Obviously, this behaviour of the pitch angle is quite impossible to be prescribed with ordinary equations.\(^5\) The PP damping obtained by this measurement is compared with the structural damping in absence of wind in Figure 30a. It can be clearly observed that the two cases appear very similar, and quantitatively in the same order of magnitude. The difference between the natural frequency (case without wind) and the oscillation frequency (during fluttering) can be also observed.

The part of the angular damping in interest can be observed in the Figure 30b from about 1.5 [s], time at which the wind is deviated, to about 1.8 [s], time from which a divergence of the angle occurs. The damping coefficient $\beta$ was evaluated in the order of magnitude of $10^{-6}$ [N s/m]. At this state of the research, this is the most accurate method providing a vague idea of the entity of such parameter.
The value has to be compared with whom Poirel et al. provided in their study on the motion of an airfoil constrained to rotate in pure pitch: their structure was composed by an airfoil of $C = 0.156$ [m], $S = 0.61$ [m], $m = 0.771$ [kg], $I = 1.3510^{-3}$ [kg m$^2$], $D = 0.002$ [N m$^{-1}$s]. Instead our system is equipped with an airfoil of $C = 1510^{-3}$ [m], $S = 7010^{-3}$ [m], with $m = 0.1510^{-3}$ [kg], $I = 1.110^{-8}$ [kg m$^2$], $D = 5.510^{-8}$ [N m$^{-1}$s]. The angular damping ratio $D/I$ in $^{54}$ is about $1.5$ [s$^{-1}$] while in our case is $5$ [s$^{-1}$]: this means that we are more over-damped, probably also because the absence of the torsional spring in our system reduces the storing of the energy from the moment $M$. The ratio $(D/M)^{0.5}$ is the pulsation of the damped system, that in $^{54}$ is $\omega_d = 15$ [rad/s], while in our case $\omega_d = 31$ [rad/s]. These differences in mechanical properties should explain the differences in their observations and their conclusions on the phenomena with respect our evidences.

Another difficulty found in the application of the equations of the system was that the difference of the force contributes reported in Eq. (17) and (18) were affected by the low accuracy of some parameters such as the static imbalance $S_i$. Moreover, the estimation of the aerodynamic coefficient, for example, seemed too sensible to the derivation of the parameters in interest, such as $\dot{y}$, $\dot{y}$, $\dot{\theta}$ and $\ddot{\theta}$ (see Eq. (17),(18) and Eq. (1),(2)). Therefore, only a raw estimation of the Lift, Drag and Moment coefficient versus pitch angle for different configurations has been obtained, founding out that not only the magnitude, but also the shape of the curves appear modified with some arbitrary choices, due to the sensibility of the parameters.

For these reasons, despite an extensive casuistry, this part was not reported in the thesis.
3.6 Phenomenological model

Seeking for an analytical description of the system and to confirm our findings from numerical simulations in a complementary way, we adopt and modify the phenomenological, quasi-steady model originally developed for falling plates by Andersen et al. and later used for insect flight. We note that a similar attempt has been recently made for an active fluttering foil energy harvester. The model consists of a system of first order Ordinary Differential Equations (ODEs) written in the frame of reference $(x_0, y_0)$ co-rotating with the plate. We modify the equations to be suitable for our idealized model by taking into account the elastic force due to the elastomers and the incoming flow, so that they become:

\[
(m + m_{11})\ddot{v}_x = (m + m_{22})\dot{\theta}v_{y'} + F_{x'}^{el} - \rho_f \Gamma \tilde{v}_y - F_{x'}^{\nu}
\]

\[
(m + m_{22})\dot{v}_y = (m + m_{11})\dot{\theta}v_x + F_{y'}^{el} - \rho_f \Gamma \tilde{v}_x - F_{y'}^{\nu}
\]

\[
(l_G + l_a)\dot{\omega} = (m_{11} - m_{22})v'_x v'_y - c r F_{y'}^{el} - l_c \rho_f \Gamma \tilde{v}_x - \tau^v
\]

\[
\dot{x}_G' = v_x' + \dot{\theta} y_G'
\]

\[
\dot{y}_G' = v_y' + \dot{\theta} x_G'
\]

\[
\dot{\theta} = \omega
\]

where $v_x'$ and $v_y'$ denote the center of mass velocities, $\tilde{v}_x' = (v_x' - U \cos \theta)$ and $\tilde{v}_y' = (v_y' - U \sin \theta)$ are those relative to the unperturbed flow, $m$ is the plate mass, $I_G$ is the moment of inertia with respect to the center of mass, $r$ represents the distance between the center of mass and the Pivot Point normalized with the chord, $x'_G$ and $y'_G$ are the coordinates of the plate center of mass, $\theta$ is the pitching angle, $F^{el}$ is the elastic force and $l_c = C/4 \cos(\theta)$ is the moment arm of the circulatory force. The added mass coefficients $m_{11}$, $m_{22}$ and $l_a$ are expressed for the plate of rectangular cross-section following Huang et al.:

\[
m_{11} = (3\pi/8) \rho c \delta^2, \quad m_{22} = (3\pi/8) \rho f C^2, \quad l_a = (5\pi/256) \rho f (C^2 - \delta^2)^2.
\]

The circulation $\Gamma$, the viscous force $F^v$ and the dissipative fluid torque $\tau^v$ are expressed in a semi-empirical manner using several free parameters:

\[
\Gamma = -C_{rC} \frac{\tilde{v}_x' \tilde{v}_y'}{\sqrt{\tilde{v}_x'^2 + \tilde{v}_y'^2}} + \frac{1}{2} C_{bC} C^2 \dot{\theta}
\]
where $C_t$ and $C_R$ are the translational and rotational Lift coefficients, respectively, $C_D(0)$ and $C_D(\pi/2)$ are the Drag coefficients while $C_T$ is the dissipative torque coefficient. On the numerical side, the evolution of such equations is performed using a third order Adams-Bashforth integration:

$$X^{n+1} = X^n + c_{f_1}H^n + c_{f_2}H^{n-1} + c_{f_3}H^{n-2}$$

where $X$ is a vector containing the six unknown quantities and $H$ collects the right-hand-side of the equations; the superscript denotes that the quantity is evaluated at a certain discrete time $t_n = n\Delta t$ while expressions of the coefficients are: $c_{f_1} = 23/12 \Delta t$, $c_{f_2} = -4/3 \Delta t$ and $c_{f_3} = 5/12 \Delta t$.

The model seems to fit quite well with the CFD simulations provided by our collaborators: for more details on the numerical simulations the reader is referred to Ref. $^{58}$. In Fig. 31 the oscillation frequencies related to the change of $K_{y}^{eff}$ and $U$ are reported.

A prototype having wing chord $C = 20$ [mm], span $S = 70$ [mm], pivot axis $PP$ located at 0.5 [mm] from the leading edge $LE$, total mass of the moving system $m = 0.85$ [g], elastomers with diameter $2 \times r = 1.2$ [mm], rest length $L_0 = 47.3$ [mm] and stretched to $L = 66.3$ [mm], is used for verifying the model. The experimental kinematics was captured and compared with the prediction of the model, as shown in Figure 32 for the shape of trajectories and Figure 33 for the fluttering frequency, amplitude and phase between pitch and plunge.

Figure 31 - Fluttering frequency as a function of $K_{y}^{eff1/2}$ for the unstable cases closest to the marginal line (a). The dotted line represents the natural frequency $\nu_n$ (Eq. 10). Fluttering frequency as a function of the free-stream velocity (b). Circles: numerical simulations, squares: phenomenological model.
It is remarked that the set of free parameters is fixed in this parametric exploration. From the figures one can see that good agreement is found overall. The optimal condition in which both frequency and amplitude are maximized (corresponding to $U = 4$ [m/s]) reveals to be the most critical one, the model smoothing the maximum in frequency and $PP$ amplitude, compared to the experimental finding. Moreover, it has to be noted how the resulting phase between pitch and plunge suddenly changes when increasing the wind speed from this condition, as previously observed in the LCO analysis section.

**Figure 32** - Trajectories from experiments (a) and phenomenological model (b), increasing $U$.

**Figure 33** - Fluttering (a) frequency, (b) amplitude (normalized with the wing chord) and (c) phase between pitch and plunge, from experiments (squares) and phenomenological model (triangles).

### 3.7 Aeroelastic stiffening

Despite the proposed phenomenological model provides good agreement with the experimental results, it still depends on a numerous set of arbitrary parameters. These include the terms of added mass, theoretically exiguous, and the semi-empirical terms of dissipation. The choice of such parameters explains why the oscillation frequencies appear quite similar, after a proper calibration. On the other hand, the CFD simulations are still unable to predict some bifurcations of the system, while if the trajectories result quite well fitted, oscillation frequencies and pitch and plunge phase cannot match for all wind speed, and vice versa.
The author observed that oscillation frequencies and the PP amplitudes shown a very similar behaviour increasing the wind speed $U$. If the effective elastic constant is calculated considering the value of maximal stretching of the elastomers, instead of the effective pre-stretched one (considered for the calculation of the natural frequency, see Eq. 8), the results appear interesting. Considering the experimental case where $CM$ is varied along the chord $C$ (sec. 3.4.2.1), the maximal stretching $\lambda_{\text{max}}$ calculated as the ratio of the maximal experimental deformation $L'_{\text{max}}$ and the rest-length of the elastomers $L_0$ reads:

$$\lambda'_{\text{max}} = \frac{L'_{\text{max}}}{L_0} = \frac{\left(\frac{F_{\text{max}}}{2}\right)^2 + L_0^2}{L_0}$$

(29)

where the pre-stretching length is defined as $L_s = L_0 \lambda_s$. Therefore, following Eq. (8), the effective elastic constant $K_{y}^{\text{eff}}$ can be defined as:

$$K_{y}^{\text{eff}} = 4 G \frac{A_0 (\lambda_{\text{max}} - \lambda_{\text{max}}^{-2})}{L_0 \lambda_{\text{max}}}$$

(30)

In this way, the oscillation frequency is then calculated by the equation:

$$f_{\text{calc}} = \frac{K_{y}^{\text{eff}}}{\sqrt{m}}/2\pi$$

(31)

The results are compared to the experimental oscillation frequencies, and reported in the Fig. 34. As it can be observed, the agreement is very good for CM0 for the LCOs before the transition, while the trends is opposite for the other cases: for CM1, 3 and 4 the calculation seems to agree with the measurements after transition, while it results overestimated before it. This could be attributed, for example, to an higher viscous contribute due to higher $CM$. In other words, such contribute could be addressed in the so called added mass (as the angular damping was individuated as the main responsible of the adsorption of the flow energy): despite it is commonly neglected because of its very low values in air, it could increase in conditions of high reduces frequencies (0.2-1 in our system) and large pitch angle (30-90 [°] in our system). During the coupled-mode fluttering of the wing, an hysteresis resulting from the phase lag between the motion and the aerodynamic forces occurs, especially during the detachments of the LEVs. These phenomena is the result of the highly unsteady flow and the dynamic stall encountered. For this reason, considering $\lambda_{\text{max}}$ instead $\lambda_s$ in the calculation of $K_{y}^{\text{eff}}$ may not surprise. Interestingly, this approach does not take into account any time dependence, as the unit of measure [s$^{-1}$] equals [Hz], and it comes from the recall force of the elastomers ([N]=[kg m s$^{-1}$]). Anyway, by increasing the values of the pre-stretching, the agreement tends to diverge (see the nonlinearity elastic force in section 3.3). The application of this approach is currently on investigation, by its integration in the phenomenological model.
3.8 **Flow visualization**

Since the aeroelastic study provided some interesting information, but the simulations, the phenomenological model and the analytical model missed a real prediction and explanation of the effects observed in our system, a flow visualization study has been carried out for investigating the evolution of the vortex and their effects on the structure during the main LCOs exhibited (see Fig. 35). Using the setup described in section 2.4, a careful study on the vortex shedding and its evolution varying some mechanical and aero-dynamical conditions has been conducted.

Along the different trajectories of the system, some LCO has been individuated also following the evidences emerged in the sub-section 3.4.2.

**Figure 35** - Flow visualization of LE vortex of a FLEHAP device in config.2 without extraction at 2.5 and 4.0 [m/s] obtained by smoke generator apparatus
Figure 36 - Smoke vortices visualization of static cases increasing $U$ with high stiffness, (a) at 2[m/s], (b) at 4[m/s], and dynamical ones with low stiffness at 3[m/s]: wind vanes in the middle of the span wise (c) and close to the chord wise axis (d), where end span vortex is observed.

Analyzing the coupling of the flow with the system when the fluttering conditions still not occur (Fig. 36a and 36b), several vortexes were observed at the confine layer of solid and fluid, while no LEVs were crossing the wing length; in these cases the flow was not modified before meeting the wing. Conversely, when the system exhibits a LCO, the flow direction was clearly modified (Fig. 36c), providing conditions in which the LEV, after having formed, run along the wing length (chord) for a certain distance. It is remarked that the system is characterized by a 3D behaviour, that means that the length of the span $S$ plays a rule in the contribution of the creation of lateral end wing (tip) vortexes, reported in Fig. 36d. This is the reason why an aspect ratio major of 2 is typically used in the experiments. The system showed different LCOs in the space of parameters investigated, which corresponded to different ways of exchanging the energy from the structure to the flow, and from the flow to the structure, basically, for the Reynolds number and reduced frequencies explored. The latter, as already stated, seemed to be regulated to the phase between the two main degrees of freedom of the system, pitch and plunge. The system, in fact, totally passive with no recall force on the pitch, as exposed in the sub-section 3.4.2, autonomously set such phase, following complex bifurcations.
3.8.1 Period Doubling

Concomitance of two different periods in the same LCO, in aeroelasticity and other fields named period doubling, in our system is characterized by an alternated phases between pitch and plunge. The study case of \( C=15 \text{ [mm]}, S=70 \text{ [mm]}, K_y^{\text{eff}} = 15 \text{ [N/m]}, U=3.5 \text{ [m/s]} \) is reported in Fig. 37 and Fig. 38 for a period doubling examination. This case coincides with the case of 3.5 [m/s] of Fig. 39, depth in this sub-section.

Figure 37 - Trajectory (a) with LE in blue and TE in red, and time-progress of PP and \( \theta \) (b) of the study case \( C=20 \text{ [mm]}, S=70 \text{ [mm]}, K_y^{\text{eff}} = 15 \text{ [N/m]}, \) at 3.5 [m/s].

Figure 38 - Analysis of the evolution of vortexes during the concomitance of two different alternated periods, through the flow visualization, in the study case \( C=15 \text{ [mm]}, S=70 \text{ [mm]}, K_y^{\text{eff}} = 15 \text{ [N/m]} \) at 3.5 [m/s].
With the measurement accuracy of our experimental setup, the two LCO show the same frequency oscillation and PP displacement, but very different pitch angles and detachment time of LEVs, and sensible different phase between pitch and plunge motions. During the period doubling considered, the wing reaches very large pitch angles, close to 90 [°], near the inversion of the displacement, which imposes the inversion to the angle. For the first period the rotational inversion is enabled by the large leading edge vortex (LEV) shedding (see Fig. 38_4), while for the second period it is avoided, as the vortex is still attached to the wing layer, letting the wing creates a TE vortex (see Fig. 38_14). From a chronological point of view, the second period is the first one which is formed, as it came from the evolution of the LCO at lower wind speed, and energetically it corresponds to a lower energy state, with respect the first period. In fact, the latter seems to anticipate the formation of the LEV, and thus it’s shedding, providing a faster and freer inversion of the wing. This anticipation is translated in a sensitive increase of the phase between pitch and plunge, observable in the negative part of PP displacement in Fig. 37b. It is curious as the system use such phase between its main two modes as like as a mechanical drive in order to handle the input energy from the flow, which is quasi-laminar before interacting with the structure.

3.8.2 LCO Analysis

From Figure 39 three very different LCOs can be distinguished:
- In the first one the fluttering is characterized by a Light Dynamic Stall with phase close to zero, the frequency can be close or higher of several [Hz] from the natural one, the amplitude is modest, less than one chord (see Fig. 35a, Fig. 36c and Fig. 39 at 2.5 [m/s]).
- The second is characterized by a Deep Dynamic Stall with very large Leading Edge Vortex (LEV) at positive pitch angles which can reach 90 [°] in the asymmetric cases (see Fig. 35b and Fig. 39 at 4.0 [m/s]). The inversion of the wing is typically close to the inversion of the displacement, with a pitch and plunge phase typically around 20 [°], and when the vortex detached from the wing the Pivot Point still does not invert its motion. Both the PP and frequency reach a maximum before decreasing. The more evident effect of this LCO is that the amplitudes of PP, which can reach more than two chords, are always bigger than the Trailing Edge one. This seems to be the condition for which the system absorbs the maximal energy from the flow. Strangely, the typical phase between pitch and plunge for which the maximal work from the flow to the wing reported in the literature study is 90 [°].
- This last condition is satisfied by the third case, for which both the amplitude of PP and θ suddenly decrease by changing the phase to value slightly less or more than 90 [°]. In most of cases this brings to the first condition of symmetric motion, but this is not the case for many experiments, as the latter also depends on CM and PP positions.
Figure 39 - Flow visualization of the fluid-solid interactions during the different LCO (C20S70K15).
A symmetric Deep Dynamic Stall is characterized by a detachment of the LEV in forward with respect the maximal stretching of the elastomers (see Fig. 39 at 5 [m/s]). The LEV detached at about the pre-stretching position of the elastomers, then the wing start to invert the motion and the moment acting on by, united to the inertia of the system, provide sufficient force to continue the stretching until the maximal amplitude of \( P \). When the \( P \) starts to decrease, the wing is already in negative pitch angle, ready to form another LEV, later detached at about the equilibrium position of the elastomers.

### 3.9 Summary

The aeroelastic study provided valid information about the physic of the device, which is not described by any model in the literature, presenting many relevant aspects, important for the comprehension and the optimization of the energy harvester. These include:

- Merge of mechanical and hydro dynamical frequencies (marginal curves) as result of the coupling between the oscillating wing and the flow wake. This is important for the estimation of the onset speed at which the device starts to show a stable LCO.

- Presence of different LCOs in a wide range of flow speed when the parameters are fixed.

- Increase of oscillation frequency and presence of frequency branches during LCOs. Its maximization is important for the electrical extraction.

- Autonomous change of phase between pitch and plunge. This is related to the attitude of the system to manage the incoming energy in various conditions. At least two very different behaviours occur in the change of phase.

- Individuation of a specific LCO as the condition of maximal adsorption of energy from the flow. The phase between pitch and plunge in this case is always less than 90 [°].

With these observations in mind, some considerations for the design of the aeroelastic system can be done. Fixing the elastic constant \( K_{yy}^{eff} \) and increasing the length of stretching of the elastomers \( L_s \), decreases the onset speed \( U_{cr} \); increasing the translational mass \( m_{plunge} \) or the centre of mass \( CM \), decreases \( U_{cr} \), but increasing \( m_{plunge} \) the frequency decreases. By decreasing the shear modulus of the elastomers \( G \) and increasing their section \( A_0 \), can decrease the total length of the device, as large amplitude at small deformation forces can be obtain with small stretching, with high frequencies. This also increases the non-linearity of the elastic system, which helps to rapidly increasing the oscillation frequency along the wind speed. The condition of the pre-transient LCO, for which the parameters in interest show the optimal values for the electrical extraction (maximization of amplitude and frequency of oscillation and minimization of the pitch and plunge phase), must be guaranteed for the widest possible flow speed range of the specific application.
4 Electromagnetic Coupling

The electromagnetic coupling (EMc) is the main strategy adopted to extract the electrical energy from the wind. This also corresponds to the more mature one, since the literature reports many similar studies, and since the studies led in my Master thesis already explored its application in FLEHAP device. This Chapter aims to present the main founding on this topic.

Figure 40 shows a representative centimetre-sized FLEHAP prototype using EMc, and Fig. 42 shows the specific EMc adopted for the device, obtained by a Finite Element Method Magnetics (FEMM) simulations. An array of permanent magnets in alternated polarity configuration allows obtaining a high spatial magnetic flux gradient.

The change of magnetic flux within the coil, represented in section in Fig. 41, gives rise to an electrical potential at the ends of the wire when the system enters in a LCO, in which the PP exhibit a vertical (in Fig. 41 horizontal) quasi-harmonic motion. When the latter are connected to an electrical load resistance, the current flows from the coil, then the power is delivered to the load. It should be mentioned that the electromagnetic coupling would produce an additional force that interplays with the structural responses of the oscillating system 34,59.

According to Faraday’s law, the electromotive force $V_{emf}$ which is produced on the terminals of the coil with time constant electromagnetic field equals to 59:

$$V_{emf} = \oint_{ coil} (\dot{y} \times B) \, dl$$

(32)

where $y$ is used to represent the displacement of the structure, the dot means differential to the time and hence $\dot{y}$ is the relative velocity of the coil and the magnet, $dl$ is the vector of each segment of the coil and $B$ is the flux density at each coil segment.

Figure 40 - A centimetre-sized FLEHAP prototype using EMc, during Fluttering (a), and the same prototype without wind and equipped with the storage system (b). The magnets width is 10 [mm].
The acquired electrical signal from the coils for a representative case of fluttering, using the prototype depicted in Fig. 40, is reported in Fig. 42. As it can be seen, an alternated voltage is generated during the harmonic motion of the coils, characterized by a zero voltage when the coils are at the maximum displacement. In fact, as in these conditions the velocity is \( \dot{y} \) null, \( V_{emf} = 0 \).

4.1 Simple damped model

In a first reasonable approximation, the relationship between the amplitude of the velocity and the electromotive force can be considered linear:

\[
V_{emf} = B_{avg} l_{coi} \dot{y} = C_{Bl} \dot{y}
\]  

(33)

and the electromagnetic force acting on the Pivot Point of the wing can be given by:

\[
F_{em} = -B_{avg} l_{coi} i = C_{Bl} i
\]  

(34)
where \( i \) is the induced current in the coil, the proportionality constant \( C_{Bl} = B_{avg} l_{coil} \) is called the electromagnetic coupling coefficient, \( B_{avg} \) is the average flux density across the coil range and \( l_{coil} \) is the total length of the coil.

Consequently, the equations of motion that couple the aeroelastic system and the electromagnetic transducer with consideration of an electrical load resistance in the electrical circuit can be written as:

\[
m(\ddot{y} + 2\xi \omega \dot{y} + \omega^2 y) = F_y(y, \theta) + F_{em}
\]

\[
V_{emf}(y, \theta) - L_c \frac{di}{dt} - (R_c + R) i = 0
\]

where \( m \) is the mass of the system, including the structural mass (wing and translational mass) and an equivalent added fluid-dynamic mass. In the above expressions, \( \omega \) denotes the pulsation of the oscillation, and the mechanical damping ratio \( \xi \) is related to the viscous damping coefficient \( D \) by \( \xi = D / (2\omega m) \). Furthermore, the coil winding is modeled by an electrical resistance \( R_c \) and an inductance \( L_c \). \( R \) is the external load resistance and \( F_y \) represents the aerodynamic forces in the considered direction.

### 4.2 Experimental setup

The experimental measurements have been performed in an aspirating subsonic wind tunnel with specifications reported in section 2.1 using a mature FLEHAP prototype (Technology Readiness Level TRL 4-5) armed with an electromagnetic coupling.

A typical centimetre-sized device for this kind of study, depicted in Fig. 40, adopts two stretched poly-isoprene elastomers of length \( L=0.12 \) [m] and \( G=0.54 \) [MPa] arranged parallel to the flow (first configuration, see Fig 11a). A brass rotational axis of length of 0.09 [m] and diameter of 1 [mm] is connected with them by 3D-printed coil-supports, placed in the middle part of the two elastomers. A wing composed by a semi-rigid poly-vinyl-acetate foil with thickness of 0.2 [mm], Span \( S=0.085 \) [m] and Chord \( C=0.035 \) [m], glued to a 3D-printed tubular support was used. Nd\(_2\)Fe\(_{14}\)B permanent magnets of 10x10x3 [mm\(^3\)] and 40 [MOe], coils of \( L_c=10 \) [mH] and \( R_c=150 \) [\( \Omega \)], were chosen for the EMc. All the parts of the frame were 3D-printed, included a fast and precise mechanism for pre-stretching and calibrating the elastomers recall force, and a mini-spring system for spacing the magnets from the coils.
4.3 **Experimental Results**

4.3.1 **Effect of a pure resistive load**

As mentioned in the previous paragraphs, an additional damping is induced to the system when an electrical current is left to pass through the coils as a result of the electromagnetic coupling. In fact, a reverse electromagnetic induction occurs as the result of the motion of the electrical charges in the wire, providing a magnetic field of opposite direction with respect the fixed magnets. The coils are connected in series to an external circuit with a variable resistive load $R$. First, the fan is turned on (at a given voltage which corresponds to a certain flow velocity) and the wing is left to reach the steady fluttering regime without energy extraction, by setting $R = 4 \text{ M}\Omega$ (i.e. equivalent to the open circuit condition). Subsequently, we reduce the resistive load to a specified value, and the resulting voltage $V_{emf}$ is measured by a PicoScope 4224 digital oscilloscope. The mean dissipated power over the applied resistance is then evaluated as:

$$P_{out} = \frac{V_{rms}^2}{R}$$  \hspace{1cm} (37)

where $V_{rms}$ is the root mean square of the alternated voltage $V_{emf}$ measured (see Fig. 42).

Figure 43 shows the effect of a pure resistance load $R$ on the power available with increasing the wind speed $U$. As it can be noted, the maximal power is observed at a resistance load $R$ always larger than the internal load $R = 300 \text{ \Omega}$.

![Figure 43 - Experimental measurement of Power $P$ dissipated on a resistance load $R$ increasing the wind speed $U$ with a cm-sized FLEHAP device exploiting an EMc.](image)
This is because, as the electrical extraction is carried out from the plunge term of the coupled-mode flutter, in which the mechanical DOF pitch and plunge (angular and spatial motions) are strictly coupled, the electromagnetic damping, in antagonism with the inertial contribution of the moving mass, drastically affects the motion of the wing. At $U = 2.5$ [m/s] the maximum power (reached for $R = 10$ [kΩ]) is around 1 [mW], already an interesting amount for previously cited applications. The power level then increases with the flow velocity while the value of the optimal load decreases, as one can expect. Note that the prediction of the optimal load while varying the mechanical and electrical parameters would require the analysis of the electromagnetic-aeroelastic system and not of the electric side only. The output power provided $P$, varies monotonically to a value of 14 [mW] at $U = 4.0$ [m/s] with $R = 1$ [kΩ], then reaches a maximum of 15 [mW] at 4.5 [m/s] with $R = 1$ [kΩ], and decreases.

The output voltage ($V_{rms}$) and the calculated current $i$ are reported in Fig. 44, in order to provide an idea about the order of magnitudes of the signals. Along with the maximal electrical power $P = P_{out}$ (Fig. 45a), we calculate the input power provided by the flow, estimated as:

$$P_{in} = \frac{1}{2} \rho_f A_{swept} U^3$$

(38)

in order to evaluate the global efficiency of the system (Fig. 46f), defined as:

$$\eta = \frac{P_{out}}{\frac{1}{2} \rho_f A_{swept} U^3}$$

(39)

where $A_{swept} = A_{PP_y} S$ and $A_{PP_y}$ is the vertical displacement swept by the wing, taken as the maximum between the leading and trailing edge oscillation amplitude, and reported in Fig. 45c as effective excursion.

Figure 44 - Root mean square of Voltage (a) and electrical current (b) measured at the end of coils. In (a) the $V_{rms}$ and $R$ related to the maximal power are reported with red circles.
As it can be observed from Figure 45f, the maximum efficiency of electrical extraction occurs when the system, in open circuit condition, is in a LCO in pre-transition. As reported in the sub-section 3.4.2 this condition maximizes both frequency and amplitude of oscillation (see Figures 45 b-c). From Fig. 45b, by comparing the relative maximum in frequency in the two curves, it can be also observed that the EM damping induces delay on the LCOs evolution. This brings important consequences in terms of the extraction of power, as if the transition is delayed, the pre-transition LCOs are able to remain stable for higher $U$, allowing the system to let flow more electrical current in the coils. Thus, despite the maximum efficiency occurred in the pre-transition LCO, at 3.5 [m/s] in this case, the maximum of power is achieved at 4.5 [m/s].

Figure 46 compares the performance of our device with those of other energy harvesters by Fluid-Solid Interaction (FSI) recently proposed $^{62-67}$. We consider the output power divided by the swept area $^{36,68}$ (defined as it follows from Eq. 38): for our device, and in particular at the highest tested value of $U = 4$ [m/s], we have $P_{out}/A_{swept} = 0.27$ [mW/cm$^2$]. If we consider the power per plan form area, we have: $P_{out}/A_{wing} = 0.47$ [mW/cm$^2$]. Based on these metrics, the system collocates in an interesting position with an efficiency close to that of micro turbines $^{63}$. Besides, we confirm the competitiveness of flutter-EMc-based harvesters $^{36}$. 
4.3.2 **Effect of change of mass on plunge with EMc**

After the kinematic study reported in the sub-section 3.4.2.3, two coil of resistance $R_c=300$ [Ω], inductance $L=10$ [mH] and external diameter of 9 [mm] have been fixed in front of the equilibrium position of the magnets (see Fig. 47), at a distance $d_{mc}=1.5$ [mm]. As it can be observed in Fig. 48, the output power varies with the increase of the number of magnets. When a first amount of mass is added to M1 (M3) the output powers decrease, but showing similar values increasing $U$. On the other hand, a stronger dependence from the external resistance loads is observed, also because the increased coupling coefficient. For M3 the system is completely damped for $R=100$ [Ω] at $U=5.5$ [m/s] and for 2 [kΩ] at 6 [m/s]; for M4 even beyond $U=5$ [m/s].
In Fig. 49b the different external load dependence occurring in the three cases is shown. As it can be noted the optimal load is about 300, 500 and 800 [Ω] for M1, M3 and M4, respectively.

As it can be observed in Fig. 27 of the sub-section 3.4.2.3, the amplitudes of oscillation resulted higher than for the case M0, but on the other hand the oscillation frequencies were lower, and this partially explains the difference on the power generated. M4 showed better performance respect M1 and M3 (see Fig. 49a). This is because, a trade-off between the oscillation frequency, \( PP \) amplitude and EM damping occurred. Furthermore, the increased inertia of the system helped to contrast the counter-magnetic field induced by the electrical dissipated on a resistance load.

Figure 48 -  
Power dissipated an variable resistance load \( R \) varying the number of moving magnets placed on the rotational axis of the wing.

Figure 49 -  
Maximum power generated changing the number of magnets (a), and related optimal external loads (b).
4.3.3 Storage by integrated electronics

In the previous sub-sections, the calculated dissipated power was estimated from the measurements of the output voltages, applying Eq. 37, over various resistance loads. By the way, in order to store the electrical energy in battery, or in our case external capacitors, the alternated current must be rectified (AC-DC conversion), and then stabilized to a constant voltage. The latter is necessary to the real electrical users, such as sensors and radio-transmitter (DC-DC conversion). To do so, the use of an integrated circuitry is necessary. Two commercial modules, showed in Fig. 50, have been utilized for the storage investigations, and are presented in this sub-section.

Figure 50 - Integrated electronics used for the storage investigation. In (a) EH300 module, from Advance Linear Devices Inc., in (b) MB39C811 module, from Cypress Semiconductor Corp. In (c) one euro coin for a scale reference of the integrated circuits.

Fig. 51 shows a comparison between the power dissipated on the optimal resistance load, obtained by the use of the device reported in Fig. 40, and the power supplied to the capacitors of two commercial integrated circuits.

Figure 51 - Experimental measurement of power \( P \) dissipated on a resistance load \( R \) increasing the wind speed \( U \) (a) with a cm-sized FLEHAP device exploiting an EMc (depicted in Fig. 41b). Comparison between the power dissipated on the optimized resistance load and the power generated during the storage of the electrical energy in capacitors using two commercial EH modules (b).
The first circuit tested is EH300 from Advanced Linear Device Inc., an Energy Harvesting Module completely self-powered and always in the active mode. It can accept energy from many types of electrical energy sources and store this energy to power conventional 3.3 [V] and 5.0 [V] electrical circuits and systems. It is designed for low power intermittent duty cycle sampled data, condition-based monitoring and extreme lifespan applications.

Such module can accept instantaneous input voltages ranging from 0.0 [V] to +/-500 [V] AC or DC, and input currents from 200 [nA] to 400 [mA] from energy harvesting sources that produce electrical energy in either a steady or an intermittent and irregular manner. The output voltage is not regulated. The second module tested is MB39C811, from Cypress Semiconductor Corporation, another high efficient buck DC/DC converter IC that adopts the all-wave bridge rectifier using low-dissipation and the comparator systems. It achieves the energy harvest solution for the energy source of the high output impedance such as the piezoelectric transducer. Both the circuit possess a similar storing logic, physically composed by two capacitors \( C_{in} \) and \( C_{out} \); the harvested energy is firstly stored on the \( C_{in} \) and then in \( C_{out} \) to operate the application block. If the size of these capacitors is too big, it would take too much time to charge energy into these capacitors, and the system cannot operate frequently. On the other hand, if these capacitors are too small, enough energy cannot be stored on these capacitors for the application block. Consequently, the sizing of \( C_{in} \) and \( C_{out} \) is very important.

Fig. 52 shows the functional description of the EH300 module. The voltage on the onboard storage capacitor bank is \( +V \), which is also the positive supply voltage switched to power the output load. Initially, \( +V = 0 \) [V]. The module's internal circuit monitors and detects the \( +V \) value. When the EH device is working, \( +V \) increases and when it reaches \( V_H \), the module output (\( V_P \)) is enabled and turned to the ON state, supplying power to the load, such as a microprocessor and/or a sensor circuit. If the external energy input is available, output \( V_P \) remains in the ON state continuously, unless the power required by the load exceeds the power generated by the EH device. In this case, \( +V \) decreases until it reaches \( V_L \) and output \( V_P \) switches to the OFF state. At this moment, the EH device charges again the capacitor and the cycle restarts. A similar logic is adopted also by the MB39C811 IC.
Figure 53 - Digital oscilloscope signals from EH300 connected to FLEHAP device at 5 [m/s], changing $R$ in order to simulate the effect of an dissipation (for instance a radio transmission) on the storage of the system.

Fig. 53 presents how the EH300 module works with FLEHAP device in condition of 5 [m/s] when a resistive load $R$ is applied to $C_{out}$. For $R=100$ [$\Omega$] the system cannot maintain the threshold voltage of 3.61 [V], causing an ON-OFF cycle with a period of few seconds. If $R=1$ [k$\Omega$], the system keeps a constant voltage of 3.4 [V], until $R=100$ [$\Omega$] is again applied. When $R=3$ [k$\Omega$] the system charges the output capacitor to the maximum value of 6.9 [V]. A similar behaviour is observed using the MB39C811 IC.

As reported in Fig. 51b, the power available at the output of these modules is always lower than the power available on the optimal resistive load. In fact, the equivalent load seen by the EH device is determined by the IC circuitry, then limiting the efficiency of the system.

A new approach for the design of a dedicated electronic, able to operate a maximum power point tracking (MPPT) of the input signal, has been published in 69, and is reported at the end of this thesis in the Appendix A.
5 **Dielectric Elastomer Generators**

Dielectric elastomers, extensively used and studied as active material in actuation mode, can also be exploited as generators to create electrical energy\(^{70,71}\). Coupling them with two stretchable electrodes, if they are stretched by an external force and electrical charges are placed on their surfaces, elastic stresses acts in the relaxing works against electrical field pressure, increasing electrical energy. In this process, opposite charges on the two electrodes are pushed further apart as the film contracts and increases in thickness, while similar charges are brought closer together as a result of the decreasing area of the elastomers (see Fig. 54), thus increasing energy density. These changes increase the voltage of the charges, and the created energy can be harvested.

A DEG cannot be an autonomous energy scavenger, because it needs to be supplied, or ‘charged’, in order to create energy, therefore the use of both DEGs and EMc strategies consists in a beneficial synergy.

Nowadays, because of the complicated inter-correlations between their parameters, the design of this kind of materials in both the generator and actuator modes, are not yet optimized\(^{72,73}\). Several efforts have been obtained in different areas of application, while a common procedure in order to find a good trade-off between advantages and disadvantages in terms of final global efficiency, is still unknown and on research.

For instance, many applications of DEGs have been reported for energy harvesting from the sea current, providing stimulations and suggestions for their architecture designs\(^{74-78}\). Nevertheless, each application must be properly investigated for the system optimization, as the input forces, electrical voltages, and operational times vary from one case to the another.

It is important to note that the actual state of art misses complete studies regarding the durability and environment conditions behaviour of this materials, crucial aspects of their technological applications. Anyway, because of their promising energy density with respect other technologies\(^ {71,79} (>1 \text{ [J/g]})\), this next decade will sure see an huge development of DEGs, in the Research and consequently in technological applications.

![Figure 54 - Illustration of the nature of a Dielectric Elastomer Generator and its principle for scavenging energy: the elastic recall works against the electric field pressure due to the polarization.](image-url)
5.1 Operation of DEG in FLEHAP

In order to describe the operations of DEGs in the FLEHAP device, and find out the main important parameters to optimize it, an analytical model has been developed.

A pure shear extension model was adopted to describe the deformations because of the geometry of the DEGs in the system (Figure 55), consisting in belt-shaped elastomers (e.g. in a centimetre sized device: length $L_{1s} = 10$ [cm], width $L_{2o}=2$ [cm], thickness $z_0 = 150$ [μm]). Here, the bounds involve, in both side, all their width length $Y$: since the maximal stretching $y_{max}$, as the aeroelastic experiments provided (sub-section 3.4.2), should not exceed 1.5, the decrease in width $dL_{2o}$ can be considered negligible in this first approximation. The assumptions of this model are the isotropy, incompressibility, homogeneity and pure elasticity of the dielectric material, in absence of defects and mechanical losses. Therefore, $0 \leq \nu_d \ll \tau_m^{-1}$, $T >> T_g$, $\nu_p = 0.5$, $G'' = 0$, where $\nu_d$ is the deformation frequency, $\tau_m$ is the elastomer characteristic timescale ($\approx 10^{-4}$ sec), $T_g$ is the Glass Transition temperature ($\approx -120[°C]$ for silicones), $\nu_p = \frac{Y}{2\mu}$ − 1 is the Poisson ratio, around 0.49 for silicones, $Y$ is Young modulus, $G$ is the shear modulus and $G''$ is the shear loss modulus. A perfect stretchability of the electrodes, without resistivity and stiffness induced, and the geometrical coincidence of electrodes and elastomers surfaces were also assumed.

![Figure 55 - Schematic and description of variables of the aeroelastic system exploiting EMc and DEGs.](image-url)
5.1.1 Deformation of DEGs in FLEHAP

Considering the deformation of half a portion of a single elastomer due to the relative movement of the Pivot Point of the wing (PP) with respect the bounds ($V_1, V_2$), assuming a pure vertical harmonic motion of PP, the following analysis can be conducted with referring to.

Defining the pre-stretching length $L_s = L_0 \lambda_s$ and the deformation length $L' = L_0 \lambda_s y = L_0 \lambda'$, and considering a transversal deformation with respect the elastomers pre-stretching position and the wind direction, the plunge motion $PPy$ is written as

$$PPy(t) = L'(t) \sin(\alpha) = \sqrt{(L'^2 - L_s^2)} = L_0 \sqrt{\lambda_{max}^2 - \lambda_s^2} = L_0 \lambda_s \sqrt{y^2 - 1}$$ (40)

Relative deformations in terms of $y$ direction can be expressed by the follow relations:

$$\lambda' = \frac{PPy^2}{L_s^2} + 1 = \frac{PPy^2 + L_0^2}{L_s^2} = \sqrt{y^2 + 1}$$ (41)

At equilibrium, that means in absence of external forces and applied voltage, $F_y = 0$ and $L = L_s$; an external vertical mechanical perturbation is managed by the aeroelastic the system as:

$$F_y = 4 F_{el} \frac{PPy}{L_s} = 4 \sigma m A_0 \sin \alpha = \frac{4 \sigma m A_0}{L_s} L_0 \sqrt{\lambda'^2 - \lambda_s^2} = K_y^{eff} PPy$$ (42)

where $\sigma_m$ represents the mechanical stress on the active section of the elastomers $A_0$, and $F_{el}$ is the force perceived by one portion of elastomer,

$$F_{el} = \sigma_m A_0 = A_0 \left( \lambda' - \frac{1}{\lambda_s^2} \right) \left( G + 4 C_2(I_1 - 3) + 6 C_3(I_1 - 3)^2 \right)$$ (43)

and $K_y^{eff}$ is the effective elastic constant of the system (four portions of elastomer) in the $y$-direction, made explicit as

$$K_y^{eff} = K_0_y = \frac{4 \sigma m A_0}{L_0 \lambda_s}$$ (44)

The mechanical frequency of the system (natural frequency) can be written in terms of the pre-stretching

$$\nu_n = \frac{1}{2\pi} \sqrt{\frac{K_y^{eff}}{m}} = \frac{1}{2\pi} \sqrt{\frac{4 \sigma m (\lambda_s) A_0}{m L_0 \lambda_s}}$$ (45)

where $m$ is the mass of the translational part of the system, composed by the wing, its rotational axis and connectors, and the coils (the mass of the elastomers are neglected). The system prototype oscillates at a frequencies $\nu_o$ higher than $\nu_n$, as a result of the nonlinearity of the
aeroelastic system, reported in Chapter 3. However, in order to maximize the EMc extraction, a $v_0$ between 15 and 25 [Hz] is typically used, thus a reference value of $v_0 = 20$ [Hz] is considered in this model. When the system exhibits the coupled-mode fluttering self-excitation, the vertical harmonic motion of the $PP$ and periodical angular motion of the wing $\theta$ can be expressed, in a first approximation, respectively as

$$PPy(t) = A \sin(2\pi v_0 t)$$  \hspace{1cm} (46)

$$\theta(t) = (\theta_{\text{max}} - \theta_{\text{min}}) \sin(2\pi v_0 t + \phi)$$  \hspace{1cm} (47)

where $\theta_{\text{min}}$ and $\theta_{\text{max}}$ are the maximum and minimum measured pitch angles of the wing (counter clock wise / nose-up), $2\pi v_0 = \omega_0$ is the pulsation of the oscillation, and $\phi$ is the phase between pitch and plunge motions ($\theta$ and $y$ displacement respectively). As observed from the experimental study of the LCO (sub-section 3.4.2) and the study related to the electrical energy extraction through EMc (section 4.3), the system autonomously adapts $\phi$ in order to achieve a proper condition of exchange between the aeroelastic and electrical energy. For each LCO of the wing, the deformation of the DEGs will be twice, due to the geometry adopted, so the frequency of deformation reads:

$$v_d = 2 \ v_0$$  \hspace{1cm} (48)

5.1.2 **DEG Cycles**

The operation acting during a period of oscillation depends on the approach that has been identified with respect the application of the system. A typical generator mode approach utilized in DEGs is the operation cycle reported in the schematic of Fig. 56, following the order of states 0-A-B-C-D-A: after a pre-stretching (0-A), the system is deformed by an external force (B), polarized when it reaches the maximum deformation (C), and depolarized when it is contracted by the recall force of the elastomer (D).

A voltage amplification occurs if the active phase is conducted at constant charge.

During the phases of polarization and depolarization an electro-mechanical balance occurs: when a voltage $V_{in}$ is applied on the surfaces of DEG $A' = L'L_{2\theta}$ (in the maximal stretching of elastomers $\lambda' = \lambda_{\text{max}}$), the electric displacement $D$ reads

$$D = \varepsilon_0 \varepsilon_r E = \frac{Q}{L_{2\theta} L_{1\theta} \lambda'},$$  \hspace{1cm} (49)

where $\varepsilon_0$ is the vacuum permittivity constant $\varepsilon_0 = 8.85 \times 10^{-12} \ [\text{F} \ \text{m}^{-1}]$ and $\varepsilon_r$ is the relative permittivity of the material.

The charge $Q$ on the surfaces of DEG can be estimated as

$$Q = \varepsilon_0 \varepsilon_r E \left( L_2 L_1 \right) = \frac{\varepsilon_0 \varepsilon_r V_{in} L_1 L_2}{z} = \frac{\varepsilon_0 \varepsilon_r V_{in} L_1 L_2 L_{2\theta} \lambda_{\text{max}}^2}{z_0} = CV_{in}$$  \hspace{1cm} (50)
where $C$ is the effective capacitance of DEG, expressed as:

$$C = \varepsilon_0 \varepsilon_r \frac{L_\lambda L_\varepsilon}{z_0} \lambda_{\text{max}}$$  \hspace{1cm} (51)$$

Such voltage $V_{in}$ causes an electric field $E = \frac{V \lambda'}{z_0}$ across the thickness of DEG.

Analogously to Dielectric Elastomers Actuators (DEA), an induced deformation due to the well-known Maxwell stress, can occurs when a voltage $V = Ez$ is applied to the DEG: its true stress reads a magnitude of

$$s_{\text{maxwell}} = \varepsilon_0 \varepsilon_r E^2$$  \hspace{1cm} (52)$$

while its nominal stress $\sigma_e$ can be written as

$$\sigma_e = \varepsilon_0 \varepsilon_r E^2 \lambda_D^{-1} = \varepsilon_0 \varepsilon_r \frac{V^2 \lambda_D}{z_0^2}$$  \hspace{1cm} (53)$$

As $\sigma_e$ is applied when the DEG is already under the application of mechanical stress $\sigma_m$
\[
\sigma_m = \left(\lambda_D - \frac{1}{\lambda_D^2}\right) \left(G + 4C_2(\lambda_D^2 + \lambda_D^{-2} - 2) + 6C_3(\lambda_D^2 + \lambda_D^{-2} - 2)^2\right) \tag{54}
\]

every instant is accompanied by an stabilization for which such balance occurs. When the DEG is subject to a combination of external force \(F\) and applied voltage \(V\), both cause a reduce in thickness and an increase in area when charging occurs, while when the elastic recall force occurs, they rules antagonistically, as the mechanical energy is transformed in electrical one

\[
\sigma_f(t) = \sigma_m(t) + \sigma_e(t) \tag{55}
\]

where \(\sigma_f(t)\) is the nominal stress of DEG when the balance occurs.

Both the phases B-C and D-A can show a change in shape due to this actuation effect, if the operation voltage is higher of a certain value, depending on the characteristic of the DEG.

As the use of this kind of smart materials started several decades ago for actuation proposes, significant efforts have been made to decrease the driving voltage of DEAs while maintaining good actuation performance. High performance materials, with dimensions of DEGs used in FLEHAP, start to exhibit actuation phenomena over about 1 [kV] [20]; it is easy to demonstrate that until a certain value of tension, namely \(V_{act}\), the system will not be affected by the electric field through the film, while over it, an increase of stiffness of the aero elastic system can be observed.

For example, \(V_{in}=1 \text{ [kV]}\) correspond in an experimental prototype of FLEHAP using \(G=0.5 \text{ [MPa]}, L_s=100 \text{ [mm]}, L_2=20 \text{ [mm]}, z_0=0.15 \text{ [mm]}, \varepsilon_r=3\) and \(\lambda=1.4\) to a net maxwell force of 10 [mN], while the mechanical force is about 200 [mN]; in this case the electrostatic force can be neglected, and any balance has to be taken into account.

Let’s call this case, in which the electrostatic contribution can be neglected, CASE a, and CASE b the cases in which an actuation component affects the stiffness and the balance states of DEG.

5.1.2.1 **CASE a: Generation**

In the CASE a, the change in capacitance between B and C is proportional to the relative deformation, following the expression

\[
\frac{C_B}{C_c} = \left(\frac{\lambda_{max}}{\lambda_s}\right)^2 \tag{56}
\]

As the charge remain \(Q_{in} = Q_B = Q_c\) and \(Q = CV = constant\), considering no electrical dissipation, \(C_BV_B = C_cV_c\), \(V_{out} = \frac{V_c}{V_{in}} = \frac{C_B}{C_c}\), the voltage \(V_c\) can be estimated as
\[ V_C = \frac{C_B}{C_C} V_B = \left( \frac{\lambda_{\text{max}}}{\lambda_s} \right)^2 V_B \]  \hspace{1cm} (57)

As the electrical energy stored in the DEG reads \( U_e = \frac{1}{2} CV^2 \), the electrical energy associated to one semi period of wing oscillation is

\[
U_{\text{scav}_{T/2}} = U_{e_C} - U_{e_B} = \frac{1}{2} C_C (\lambda_{\text{max}}) V_C^2 - \frac{1}{2} C_B (\lambda_s) V_B^2 \\
= \frac{1}{2} \varepsilon_0 \varepsilon_r \frac{L_1 L_2}{Z_0} \left( \lambda_{\text{max}}^2 V_C^2 - \lambda_s^2 V_B^2 \right)
\]

Specifically,

\[
U_{\text{scav}_{T/2}} = (U_{e_{T/2,\text{ out}}} - U_{e_{T/2,\text{ in}}}) - U_{e_{\text{loss}}} = \int_0^{T/2} \mathcal{V} \mathcal{I} \, dt \hspace{1cm} (59)
\]

where \( U_{\text{loss}} \) represents the electrical losses, and, as the storable electrical energy directly depends by the mechanical \( U_{m_{\text{loss}}} \) (viscous) and the electrical \( U_{\text{loss}} \) (leakage and supplier circuit) losses, that can be summarized in the term \( U_{e,m_{\text{loss}}} \), the mechanical adsorbed energy \( U_{m_{T/2}} \) reads

\[
U_{m_{T/2}} = U_{e_{T/2,\text{ out}}} - U_{e_{T/2,\text{ in}}} + U_{e,m_{\text{loss}}}
\]

In fact, as a part of the mechanical energy from the ambient \( U_{\text{amb}} \) is returned to the environment, \( U_{m_{T/2}} \) is the effective mechanical energy stored in the elastomers when a deformation occurs.

Consequently, the final DEG efficiency, taking into account the viscoelastic losses, the dielectric losses and the Ohmic losses at bulk and surface level, is defined by

\[
\eta_{\text{DEG}} = \frac{U_{\text{scav}_{T/2}}}{U_{m_{T/2}}}
\]

The energy density express in terms of mass of DEG is commonly used to compare different system; for a semi period of oscillation it then reads:

\[
u_e = \frac{U_{\text{scav}_{T/2}}}{L_1 L_2 L_3 \rho_{\text{DEG}}}
\]

where \( \rho_{\text{DEG}} \) is the density of the composite material (dielectric + electrodes), and it contains both the active and the passive part of material (in this model assumed to be overlapped).

In such CASE a, the mechanical energy is not affected by the electrical one, as the stiffness of the system remains the same before and after applying the voltage: a simple balance within them will occur.

For this case, an LCO (two semi-period of oscillation) is described by the operations cycle of Fig. 56: 0-A-B_{T/2}-C_{T/2}-D-A-B_{T}-C_{T}-D-A.
If the charge $Q = C(t)/V(t)$ is maintained constant on DEG during a deformation, considering a cyclic deformation in time expressed by

$$\sigma_{m}(t, \omega_{0}) = \frac{F(\lambda_{\text{max}})}{A_{0}} \cos(\omega_{0} t)$$

(63)

the change in capacitance $C(t)$ can be written as

$$C(t, \omega_{d}) = \varepsilon_{0} \varepsilon_{r}(\omega_{d}) \frac{L_{10} L_{20}}{\varepsilon_{0}} (\lambda'(t)/\lambda_{s})^{2}$$

(64)

where $\omega_{d} = 2\pi \nu_{d}$ is the pulsation, $\lambda_{s}$ is the pre-stretching and $\lambda'$ is the stretching due to the deformation. In general, the system can deform the DEGs in a large spectrum of frequency (from few [Hz] to 50 [Hz], depending by the application, i.e. the kind of fluid and flow speed), and such frequency dependence is normally much stronger on the final performances as lower is $E$ and higher is $\varepsilon_{r}$.

Being the system typically characterized by small stretching ($\lambda_{\text{max}} < 1.5$), the dependence of the permittivity by the stretching can be neglected, as the studies of Tagarielli et al. reported.

This described condition (deformation with constant charge), has been already demonstrated experimentally with the FLEHAP device, measuring a change in voltage of DEGs $V_{c}(t)$ due to the change in $C(t)$ when a charge $Q$ is maintained constant on their surfaces.

5.1.2.2 CASE b: Actuation and Generation

The CASE b considers a voltage applied on the electrodes $V_{in}$ comparable or larger than $V_{act}$, and this condition can be obtained by a unique polarization, by placing the charge $Q_{in}$ in ‘one shot’ (CASE S), or by a sum of polarizations obtained by a ‘charge pumping’ process (CASE M). The first approach is described with the operations cycle 0-A-B-C-D-A-B-C-D-A, while the second one can be defined as 0-A-B-C-D-(B-C-D)$_{n}$-A, where $n$ is the number of semi periods of oscillation in which the system is not discharged, while the operations (B-C-D) represents the phases of further charging ($T - T/2$).

The ‘one shot’ polarization (charge in C and discharge in D every DEG cycle, CASE S), in which $V_{in} = V_{act}$, need the use of a very efficient voltage supplier and in order to put very quickly charges inside your transducer.

The second approach (CASE M) can be considered a good way to harvest energy: McKay et. ali demonstrated by the use of a self priming circuit the amplification on the voltage, by gradually boosting its voltage from 10 [V] up to 3.25 [kV], where an electrical breakdown occurred (Fig. 57).

This is highly advantageous also because the efficiency of DEG, and so the power generation, increases monotonically with DEG voltage.
It can be mentioned that in our system a voltage of some hundreds of volts could be sufficient for the applications introduced in the previous chapters. Anyway, reaching higher values could provide the requirements for the exploitation of shape-change-effect, as it will be clear during this sub-section.

Consequently of the fact that the phases $B_n$ ($n>1$) are characterized by a stretching while a charge $Q_{D_{n-1}} = C_{D_{n-1}}V_{D_{n-1}}$ is placed on $L_{1_{(n-1)}}L_{2_{(n-1)}}$, and $V_{D_{n-1}} > V_{act}$, the mechanical behaviour of DEG in $B_n$ is affected by a change in its stiffness.

Because of the geometry of the system, where the two couples of elastomers are bounded in the way that their equilibrium position is independent by the pre-stretching, an applied voltage $V_{in} > V_{act}$ on DEGs in the phase A allows a pure change in stiffness without any equilibrium shift in the direction of stretching (mostly in thickness). As the capacitance read

$$C = \varepsilon_0 \varepsilon_r \frac{L_1 L_2}{X_0} \lambda(t)^2 = \varepsilon_0 \varepsilon_r \frac{L_1 L_2}{X},$$ (65)

defining $b = \frac{\varepsilon_0 \varepsilon_r L_1^3}{vol}$, it gets $C = bL_1^2$, and the stored energy in a DEG can be written so as

$$U_e = \frac{1}{2} CV^2 = \frac{1}{2} \frac{Q^2}{bx^2}$$ (66)

Considering $x$ the direction where the elongation $L_1$ lies, we can define the effective elastic constant of one portion of elastomer, $K_{x1}$, as

$$K_{x1} = \frac{dF(x)}{dx} = \frac{d^2(U_m + U_e)}{dx^2} = K_x^{eff} + \frac{d^2U_e}{dx^2} = K_x^{eff} + \frac{d^2\left(\frac{1}{2} \frac{Q^2}{bx^2}\right)}{dx^2} = K_x^{eff} + \frac{3Q^2}{bx^4}$$ (67)

where $K_x^{eff}$ is the horizontal mechanical effective elastic constant due to the pre-stretching.

As the relation between $K_x^{eff}$ and $K_y^{eff}$ is dependent by purely mechanical parameters, we define the elastic reciprocal function as:

![Figure 57 - Voltage output versus time for a self-priming DEG operating in voltage-boost mode](image_url)
\[ f^* = f(G, A_0, L_0, \lambda_1, \lambda') = \frac{K_y}{K_x} \] (68)

which exclusively depends by materials and configuration design.

The final stiffness induced by both the mechanical and electrical contribution in the \( y \) direction for each elastomer can be written as

\[ K_y(Q, \lambda) = \pi^2 v_n^2 m + \frac{3Q^2}{b x^2} f^* = \pi^2 v_n^2 m + \frac{3Q^2 \varepsilon_0}{\varepsilon_\varepsilon r L_2 L_1^3} f^*. \] (69)

That means that various combinations of the geometrical, mechanical and electrical parameters lead to different working conditions of the system, so characterized by a variable stiffness.

Consequently, the expression of the equilibrium of the mechanical and electrical stresses can be rewritten as

\[ \sigma_m(t, \lambda, V) + \sigma_e(t, \lambda, V) = \sigma_f(t). \] (70)

This implicates that the phases C and D, are characterized by furthers equilibrium positions different from B and A respectively, in which the actuation contribution cannot be neglected, and for which the mechanical stress is strongly affected, showing a change in dimension (thickness and surface areas) and in stiffness.

On one hand, it can be highlight that the maximal deformation will increase proportional to the voltage during the phases (B-C-D), but in the other hand the oscillation frequency can moderately grow because of the increasing of stiffness \( K_y(Q, \lambda) \).

Since the complete case of the system is formed by 4 portions of elastomers for which the treatment has been carried out, the global effective elastic constant of the system in the \( y \) direction reads:

\[ K_{y\text{global}}(Q, \lambda, \lambda', t) = 4\pi^2 v_n^2 m + \frac{12Q^2 \varepsilon_0}{\varepsilon_\varepsilon r L_2 L_1^3} A_0 \lambda' + \frac{12Q^2 \varepsilon_0}{\varepsilon_\varepsilon r L_2 L_1^3} \lambda'^4 \] (71)

\[ = \frac{4\sigma_m(\lambda_1) A_0}{L_1 \lambda_1} + \frac{12Q^2 \varepsilon_0}{\varepsilon_\varepsilon r L_2 L_1^3} \lambda'^4 \]

\[ = \frac{4}{L_1} L_2 z_0 \left( 1 - \frac{1}{\lambda'^2} \right) \left( \bar{G} + 4C_2(\lambda_1^2 + \lambda_1^{-2} - 2) \right. \]

\[ + 6C_2(\lambda_1^{-2} + \lambda_1^{-2} - 2) + \frac{12Q^2 \varepsilon_0}{\varepsilon_\varepsilon r L_2 L_1^3} \lambda'^4 \]

where \( Q = Q(t) \) and \( \lambda_1 = \lambda_1' \) and \( \lambda' = \lambda'(t) \).

In other terms, pumping the charges \( Q \) on the surfaces of the DEGs, a voltage \( V(t) \alpha \left( \frac{\lambda'(t)}{\lambda'^2} \right)^2 \)

will affect the stiffness as
The oscillation frequency will be thus affected by a variable stiffness following the expression

$$K_{y_{global}}(V, \lambda_s, t) = \frac{4}{L_{10}} L_2 Z_0 \left(1 - \frac{1}{\lambda_s^2}\right) \left(G + 4C_2(\lambda_s' - 2) + 6C_3(\lambda_s'' + \lambda_s' - 2)^2 + \frac{12K_0 L_2}{L_{10} Z_0} f' V^2 \right)$$ (72)

The estimation of the electrical contributions is crucial to understand its effect on the behaviour of the device, and it is highlights that the main parameters that can discriminate such effect are the geometries of DEGs $L_1, L_2, Z_0$, the charge $Q$ placed on the electrodes in each cycle and the number of cycles of charging $n$, and the Shear Modulus $G$.

By changing these few parameters several conditions take place on the DEG system: acting on the thickness of the same material, designing a device able to change the stiffness in order to match the characteristic frequency of the aeroelastic system (see Chapter 3) with voltage $<1$ [kV] is possible. Synergistically, the electromagnetic damping of the EMc obtained by changing its external resistance load, and the variable stiffness effect shown by DEGs could lead to the development of an highly versatile and flexible device, while even adopting lower working voltages permits a large increase of its global efficiency. Further studies can identify the spaces of parameters in which these described conditions occur, integrating the model also with the electro-mechanical losses contributions, temporarily neglected.

5.1.3 **Equivalent circuit**

The electrical model of DEG consists in a variable capacitor connected in parallel to a resistor $R_s$, representing the electrical resistance of the dielectric film, and connected in series to a resistor $R_e$, representing the electrical resistance of electrodes and wires, as shown in Fig. 58.

This circuit is presented to give an idea of the electrical conditions in which the system is managed, though it could be used only for the CASE a.

The charge $Q$ exchanged by the system is given by the sum of the time integral of the leakage current and the product of capacitance and voltage of the soft variable capacitor,

$$Q(t) = \int_0^t i_{R_s}(t) \, dt + C(t) V_C(t) .$$ (74)

The generator operates in an electrical circuit achieved by connecting the DEG in parallel to a battery through a diode and to an electrical load, as illustrated in Fig. 58.
The battery supplies the circuit with a difference in the electric potential $V_0(t)$. We assume that the voltage supplied by the battery is zero at the initial time $t = 0$ and then increases linearly during the semi-period $T/2$ of the stretch oscillation ($T/4$ respect the fluttering oscillation) up to the value $V_0$, namely

$$V_0(t) = t \frac{V_0}{(T/2)} \text{ for } 0 < t < T/2.$$

Thereafter, for $t > T/2$, the supplied voltage is kept constant, i.e. $V_0(t) = V_{in}$ for $t > T/2$.

The electrical load is represented by the external resistor $R_{ext}$, while the impedance of the load has to be sufficiently high so that the charge is maintained constant during the release of the elastomer and, as a consequence, the voltage on the dielectric elastomer is increased with respect to the constant value $V_0$ supplied by the battery. A diode must prevent the flowing of the charge from the generator to the battery during the release phase.

Note that a change in $R_{ext}$ can affect the damping of the system; however with this passive components in parallel i.e. resistors, the variable damping is more complex, as it strongly depends on the operations time and frequency.

As it can be deduced, the choice of the electrical components adopted in the electric circuit is very important, because of the importance of the time scale of the process of charging and discharging acting in the Energy Harvester phases of the hybrid system.

A more complicated dedicated electronic circuit has to be designed for the proposes of CASE M: an idea of such circuit can be provided by a similar application reported in [81].

A draft of the expected results of the operations described in the CASE M of DEGs management is reported in Figure 59: a charge pumping process ensures the increasing of the voltage on the DEG during several cycles until a discharge at higher voltage occurs. The zoom reports an experimental measurement conducted on FLEHAP device, demonstrating the change in voltage on the electrodes of the elastomeric capacitors due to the change in capacitance during a cycling stretch imposed by the wind at low flow speed.
5.2 Elastomeric dielectric composites

The experimental activities related to the manufacture and characterization of a new type of dielectric elastomer materials have been carried out partially in DCCI of University of Genova, partially in IMP of INSA de Lyon and partially in G2Elab of INPG de Grenoble.

Silicone elastomers are one of the most studied materials for DEGs due to their high efficiency, reliability and fast response times. The major advantage of silicone elastomers over other materials is that they produce repeatable, reproducible actuation upon activation, they show little tendency for Mullins (stress softening) and ageing effects, and they have been shown to actuate (at low strains) to more than 400 million cycles, without failure. They also possess significantly lower viscous losses than acrylics, indicating that they can be operated at higher frequencies with lower losses and less heat generation. Furthermore, silicones can operate on a broader temperature range and possess inherent softness and stability. The disadvantage of silicone elastomers is that they show relatively low dielectric permittivity, compared to other matrixes such as poly-urethanes and acrylics. A common strategy to increase the dielectric permittivity of DEG is to add a high permittivity filler into the matrix, although in most cases it leads to increase of the stiffness of the composite material: furthermore, a too high dielectric permittivity contribution of fillers normally worsens other properties such as lower Electrical Breakdown Field $E_b$, electrical losses $\varepsilon'$ and, in general, the stability of the DEG in both thermal and life time points of view.

In order to find a reasonable trade-off through the complicated, and often inter-dependent main properties of this kind of materials, referring to the advantages mentioned, silicone elastomers have been chosen as matrix, and a functional filler has been individuated and added varying its amount in weight. The aims were obtaining soft, stable and high lifetime composites.
5.2.1 **Film realization**

The realization of the materials was carried out in the laboratories of Ingénierie des Matériaux Polymères (IMP) of INSA de Lyon.

5.2.1.1 **Materials**

The use of commercial silicone-based products is experimentally easier, because of better reproducibility in short times to investigate the composites materials from the mechanical to the electrical point of view.

5.2.1.1.1 **Elastomer matrix**

The choice of the two silicone matrixes has been done mainly through their elastic properties, because, as reported in literature, all silicones have got similar dielectric relative permittivity, between 2 and 3\(^79\).

Sylgard 186 was acquired from Dow Corning. It consists in a two components room temperature vulcanizing (RTV) silicone rubber: part A is the pre-polymer; part B is the curing agent. The elastomers obtained using a mix ratio of 10:1 (part A:part B), exhibit a hardness of about 24 shore A after 2 hours at 25 [°C].

Silbione 4310 PEX was purchased from Bluestar Silicones. It is a bi-component healthcare grade liquid silicone rubber (LSR); the two parts (part A with the catalyst and part B without catalyst) were mixed in a equal ratio (curable in 15 minutes at 175 [°C] in a oven) providing an elastomer with a hardness of about 10 shore A.

Both materials contain fumed silica, are cross-linked by a hydrosilylation reactions catalysed by highly active platinum catalysts (silicone-soluble Karstedt's catalyst), and show medium-high viscosity when mixed (around 10\(^5\) [cP]).

LSRs are a family of tough and versatile silicones, enabling fast-cured, high-precision injection moulding for high-performance parts such as transducer devices\(^88\). They can be moulded into very intricate and complicated geometries, remaining elastic without tearing\(^89\). Since they are cured by addition reaction, the formulations enable a high degree of control over the network topology – and thus on the mechanical properties – of the final material\(^90\).

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<td>Sylgard 186</td>
<td>24</td>
<td>0.9</td>
<td>250</td>
<td>2.7 (\text{@}100) [Hz], (2.68 \text{@} 1) [kHz](^*)</td>
<td>1:10</td>
<td>(15min@150(^\circ))</td>
</tr>
<tr>
<td>Bluestar</td>
<td>Silbione LSR 4310</td>
<td>10</td>
<td>0.55</td>
<td>1000</td>
<td>Not reported</td>
<td>1:1</td>
<td>5min@185(^\circ)</td>
</tr>
</tbody>
</table>

Table 5 - Main properties provided by the supplier of commercial silicone matrixes chosen. *Dielectric Spectroscopy.
LSRs may be clear or translucent and can be manufactured to have unique properties such as solvent resistance, increased thermal stability or low outgassing. Furthermore, they can incorporate additives and unique fillers, such as pigments and active pharmaceutical ingredients, with minimal changes to the key characteristics of the cured elastomer, i.e. toughness and elasticity. In most cases LSRs use a platinum catalyzed addition cure system and require a curing temperature above 80 °C.\(^1\)

They typically comprise ~ 75 wt.% linear silicone polymers and ~ 23 wt.% fumed silica, with the remainder consisting of a combination of curing additives such as hydride cross-linkers and inhibitors.\(^2\) Fumed silica, also known as pyrogenic silica because it is produced in a flame, consists of microscopic droplets of amorphous silica fused into branched, chainlike, three-dimensional secondary particles which then agglomerate into tertiary particles (see Fig 60). The resulting powder has an extremely low bulk density and high surface area. Its three-dimensional structure results in viscosity-increasing, thixotropic behaviour when used as a thickener or reinforcing filler.

The majority of commercially available LSRs are formulated as two-part systems: 1:1 mix ratio silicones with viscosities ranging from 50,000 to >1,000,000 [cP]. Part A contains a platinum catalyst and Part B contains a cross-linker and an inhibitor. Fumed silica improves the ultimate mechanical properties of the silicone and has the ability to interact non-covalently with the polymer (see Fig. 61), thereby allowing stress relief when a shear is applied in an uncured or a cured state. Moreover, fumed silica also increases the tear growth resistance strongly.

Figure 60 - Preparation of aggregated fumed silica from flame pyrolysis of silicon tetrachloride vaporized in a 3000[°C] electric arc, \(^3\), and its relative organic surface modification.
Semi-volatile inhibitors control the reaction rate, in order to provide an acceptable liquid elastomer pot life until heat is applied. The combination of medium-high viscosity polymers and silica gives uncured LSRs a creamy consistency similar to petroleum jelly. Processing LSRs successfully depends on using equipment designed to mix and pump thick materials without introducing air. For thin films this requirement becomes even more important.

5.2.1.1.2 **Nanocomposites**

Nanocomposites are a fairly new class of composite materials where a filler having at least one dimension in the nanometer ($10^{-9}$ [m]) range is dispersed in a continuous matrix. They got recognition after the first successful development of Nylon nanocomposites having enhanced mechanical properties by the Toyota researchers. Since then, nanocomposites have been major area of research. The nanocomposites have shown improved mechanical and thermal properties, barrier properties and decreased flammability with respect both micro and macro composite materials. The filler shape, aspect ratio and their affinity towards matrix material are among distinctive parameters that play a vital role in modifying the properties. The nanocomposites could be prepared by different methods including in situ polymerization, melt intercalation and direct mixing. The dispersed filler can be of the shape of sphere, tube, fibre or lamellae. The correlation of properties of materials with filler size has gained a great deal of importance with the
recent advancement in the field of nanotechnology. Owing to their exceptional potential to exhibit peculiar characteristics that could not have been achieved with their traditional micro-scale counter parts, they have attracted a great deal of interest to exhibit enhanced and novel properties.

### 5.2.1.1.3 Nanoclay

Nanoclays are rigid fillers, and generally the improvement in the moduli of matrix material with the incorporation of these nanoclays is attributed to their high stiffness. Polymeric matrices are soft materials and their reinforcement with rigid nanoclays impede the free movement of polymer chains neighbouring to the filler and if the interfacial adhesion between the filler and the chains is satisfactory, the filler behaves as load bearing constituent.

Improvement in the mechanical properties of polymer matrices with the addition of nanoclays have been published in many reviews. Beside improvement in the mechanical properties of polymers, nanoclays also have potential to reduce the permeability of gases, like oxygen, carbon dioxide, and many organic compounds. Uniform dispersion of rigid impermeable nanoclays impede the diffusion of permeating molecules by forcing them to follow a tortuous path. Increased flammability resistance and enhanced thermal stability of nanocomposites are among the various other benefits attributed to the nanoclays. Nanoclays has been also explored as filler to avoid an high charge paths across the DEG (electrical failure) and a possible damage due to cycle deformations (mechanical failure).

Montmorillonite (MMT), showed in Fig. 62, is a low cost hydrated alumina-silicate clay that consists in multiple planar sheets (tethraedral-octaedral-tethraedral) of about 1 [nm] of thickness and 100-200 [nm] of width.

![Crystal structure of Montmorillonite layered silicate](https://example.com/crystal-structure.png)

**Figure 62** - Crystal structure of Montmorillonite layered silicate; silicon atom located in the centre of each tetrahedron is not illustrated in the figure for simplicity. Thickness of fillosilicate about 1 [nm].
These sheets are normally packed in micro agglomerates by the presence of anions and cations between them (few [nm]). They can be exploited as reinforcement fillers, because their segregation also allows the adhesion of portions of the matrix, increasing in some case the mechanical properties.

Alkyl-ammonium cations in the organo-silicates, originally proposed in 90ies from Toyota Research, lower the surface energy of the inorganic host and improve the wetting characteristics of the polymer matrix, and result in a larger interlayer spacing; this can also lead to a good rate of reaction sites, as well charge traps, and so an interesting composite material with mechanical and dielectrical properties can be obtained. These organo-surface modified benthonites are called OMMT. The organic functionalization also prevents the water absorption of material. Summarizing the main effects that this filler could imparts to the silicone composite material, we can report: high interfacial polarization and high space charge (improve dielectric permittivity \( \varepsilon_r \)), thermal resistance/flame retardant properties (increase \( E_b \)), and, if exfoliated with a further use of additives such as plasticizers, a good trade-off in terms of the global stiffness.

A schematic illustration of a polymerization with OMMT is reported in Fig.63, where different conditions of dispersion by presence of entanglements at different filler loadings are illustrated.

The commercial product chosen as filler was Cloisite 20A from the supplier BYK Additives. It is an organo-modified nanoclay (montmorillonite) with a dimethyl, dehydrogenated tallow, quaternary ammonium salt surface-modifier (95 meq/100g of clay). Such surface modification permits to improve the compatibility of the nanoclay with the silicone chains. The main properties of Cloisite 20A are summarized in Table 6.

![Figure 63 - Schematic illustrations of the formation process of PE-OMMT nanocomposites during in situ polymerization in the presence of OMMTs with different concentration. From 114,115](image-url)
5.2.1.2 **Methods and Protocols**

The pristine silicone samples (RTV rubber and LSR, without filler) were realized with a protocol different from what was used for nanocomposites. After correctly weighting the two parts of silicones (A+B about 10 [g]), they were mixed thanks to a Hauschild Speed Mixer 2750 with the procedure specifications reported in Tab. 7.

<table>
<thead>
<tr>
<th>Steps [#]</th>
<th>Time [s]</th>
<th>Speed [rpm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>30</td>
<td>800</td>
</tr>
<tr>
<td>2</td>
<td>30</td>
<td>1200</td>
</tr>
<tr>
<td>3</td>
<td>60</td>
<td>2000</td>
</tr>
<tr>
<td>4</td>
<td>120</td>
<td>2750</td>
</tr>
</tbody>
</table>

Table 7 - Procedure steps of speed mixing process.

The mixture was then placed on a 130x130 [mm²] Pyrex substrate covered by a textured Teflon coating, having two additional 150 [μm] stripes at its lateral sides. Such substrate was positioned on an auto bar coater applicator. The bar of the instrument consisted in a cylinder of 10 [mm] of diameter with 16 [μm] spiral wire, the weight on the moving bar was 100 [g], and the bar moving speed was set to 50 [mm/min]. The obtained films were then placed under a vacuum chamber for 10+10 [min] at 0.7 [bar] in order to remove the gas bubbles inside the material. Next, the films were cured in an oven following the specifications reported in Table 5.

In the case of filled silicones, a pre-dispersion of the fillers in the base of the elastomer (pre-polymers) by a roll to roll milling process was led (model EXACT 80E). The base and the filler were placed between the first two cylinders (c1-c2) for a certain number of times, while the distances of each cylinders to each other were varied, as well as the turning speed of the third cylinder, for each of the three steps reported in the Table 8. After the roll mill process, the procedure adopted was the same used for to the unfilled samples. For each silicone matrix the amount of filler was varied to 0, 2, 4, 6, 8 and 10%.

<table>
<thead>
<tr>
<th>Distance between cylinders [μm]</th>
<th>Speed [rpm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Step 1 5 c</td>
<td>c1-c2</td>
</tr>
<tr>
<td>1</td>
<td>30</td>
</tr>
<tr>
<td>2</td>
<td>20</td>
</tr>
<tr>
<td>3</td>
<td>15</td>
</tr>
</tbody>
</table>

Table 8 - Procedure for the roll milling process.

---

Table 6 - Main properties of Cloisite 20A.

<table>
<thead>
<tr>
<th>Moisture [%]</th>
<th>Typical dry particle size, d_{50} [μm]</th>
<th>Packed bulk density [g/l]</th>
<th>Density [g/cm³]</th>
<th>d_{40} X Ray results [nm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>&lt;3</td>
<td>&lt;10</td>
<td>175</td>
<td>1.77</td>
<td>3.16</td>
</tr>
</tbody>
</table>

---
5.2.1.3 **Results**

The composites obtained by the use of the described procedure, didn’t catalyze in the cases of an amount of OMMT >4%wt, as reported in Table 9.

This was attributed to the effect of the Pt catalyst contained in the cure agent of silicones on the ammonium salt of the organic functionalization of MMT; in fact, the amines played as competitors in the catalyzed reaction, so that the catalytic reaction was partially inhibited. In order to obtaining materials with higher loading of OMMT, Dicumyl peroxide, acquired from Sigma Aldrich (DP), was adopted with an amount in weight of 1% as cross-link forcing agent in Sylgard 186. Samples with 0%, 4% and 10% in weight of OMMT were realized at 180[°C] using a hydraulic press at 70 [bar] for 40 min. A de-vacuum process was also adopted in order to extracting the un-reacted peroxide.

Such samples, obtained by using a metal mask frame of 400 [μm], were well catalyzed also in the case of the largest amount of OMMT (SYDP10). A percentage in weight, and not a molar weight, was used for DP, because the precise composition of Sylgard 186 Base was unknown. Probably, a smaller amount of DP was sufficient; nevertheless, a complete cross-link process, in this way, was ensured.

**Table 9** - Nomenclature of the samples realized: barred samples represent the not cross-linked ones.

<table>
<thead>
<tr>
<th>RTV Sylgard 186 (1:10)</th>
<th>OMMT [%wt]</th>
<th>LSR Silbione 4310 (1:1)</th>
<th>OMMT [%wt]</th>
<th>RTV Sylgard 186 + 1%wt peroxide</th>
<th>OMMT [%wt]</th>
</tr>
</thead>
<tbody>
<tr>
<td>SY0</td>
<td>0</td>
<td>B0</td>
<td>0</td>
<td>SYDP0</td>
<td>0</td>
</tr>
<tr>
<td>SY2MT</td>
<td>2</td>
<td>B2MT</td>
<td></td>
<td>SYDP4</td>
<td>4</td>
</tr>
<tr>
<td>SY4MT</td>
<td>4</td>
<td>B4MT</td>
<td>4</td>
<td>SYDP4</td>
<td>4</td>
</tr>
<tr>
<td>SY6MT</td>
<td>6</td>
<td>B6MT</td>
<td></td>
<td>SYDP10</td>
<td>10</td>
</tr>
<tr>
<td>SY8MT</td>
<td>8</td>
<td>B8MT</td>
<td></td>
<td>SYDP4</td>
<td>4</td>
</tr>
<tr>
<td>SY10MT</td>
<td>10</td>
<td>B10MT</td>
<td>10</td>
<td>SYDP10</td>
<td>10</td>
</tr>
</tbody>
</table>

5.2.2 **Mechanical characterization**

The mechanical characterization of the materials previously prepared was conducted in the LAMCOS Laboratories of INSA de Lyon. The studies, when possible, have been led in the operative conditions of FLEHAP when it is working in wind and it is extracting electrical energy by the EMc: the relative deformations λ do not exceed 1.5, so the range 1 ≤ λ < 2 was considered in the treatment.
5.2.2.1 Introduction to the elastic characterization

Different methods for estimating Young’s modulus are commonly used: Dynamic Mechanical Analysis (DMA), Rheometric Dynamic Analysis (RDA) or Moving Die Rheometry (MDR). Common data outputs from both are in the form of storage ($G'$) and loss ($G''$) moduli. The complex storage modulus ($G*$) is defined as $G*^2 = G'^2 + G''^2$. Young’s modulus ($E$) is approximated as $E = 2G * (1 + v)$, where $v$ is Poisson’s Ratio. For silicones, Poisson’s ratio is commonly taken between 0.48 and 0.495\(^7\).

Testing of Young’s modulus is most commonly done by generating a stress/strain curve in tension. Young’s modulus is defined as the initial slope of the stress/strain response. However, measuring the slope is not as easy, if not even quite arbitrary, while taking a tensile strength at a given elongation cannot provide a comparable parameter for all kind of elastomers. Tensile strength divided by elongation is a “Secant” modulus. Such a Secant modulus = Young’s modulus only when the stress/strain response is linear. For products that have such a very linear response over a long range of strain, the simplest reasonable estimate of Young’s modulus is the tensile strength at 100% elongation. However, most products do not have such a linear stress/strain relationship. It is common to see an initial steeper slope, followed by a much lower slope that eventually steepens again as the material approaches failure/breakage.

Very often the elastic properties of rubbers are simplistically reduced to scales referred to fast characterization methods adopted in industries, such as durometers obtained by measuring the depth of an indentation in the material created by a given force on a standardized presser foot. This depth is dependent on the hardness of the material, its viscoelastic properties, the shape of the presser foot, and the duration of the test. The reader is referred to ASTM D2240 a complete specification of the standards.

![Figure 64 - Representation of equivalent Young's Modulus with respect the ASTM Shore scales, from 118.](image)
Briefly, for measuring Shore A, a foot indents the material while for the Shore D foot penetrates the surface of the material. The most widely known correlation of durometer values to Young’s modulus was put forth in 1958 by A. N. Gent:

\[
E = \frac{0.0981 \times (56 + 7.62336 \times S_A)}{0.137505 \times (254 - 2.54 \times S_A)}
\] (75)

where the Young’s modulus \(E\) is expressed in [MPa] and \(S\) is the ASTM D2240 Type A durometer hardness. This equation is considered a good first-order approximation of Young’s modulus from A hardness of 80 down to 20, though some have considered it of less value below a hardness of 40A.

Other equations have also been postulated by Ruess for the conversions of Shore A and Shore-D hardness to Young’s Modulus expressed in [MPa], relatively:

\[
E = e^{0.0235 S_A - 0.6403}
\] (76)

\[
E = e^{(0.0235 (50+S_D) - 0.6403)}
\] (77)

Comparing the Secant and RDA data to the Gent and Ruess equations shows reasonably good correlation, with the Ruess equation perhaps fitting somewhat better. For the family of LSRs the Gent’s equation appears to model the data quite well.

The D durometer hardness scale typically aligns with silicone products with \(80 S_A = 20 S_D\). There was considerable scatter in the data when the durometer values moved into the 00 scale, where a 65 \(S_{00}\) usually about 8 \(S_A\). Looking at just the stress/strain data, a reasonable fit can be made with an exponential curve, letting find a good fit for low stiffness materials with low viscous losses.

Besides the obviously large scatter in the data reported in Fig. 64, especially at low hardness values, other factors also influence how a material performs in application induced extensions (and compressions).

- Cyclic stress gives way to the Mullins effect, where stress/strain responses are impacted by prior maximum loading stress. Such changes are often recoverable at low strains, but can become permanent at higher strains.

- Material fatigue over time and/or cycling count can induce defects such as tears or cracks that dramatically reduce the stress required to cause failure.

- Exposures to high temperatures, and especially when coupled with compressive pressure can often lead to further cross-linking which can increase modulus and hardness values. This is commonly observed in compression set testing. Such changes can be thought of as further curing at milder temperatures, or they can be a result of oxidative or other degradation mechanisms at higher temperatures.
• Exposure to soluble liquids and oils can cause plasticization, which lowers modulus and hardness. Such changes can be temporary or permanent, depending on the volatility of the contaminant.

• Stress/strain characteristics can be profoundly affected by temperature. Most silicones undergo a slight crystallization around -48[°C]. Below that transition temperature, these elastomers will be significantly harder and higher modulus. Some silicones contain semi-crystalline silicone resins which can exhibit a melting or softening point. Durometer and modulus characteristics can significantly change when the temperature crosses such transitions.

• Modulus can be impacted by cyclic strain frequency. DMA and RDA are commonly used to characterize property responses over temperature and frequency ranges.

Furthermore, the properties of commercial products are designed to meet standards usually called out in technical data sheets and sales specifications. Modulus and hardness can be unwittingly or purposely modified in several ways. For two-part products, the specified mix ratio will create a mixture that meets the intended property profile. Products are formulated to allow for expected mix tolerances from dispense equipment, which typically should be ±3% from reputable vendors. Many products can tolerate ±5% in the mix ratio without significant impact on cured properties. Going beyond that tolerance can shift modulus, durometer and other properties outside of the product specifications and warranties. In general, using less cross-linker will make cured products softer and lower modulus, while adding more has minimal impact (note, for the extremely soft gels, adding more cross-linker can significantly increase hardness and modulus). As mentioned in the Limitations, exposure to soluble liquids and oils will essentially plasticize silicones, lowering hardness and modulus. Some formulators will purposely add silicone or organic oils to achieve the same purpose. However, such practices can lead to unintentional consequences, such as oil migration out of the cured silicone (called bleed) which can cause visual blemishes and negatively impact adhesion of adhesives, sealants, paints and inks on nearby surfaces. Over time, such modified silicones may appear to harden and stiffen as the plasticizing oil migrates away. It is generally not recommended to use this approach to modify properties unless there is a very clear understanding of the long term durability effects on the silicone itself and on surrounding surfaces. Note that some silicones are originally formulated with small amounts of such oils – these are commonly called out as “self-lubricating” or other terminology to indicate their presence. Two-part room temperature curing condensation cure silicones also commonly include a small amount of non-curing silicone polymer used as a diluent for the cure catalyst. Modulus and hardness modifications can be achieved as above, but these can be prone to poor durability of the desired properties as well as other unintended consequences. Other methods for reducing the cross-link density in a more stable manner can create products with desired hardness and modulus characteristics.
5.2.2.2 Model

The mechanical model is based on two extensional approaches: the Uniaxial extension (Fig. 65a), describing a theoretical homogeneous deformation, and the shear case (Fig. 65b), in which the width $L_{2o}$ is considered constant during a longitudinal deformation. Because of the clamping condition of the rectangular samples by the jaws during the tests, a further case is presented in (Fig. 65c); however this effect can be considered negligible for small stretching ($\lambda < 1.5$) and high aspect ratio.

Referring to the shear case of Figure 65b, a planar extension of the material causes the relative deformations

$$L_{1o} \rightarrow L_1 = L_{1o} \lambda \quad ; \quad L_{2o} \rightarrow L_2 = L_2 \lambda \quad ; \quad z_0 \rightarrow z = z_0 / \lambda$$

The deformation tensor of the considered model is then written as

$$T = \begin{bmatrix} \lambda & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & \lambda^{-1} \end{bmatrix} \quad \text{(78)}$$

while

$$\lambda x 1 x \lambda^{-1} = 1 \quad \text{(79)}$$

The strain invariants, expressed in terms of the stretch ratios $\lambda = \frac{L_1}{L_{1o}}$ are

$$I_1 = T_1^2 + T_2^2 + T_3^2 = \lambda^2 + 1 + \lambda^{-2} \quad \text{(80)}$$

$$I_2 = T_1^2 T_2^2 + T_2^2 T_3^2 + T_3^2 T_1^2 = \lambda^2 + \lambda^{-2} + 1 = I_1 \quad \text{(81)}$$

$$I_3 = T_1 T_2 T_3 = \lambda^2 x 1 x \lambda^{-2} = 1 \quad \text{(82)}$$

In the case of Uniaxial deformation (Fig. 65a), adopted on the dog bone samples

$$T = \begin{bmatrix} \lambda & 0 & 0 \\ 0 & \lambda^{-1/2} & 0 \\ 0 & 0 & \lambda^{-1/2} \end{bmatrix} \quad \text{(83)}$$

while

$$\lambda x \lambda^{-1/2} x \lambda^{-1/2} = 1 \quad \text{(84)}$$

So, its first strain invariant reads

$$I_1 = T_1^2 + T_2^2 + T_3^2 = \lambda^2 + 2 \lambda^{-1} \quad \text{(85)}$$

Figure 65 - Uniaxial deformation (a), Shear extension (b) and nonlinear deformation (c) approaches.
The Mechanical Strain Energy described by the 3 parameters constitutive polynomial equation of Yeoh (from Rivlin model) is chosen to fit the experimental curves:

\[
W = \sum_{i=1}^{3} C_i (I_1 - 3)^i = C_1 (I_1 - 3) + C_2 (I_1 - 3)^2 + C_3 (I_1 - 3)^3
\]

The true stress is expressed as

\[
\sigma_m = 2\left(\lambda - \frac{1}{\lambda^2}\right) \left(\frac{\delta W}{\delta I_1} + \frac{1}{\lambda I_2} \frac{\delta W}{\delta I_2}\right) = \sum_{i=1}^{3} 2 i C_i \left(\lambda - \frac{1}{\lambda^2}\right) (I_1 - 3)^{i-1} =
\]

\[
\sigma_m = \left(\lambda - \frac{1}{\lambda^2}\right) (2C_1 + 4C_2 (I_1 - 3) + 6C_3 (I_1 - 3)^2)
\]

where \(2C_1\) denotes the shear modulus \(G\) of material.

The relaxation test is the second main interesting measurement conducted: this viscoelastic test is obtained by measuring the decreasing of the stress applied on a sample when a fixed deformation is imposed. The Generalized Maxwell model is the most general form of the linear model for viscoelasticity. It takes into account that the relaxation does not occur at a single time, but at a distribution of times: this is due to the distribution of the molecular segments lengths and to the density of the cross links in the volume of the silicone material in object.

The expression of the time dependent mechanical stress reads

\[
\sigma_m(t) = \sigma_{\infty} + \sum_{i=1}^{N=3} \sigma_i e^{\left(-\frac{t}{\tau_i}\right)}
\]

where \(\sigma_{\infty}\) is the equilibrium stress, \(\sigma_i\) are the characteristic stresses, \(\tau_i\) the characteristic times and \(N\) is the number of considered relaxation steps. After fitting the curves, it is necessary to individuate the characteristic relaxation parameters of the material, following a quasi-linear viscoelasticity model.

\[
\sum_{i=1}^{N} \frac{\sigma_i}{\sigma_{\infty}} e^{\left(-\frac{t}{\tau_i}\right)} = \sum_{k=1}^{N} \frac{g_k}{1 - \sum_{k=1}^{N} g_k} e^{\left(-\frac{t}{\tau_k}\right)}
\]

### Methods and Protocols

NF T 51-034 Standard, derived from Iso 527-1 / 37-2, was used for sizing the test samples used for the characterization. Type H3 was, while the thickness (E in Fig. 66) was 10 times smaller, between 0.15 and 0.3 [mm], depending of the materials.

A specific protocol was used for all samples, and 3 to 5 sample of the same were tested for each measurements. Before testing, the dimensions of each sample were measured through a digital calliper (±0.01 [mm]). Then, the end parts of the samples were sandwiched between two insulating tapes and fixed at the two jaws of the tensile machine, equipped by a 10 [N] force sensor (±0.1 [mN]). First, an elongation to rupture at 20 [mm/min] was carried out, in order to
provide information about stress-strain curves. Next, 3 cycles of deformation up to 30 [mm] ($\lambda \sim 2$) and elongation to rupture at 20 [mm/min], allowed to evaluating the mechanical hysteresis between loading and unloading. Finally, 3 cycles to 30 [mm] at 20 [mm/min] and 60 [min] hold to 30 [mm] were led for evaluating the relaxation times of the samples.

Figure 66 - Dimension of test sample, derived from NF T 51-034; on the bottom ‘type H3’ shape utilized.

### 5.2.2.4 Results

The first results concern the estimation of the coefficients able to describe the mechanical behaviour of the elastomers during a longitudinal deformation: these were obtained by fitting the curves corresponding to the last cycle of deformation of all samples with Yeoh polynomial equation previously shown, by the use of Matlab®.

The results were quite reproducible, with deviations from the reported average values around 5-10%. Over a slight dispersion of the force curves measured, the higher indetermination was related to the estimation of the initial section of the samples $A_0 = L_2 z_0$. In fact, the measurement of thickness of these very soft membranes has proved to be sophisticated.

The case of Sylgard 186 cured with 1%wt Dicumyl Peroxide showed the typical effect of adding fillers, for which the stiffness of the composites appeared increased (Fig. 67c).

On the contrary, all other materials showed a reduction of the shear modulus increasing the content of OMMT. The latter were well described by an hyper-elastic model, where the predominant term, linked to $G$, was the first coefficient $C_1$ (see Eq. 87).
Figure 67 - Stress/Strain curves of the last cycle of deformation of a) Sylgard 186 (named SY), b) Silbione 4310 (named B), and of SY cured by Dicumyl Peroxide (named SYDP).

Figure 68 - Mean values of percentage elongation to rupture.

Regarding the elongation to rupture, Silbione 4310 (B in Fig. 68 shows very impressive effect due to OMMT (>1000%), while in the other cases there are no relevant changes. This property is not interesting for the DEG application in FLEHAP, where the relative deformation shall not exceed $\lambda = 1.5$, but provides an idea of their integrity.
Figure 69 reports the cyclic deformation tests at 20 mm/min for the LSR samples, revealing that no important viscous or plastic effects occurred. Surprisingly, the samples loaded with OMMT shown smaller or similar losses with respect the pristine material. Similar results have been obtained for the RTV samples (see Fig. 70). Moreover, from the curves reported in Fig. 70, the hyper-elastic properties were confirmed: in fact, the global loss of stress measured for both materials (RTV and LSR OMMT loaded) was evaluated less than 5%. This parameter, named $\sigma_{\text{loss}}$, is defined as the stress percentage losses between the loading and the unloading of the samples. The cyclic tests conducted revealed that there are no important viscous or plastic effects on the materials and no dependence by the frequency of deformation, confirming the advantage of silicones previously cited as a fast response material. Thus, adding OMMT seems to not impart significant viscoelastic properties: the samples of SY and B with fillers need less force to be stretched at the same ratio, while the viscous losses appear reduced with respect the unfilled ones (Fig. 71).
Furthermore, as it can be observed from Fig. 71 the speed deformation (from 20 to 1000 mm/min) didn’t affect the response of the materials (SY). Similar results were observed also for the other materials.

As clearly shown in Fig. 72, SY0 exhibited similar values of $G$ reported by the supplier and other studies in literature $^{79,82}$ ($G=0.3$ MPa).

Relaxation time tests showed that the first relaxation step ($t_1, G_1$ in Table 10) is normally very fast for these materials. The related curves are reported in Fig. 73.

Figure 71 - Example of cycle test on SY double dog bone sample: 3 cycles to $\lambda = 2$ at 100, 200 [mm/min], 5 cycles at 300, 600, 1000 [mm/min] and 2 cycles at 1600 [mm/min].

Figure 72 - Mean values of the 3 Yeoh parameters (see Eq. (93)) obtained by fitting the experimental curves ($R>.995$).
Referring to Table 10, excepted the first sample SY0, for which the deformation imposed was \( \lambda = 1.3 \) and 2, the rest of samples have been deformed at \( \lambda = 1.3 \), typical value of the maximal pre-stretches of the device, where global loss in stress decrease less than 2% (value when the stabilization occurs). Only the highly filled Silbione10 (B4MT) shows an increase of \( t_1 \), while \( t_2 \) and \( t_3 \) remain similar with respect to the pristine matrix. In order to obtain the parameters able to describe the material without the dependence of the geometries and the measurement conditions (i.e. the maximal deformation imposed), a quasi-linear viscoelastic model (Eq. 89) was adopted to fit the results achieved (Tab. 10). Analyzing the results of the measurements performed at different stretching values (SY0 measured at \( \lambda = 2 \) and 1.3), it was concluded that a quasi-linear viscoelastic model cannot be formally applied, because of different behaviour showed. This was also attributed to the nature of the sample, characterized by thin films. Anyway, the global losses in stress were of less than 2%.

### Table 10

<table>
<thead>
<tr>
<th>( \sigma_{\text{rela}} )</th>
<th>( G_{\text{inf}} )</th>
<th>( G_1 )</th>
<th>( t_1 )</th>
<th>( G_2 )</th>
<th>( t_2 )</th>
<th>( G_3 )</th>
<th>( t_3 )</th>
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</thead>
<tbody>
<tr>
<td>SY0</td>
<td>SY2MT</td>
<td>SY4MT</td>
<td>B0</td>
<td>B2MT</td>
<td>B4MT</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.085</td>
<td>0.087</td>
<td>0.034</td>
<td>0.087</td>
<td>0.106</td>
<td>0.101</td>
<td></td>
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<tr>
<td>0.937</td>
<td>0.2884</td>
<td>0.2357</td>
<td>0.2522</td>
<td>0.1688</td>
<td>0.1589</td>
<td>0.2275</td>
<td>0.226</td>
</tr>
<tr>
<td>0.0085</td>
<td>0.0054</td>
<td>0.0001</td>
<td>0.0062</td>
<td>0.0142</td>
<td>0.0048</td>
<td>0.0002</td>
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</tr>
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<td>2</td>
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<td>0.0064</td>
<td>1</td>
<td>4.773</td>
<td>0.0029</td>
<td>0.0064</td>
</tr>
<tr>
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<td>0.0055</td>
<td>0.0049</td>
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<td>59.89</td>
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<td>0.0087</td>
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<td>894.1</td>
<td>864.4</td>
<td>842.2</td>
<td>277.9</td>
<td>522.1</td>
</tr>
</tbody>
</table>

Table 10 - In first row, percentage in losses in stress during a cyclic deformation (20 [mm/min]) at \( \lambda = 1.3 \). In the other rows: hyper-elastic Prony series relaxation parameters obtained by fitting the relaxation curves.
The viscoelastic contribution during the cycle deformations of the device is very low at room temperature and, in general, out of extreme conditions, for which further experiments will be conducted. From the mechanical analysis our systems seemed to belong to bimodal network features, composed by a chemical network and a physical network. The formation of the chemical (primary) network, formed by long and short chain fractions and low molecular mass molecules, was partially inhibited by the surface modifier of the fillers (quaternary ammonium salts). On the other hand, a secondary network composed by the filler loading (fumed silica and nanoclay) consisted in a physical network which provided stability to the primary one. The result of such bimodal network could explain the improvement observed on the viscoelastic properties.

5.2.3 **Rheological characterization**

Rheology is a branch of Physics which describe the deformation and flow behaviour of any kind of material. In fact, the term originates from the Greek word “rhei”, meaning “to flow”.

For any type of filler in an elastomeric matrix the first mechanism involved in reinforcement is hydrodynamic, i.e. the introduction of rigid particles creates obstacles to the materials flow, whatever the polymer–filler and filler–filler interactions.

In order to evaluate the contribution of the filler in the silicone matrix on the mechanical properties due to its secondary network, a rheological characterization was carried out in the Polymer Materials Laboratory of DCCI of UniGe.

5.2.3.1 **Theory**

Linear rheology gives insight into the structure-properties relationship and the state of dispersion. In the linear viscoelastic region, the observed moduli are not a function of the deformation amplitude. Therefore, the sample must be excited at very low values of deformation amplitude, which implies low torque values that are difficult to be measured. If the aforementioned condition is fulfilled, the molecular behaviour can be observed without significant modification of the relative position of the structure from the equilibrium. All filled materials exhibit non-linear behaviour after a certain amount of strain that is inverse to the filler level. The higher the filler level, the shorter the linear viscoelastic domain becomes. It has also been shown that the incorporation of particles will systematically increase the storage modulus ($G'$) and the loss modulus ($G''$) of the composite. Adding a more rigid material would definitely increase the response of $G'$, which reflects the elasticity of the material; while, $G''$ is increased due to the more pronounced dissipation processes that occur at higher filler level e.g., more pronounced friction between particles.
During an amplitude sweep the amplitude of the deformation, or alternatively the amplitude of the shear stress, is varied while the frequency is kept constant. The amplitude is the maximum of the oscillatory motion.

At low deformation $G'$ and $G''$ are constant; the sample structure is undisturbed. This region is called linear-viscoelastic (LVE). As soon as the moduli start to decrease, the structure is disturbed, until the end of the LVE-region is reached: the plateau value of $G'$ in the LVE-region describes the rigidity of the sample at rest; the plateau value $G''$ is a measure for the viscosity of the unsheared sample. The ratio of the two moduli gives information about the characteristic of the sample: if the storage modulus is larger than the loss modulus (see Fig. 74), the sample behaves more like a viscoelastic solid. In the opposite case in which $G'' > G'$ in the LVE region, the sample has the properties of a viscoelastic fluid. As larger the difference between the moduli as more the samples show the properties of a pure fluid and solid, respectively. With the amplitude sweep also the yield point can be determined. Therefore two special points can be used: the end of the LVE-region and the intersection of the curves for $G'$ and $G''$. In most cases the intersection of $G'$ and $G''$ is of more practical importance, and it is called crossover point.

During the frequency sweep the frequency is varied while the amplitude of the deformation - or alternatively the amplitude of the shear stress - is kept constant. For the analysis the storage and loss modulus are plotted against the frequency. The data at low frequencies describe the behaviour of the samples at slow changes of stress. Oppositional behaviour at fast load is typically showed at high frequencies. The frequency sweep is very important for polymer melts: by measuring frequency sweeps at different temperatures the characteristics like melting point, glass transition, rubbery-elastic characteristics, entanglement density, and processability can be achieved. There are some features of frequency sweep graphs we should know for characterization. First of all we should know that a typical fluid would show slope of 2 for $G'$ and 1 for $G''$ ($G'$ depends on square of frequency in a log-log plot, and $G'$ depends on frequency) at
low frequency regions, based on mathematical definitions describing small amplitude oscillatory shear tests. This increase in moduli is a behaviour indicating relaxation processes.

However, there are some cases that show different behaviour, like plateau regions. This plateau region is an indication of entanglements or network structures (gel-like) in suspensions and nanocomposites which prevents the relaxation mechanism. The latter case should show up at very low frequency regions to prove the existence of network structures. At higher frequencies (shorter times) movements are quite fast and there is only very local relaxations which are not so sensitive to material structures. That’s maybe why in a series of suspensions with different particle concentrations all graphs corresponding to different particle concentrations converge to roughly the same magnitude at very high frequency regions.

From low to high frequency, $G'$ and $G''$ increase with the frequency. In this case, if we have a low concentrated suspension below the percolation concentration, plotted in log-log scale, the slope higher than 1 for $G''$ and higher than 2 for $G'$. The slopes of 1 and 1 means that our clay concentration is so small that really there is no any interaction between particles. The clays, in fact, are separated enough to avoid any interaction, no possibility of collision by brownian thermal movement, no repulsion or attraction potential (London, Van der Walls...). In typical rheometer measurements, with maximum frequency of 628 [rad/s] = 100 [Hz], we will never see a crossover between $G'$ and $G''$ because the relaxation time is lower than $1 \div 628$ [rad/s]. If we increase a little the concentration we increase the possibility of physical interactions between particles, so the slope of $G'$ and $G''$ versus frequency will decrease. By increasing the concentration of particles a solution a more solid behaviour and solids are less dependent to frequency will be shown. So at a certain concentration we will start seeing the crossover point between $G'$ and $G''$. The inverse of the crossover in [rad/s] units will give us the relaxation time of the suspension. If we increase more the concentration, the crossover frequency will move to lower frequencies as the relaxation time moves to higher values. Of course the slopes also increase. At one point, we don’t see anymore the crossover point although we go to very low frequencies. This means that $G'$ is always higher than $G''$. At this point we have reached a gel, solid like behaviour. The interactions between particles are so high that the particles form a network with physical forces acting as bonds. But this gel is not a perfect solid. Still, there is a slope of $G'$ versus frequency, because we still have a material that his viscoelasticity depends on the observation time. It should be remarked that the relaxation time in polymers is very high but not infinite and we can vary a little his liquid like or solid like behaviour changing the observation time (frequency).

This was a general explanation regarding distributed filler and suspensions. Particle size, particle shape, the chemistry of the particles has a great influence in the dependence of the viscoelastic properties and mechanical stability of the composites materials. Therefore, in order to
better characterize the systems, it is recommended to evaluate their behaviour at low frequency regions to check if there's a gel-like behaviour, network or just an increase in moduli (terminal behaviour). Usually increasing the clay concentrations should lead to plateau behaviour in low frequency regions. So if the material still shows sharp increase in moduli probably it means it didn’t make a network or at least a weak network is obtained, or it's still in a sol state. For these reasons we focused on very low amplitude and low frequency of deformation analysis.

5.2.3.2 Methods and Protocols

The samples prepared for these measurements consisted in rectangular-shaped thin films, dimensioned following the specifications of the instrument, with a width of 10 [mm] and a length of 40 [mm]. Alton Paar Rheometer of DCCI was adopted. The samples were clamped to two gauges, equipped by traction and torque sensors and placed in a thermal insulated chamber. Liquid Nitrogen was used to cool down the chamber and resistors, permitting to archive also low measurement temperature as -80 [°C]. The upper gauge imposed torsion of the sample, while the torque sensor measured its couple.

5.2.3.3 Results

In a rectangular shaped specimen, the torsional stress is no longer linear with the distance from the axis and depends on the angular position along the specimen length. For a homogenous and sufficiently long rectangular cross-sectional sample, the maximum stress is obtained on the mid-points of the long sides, whereas it is zero at the four corners of the cross section, as the theory of elasticity states. The change of the geometry of the same sample affected the results of \( G' \) and \( G'' \) obtained by the measurements.

![Figure 75 - The effects of specimen geometry on moduli obtained by rheological tests at very low strain: the cross-over frequencies are also highlighted with red lines.](image-url)
The effects of specimen geometry (length-to-width aspect ratio) on storage modulus of rectangular rubber samples measured in torsional oscillator rheology have been investigated by C. Dessi et al., founding similar results reported in Fig. 75. They proposed a correction in order to take into account the effect of the stress distribution acting on soft samples in particular geometries of the samples due to the deformation imposed by the measurement.

In fact, the values of the moduli obtained by the analysis appeared overestimated of several orders of magnitude if compared with the results of tensile tests (see Fig. 72). This was also interpreted with the fact that the volume of the filler network involved was different.

On the other hand, by applying the correction proposed by Dessi et al. the modulus obtained were underestimated (see Figure 76).

After verifying that the Shear modulus obtained by traction (through the same instrument used for the rheometry), was perfectly comparable with that obtained by the tensile tests and with that provided from the supplier (for SY0 about 0.9 [MPa]), we didn’t focus on the absolute values of the modulus obtained. Indeed, we focused on the rheological behaviour answer of the materials in a condition of very low deformations, interpreting the results from the filler-matrix interaction point of view, as explained in the first sub-section.

In the Figures 77 and 78 the storage and loss modulus from frequency sweeps of the Sylgard 186-based materials (SY0MT, SY2MT, and SY4MT) and of the Bluesil 4310-based materials (B0MT, B2MT and B4MT) are reported.

All materials, except SY4MT and B2MT (the more loaded composites), show temperature effect on $G'$. All $G''$ tend to show the same answer at all temperature: at highest frequencies the $G''$ curves collapse on the same curve, maintaining the same or comparable values. The $G''$ behaviour is strictly linked to the rheology of particles, small molecules and dangling chains; increasing the temperature, they will impart more movement to the long chains, thus increasing the energy losses, decreasing the values of $G'$.
Figure 77 - Storage and Loss Modulus from Frequency Sweeps, obtained by rheological characterization of the Sylgard 186-based materials: SY0MT, SY2MT, SY4MT.
Figure 78 - Storage and Loss Modulus from Frequency Sweeps, obtained by rheological characterization of the Bluesil 4310-based materials: B0MT, B2MT, B4MT.
Figure 79 - Summary of Storage and Loss Modulus from Frequency Sweeps obtained by Rheological tests with thin films at very low strain.

At low frequency the dangling chains probably have enough time to react to the slow deformations (‘reptation’ effect), while at higher frequencies they tend to flow (liquid-like behaviour). Reptation is the thermal motion of very long linear, entangled macromolecules in polymer melts or concentrated polymer solutions. Derived from the word reptile, reptation suggests the movement of entangled polymer chains as being analogous to snakes slithering through one another. Pierre-Gilles de Gennes introduced (and named) the concept of reptation into polymer physics in 1971 to explain the dependence of the mobility of a macromolecule on its length.

All materials over 0.2 [Hz] of deformation (corresponding to about 1.25 [rad/s]) show this behaviour (see Figures 77 and 78). In SY4MT and B4MT, probably due to the formation of hard and soft domains topology, through a specific dispersion of the filler, this effect is difficult to be observed, as the amount of dangling chains and short chains could be smaller respect to the other cases: this could explains why the curves of $G''$ Vs T for these cases show less dispersion.

All the samples show a typical decrease of $G'$ with the increase of temperature, except for SY4MT and B4MT, which show a slight dependence on T.

SY-based and B-based materials showed the same rheological behaviour, despite B0 showed less dependence by T, with smallest $G'$ modulus respect SY0 (see Figures 79), similarly to the results obtained by the tensile tests.

Looking to the crossover point evolution by varying the temperature in Fig. 80c, for all materials excepted of SY4MT and B4M it can be observed a behaviour coherent to the theory, in which the decrease of $G'$ with T in concomitance to a linear increase of $G''$, brings a shift of the crossover point to lower frequencies. It is reminded that from the definition, the crossover point corresponds to the condition in which $\tan(\delta)=1$. For materials SY4MT and B4MT a quasi constant value is shown varying the temperature.
Figure 80 - Effect of the temperature on the Storage Modulus from FS at 0.1 [Hz] and 0.01% strain for the SY materials (a) and B materials (b). In c) and d) the crossover point frequencies are reported.

SY4MT and B4MT revealed very interesting and different rheological behaviour with respect all other materials, indicating the existence of different conditions between filler and polymer interactions. This was again attributed to the presence of an efficient secondary network in these materials, which stabilized the primary one, reducing the relaxation phenomena.

5.2.4 Dielectrical characterization

The dielectrical characterization was carried out in G2Elab of INP de Grenoble. The main subject was to estimate the dielectric permittivity of the materials synthesized. Furthermore, an Electrical breakdown study (Clàquage analysis) was conducted to estimate the electromechanical properties of the composite materials realized, and for evaluate the effect of the order of nanoclays distribution.
5.2.4.1 **Dielectric Spectroscopy**

5.2.4.1.1 **Model**

The Dielectric spectroscopy is a measurement of the capacitance \( C \) when a sinusoidal electrical current is applied on a sample, by analyzing the voltage response in terms of amplitude and phase shift respect the current:

\[
C(\omega) = \frac{Q}{V(\omega)} = \varepsilon_0 \varepsilon_r(\omega) \frac{A}{Z}
\]  

(90)

where \( A \) is the surface of electrodes, \( Z \) is the thickness of elastomer, \( \varepsilon_0 \) is the vacuum permittivity, \( \varepsilon_r \) is the relative permittivity and \( \omega \) is the pulsation of \( V \).

Relative dielectric permittivity \( \varepsilon_r(\omega) \) is a complex function of the pulsation \( \omega \).

\[
\varepsilon^*(\omega) = \varepsilon'_r(\omega) - i\varepsilon''_r(\omega)
\]

(91)

where \( \varepsilon''_r(\omega) \) can be contained in the dissipation term, named loss tangent.

\[
tan(\delta) = \frac{\varepsilon''_r(\omega)}{\varepsilon'_r(\omega)}
\]

(92)

Measuring the capacitance with changing in AC Voltage frequency, and knowing the precise dimension of the capacitors so obtained, the Dielectric Permittivity of the materials \( \varepsilon_r \) can be calculated.

Commercially, the values of \( \varepsilon_r \) at 100 [Hz] and 1 [kHz] are reported for a fast comparison of materials, however, especially for high performance ones, the dependence of \( \varepsilon_r \) on the frequency used can be important. Typically, a noble metal is used for the realization of the electrodes, and for thick samples their dimensions do not affect the measurement. Anyway it has been experimentally observed a significant fluctuation by the change of the dimensional parameters, adopting the same procedure on thin film samples. The latter were opportunely scaled in dimensions in order to avoid false probing caused by field effect: following the specifications reported in \textsuperscript{124}, the distance between the border of electrodes to the borders of sample must exceed the electrode diameter, while the distance between the electrodes must be more than 50 times less.

5.2.4.1.2 **Methods and Protocols**

A dielectric spectrometer with Capacity range of \( 10^{-6} - 1 \) [F], tension range of \( 10^4 - 3 \) [V] and frequency range of \( 10^6 - 10^7 \) [Hz] was adopted: the measurements were conducted with a 3[V] AC Voltage at \( 10^1 - 10^6 \) [Hz]. The system was able to evaluate also the dielectric losses.

The samples, placed on a conductor substrate (deposited gold wafer) inside the measuring chamber, were connected by two gold needles (one at the top electrode and one to the conductor substrate) to the AC source: this configuration, reported in Fig. 81 was adopted in order to avoid an electric field deflection effect that could affect the capacitance measurements.
A first step of the measurement consisted in the deposition of two electrodes on the top and bottom surfaces of the thin film samples, in order to obtain a capacitor with precise dimensions, next measured with digital calliper. In order to ensure a good adhesion between the polymer and the conductor, a metal deposition was realized with the use of Quorum Q150 T ES metallizer. The system created a plasma between the sample and a metal coin reference source by applying an electric current in the presence of an Argon flow in a pressure chamber, while an Azote flow was used for cooling the chamber. Au/Pd (Gold/Palladium 80:20) layer of about 50 [nm] was obtained in 100 [s] by applying a current of 40 [mA] (29 [nm/min]), while Ar pressure was 0.7 [bar] and N₂ pressure was 2.2 [bar].

The deposition was carried out on both the surfaces of each sample, sandwiching it between two identical masks consisting in two polymeric electrostatic layers in which 6 circular holes of 8 [mm] of diameter were previously prepared. About 25 [mm] x 25 [mm] samples with centred circular electrodes were obtained by this procedure. Thus, 6 samples for each material were realized and analyzed.

Two diameter dimensions of electrodes have been considered, 8 and 20 [mm], and differences less than 8% in permittivity have been valued in the measurements. The indetermination on the thickness of the sample was estimated about 5%. A samples electrode diameter of 8 [mm] has been used.

5.2.4.1.3 Results

The curves reported show the main parameters in interest in function of the AC voltage frequency in a range between 10 [kHz] and 0.1 [Hz]. This measurements have got an appreciable dispersion of the results, mainly due to the homogeneity of the thickness of the samples, obtained by a bar coater process, using medium-high viscosity materials (around 10⁵ [cP]). Each curve is the representative one for each material, with an averaged deviation from 2 to 10%.
Dielectric spectroscopy showed an enhancement of performance in almost all samples loaded with OMMT respect the pristine ones. However, the trends of both permittivity and dielectric losses possessed very different behaviour when clay is added, especially at low frequencies: SY0 showed an homogeneous behaviour in frequency, $\varepsilon_r = 2.78$ (close to the supplier value), while the Permittivity increased at low frequencies with the fillers: an amount of 2%wt of OMMT brought $\varepsilon_r@10$ Hz = 3.52 (+26%), $\varepsilon_r@40$ Hz = 3.48 (+24%), and $\varepsilon_r@100$ Hz = 3.45 (+23%), but a further amount, 4%wt OMMT, provided a smaller increase, for which $\varepsilon_r@10$ Hz = 3.10 (+11%), $\varepsilon_r@40$ Hz = 4.05 (+45%), and $\varepsilon_r@100$ Hz = 2.98 (+7%). This non-monotonic trend were explained by a different level of dispersion of the clays, which form aggregates if a proper flocculation is not guaranteed\textsuperscript{121}, thus causing different dielectric contributes. The losses, coherent with literature\textsuperscript{113}, showed an increase at low frequencies, basically due to interfacial polarization phenomena between electrode and polymer, and/or between polymer and filler, and the accumulation of space-charges at the interfaces/interphases of the materials\textsuperscript{125}. A strong increase in the losses can be observed in Fig. 82b at low frequency, probably due to interfacial polarization (between electrode and polymer, and/or between polymer and filler).
B0 showed comparable permittivity with respect to SY0, with \( \varepsilon_r = 2.7 \), and a non-monotonic behaviour was observed when filled: for 2%wt OMMT \( \varepsilon_r @ 10 \text{ Hz} = 2.75 (+2\%) \), \( \varepsilon_r @ 40 \text{ Hz} = 2.70 (+0\%) \), and \( \varepsilon_r @ 100 \text{ Hz} = 2.65 (-1.5\%) \), while for 4%wt OMMT \( \varepsilon_r @ 10 \text{ Hz} = 3.18 (+18\%) \), \( \varepsilon_r @ 40 \text{ Hz} = 3.12 (+16\%) \), and \( \varepsilon_r @ 100 \text{ Hz} = 3.05 (+13\%) \). For the same reasons stated, B2MT seemed belonging to a particularly high exfoliated condition of the clay. In fact, the losses appeared coherent with the amount of filler.

The effect of organically modified montmorillonite (OMMT) on the dielectric properties of silicone rubber was examined by Razzaghi et al.\(^{112}\). OMMT was added to this rubber in two concentration levels similar to the ones used in this study, 2 and 5 wt%. Their results showed that the order of organo-clay layers in the less dispersed structure imparts an additional ionic polarization and higher dielectric permittivity compared to clay layers that are more dispersed and have lost their order. These observations have been taken into account in the micromechanical model proposed by Yi et al.\(^{125}\) to predict the complex dielectric response of nanoclay composites.

The cure of the base of Sylgard 186 by Dicumyl peroxide shows a systematic increase of permittivity with the increase of the amount of filler: the dependence toward the frequency is again strong, due to relevant dielectric relaxation mechanisms acting with an huge amount of fillers (4 and 10% in weight), and also because of the presence of the splitted molecules of the DP: in fact, also in the case without filler the permittivity value appears greater than Sylgard 186 one, with an \( \varepsilon_r @ 40 \text{ Hz} = 3.38 \) and \( \varepsilon_r @ 1\text{kHz} = 3.37 \). It shows an increase of +66% for 4%wt of OMMT at 40 [Hz] \( (\varepsilon_r @ 40 \text{ Hz} = 5.62) \) and +79% for 10%wt of OMMT \( (\varepsilon_r @ 40 \text{ Hz} = 6.04) \).

The losses in filled materials hold a relative maximum at 1 [kHz], while at lower frequencies they don’t show the typical exponential increase exhibited in the other materials and reported in the literature and in the theory; however, it can be observed a smooth increase close to 0.1 [Hz]. In fact, if the effect of the same amount of filler (4%wt) is compared in the silicone matrixes of the commercial SY and SY catalyzed with Dicumyl Peroxide (Fig. 85) it’s clear that the use of the peroxide strongly affects the response of the material, especially in permittivity,

![Figure 84 - Effect of OMMT filler on dielectric relative permittivity \( \varepsilon_r \) and dielectric losses tan(δ) of SYDP.](image)
because its provision of dipole moment from the aromatic rings, while the losses doubles their value with respect the commercial mixture when the filler is added.

The additional space-charge and ionic polarizations of the clays enhances not only the dielectric permittivity \( \varepsilon_r \), but also the dielectric losses \( \tan(\delta) \) of the composites, which is more prominent at low frequencies; both for SY and SYDP composite it is observed a relative maximum at 1 [kHz].

Dielectric loss consists of two components, namely dipole energy loss and ionic conduction: entrapment of more charges in the polymer composite, especially in the spacing between clay platelets, also enhances the conduction current provided by inelastic displacements of charge carriers in the dielectric. This explains the larger values of dielectric loss in the composites under AC field notable in Figure 85b.

The effect of the order of dispersion on the permittivity, but also with respect the frequency was deeply investigated by Yi et al., explaining the strongly dependence on such parameter: Figure 86 reports the results of their interpretation of the phenomena.

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**Figure 85** - Dielectric relative permittivity \( \varepsilon_r \) and dielectric losses \( \tan(\delta) \) of SY(°) and SYDP(°), pristine (blue) and with an amount of 4%wt of OMMT(cyan)

---

**Figure 86** - Comparison of the effect of three order of distribution of MMT nanocomposites on \( \varepsilon_r \). From
Finally, it can be observed in the summary of results of Fig. 87 and Fig. 88 that 40 Hz is an optimal operational frequency, for which the permittivity is higher and the losses, with the exception of Bluesil samples, are lower with respect of 100 [Hz] and 1 [kHz]. Anyway, for all samples, the related losses remained less than 0.04.

**5.2.4.2 Electrical Breakdown Measurements**

When subjected to an electrical field, the dipoles will align with the field, in order for the negative part of the dipole to orient towards the higher potential, while the positive part of the dipole will orient towards the lower potential. Electrostatic energy created in the material is then converted into mechanical energy through different mechanisms, the simplest of which is Maxwell stress, whereby stress is proportional to the dielectric constant. The driving voltage that can be applied to a dielectric elastomer is limited by dielectric breakdown strength, $E_b = \ldots$
Which ideally is a constant for each material. Electrical breakdown mechanisms are complex, as they are dominated by extrinsic factors not inherent in the material, such as imperfections, as well as statistical variations in morphology and microstructure. Recent modelling has focused on determining intrinsic breakdown strengths, and a fundamental understanding thereof will lead to improved materials. Based on this fundamental knowledge, combined with continued progress in processing methods, electrical breakdown strengths closer to the intrinsic breakdown will become realisable. High dielectric breakdown strength is desired so that the DEAs do not experience premature failure. This means that it is important for the elastomer to be free from impurities or voids that cause locally reduced dielectric strength. Since in practice it is impossible to avoid defects, $E_b$ depends strongly on elastomer film thickness, since the number of defects scales with volume and thus thickness. Furthermore, it is important that the material exhibits low dielectric losses, which continuously contribute to the generation of heat in the material. If heat generated by these dielectric losses exceeds heat dissipated into the surrounding area, the temperature in the elastomer will rise, which in turn could lead to thermal breakdown.

5.2.4.2.1 Methods and Protocols

The Electrical breakdown tests were performed on an in-house-built device based on international standards (IEC 60243-1 (1998) and IEC 60243-2 (2001)), while thicknesses were measured through a digital calliper. The samples, prepared with same procedure adopted for the dielectric spectroscopy, were placed between the two cylindrical electrodes (radius of 8 mm). The conditions of breakdown failure were measured at the point of contact by applying a stepwise increasing voltage of 100 [V] from 0 to 12 [kV] in 200 [s]. The breakdown strength of each sample was given by the average of 3 measurements.

5.2.4.2.2 Results

The Breakdown Field evaluated by the electrical breakdown measurements revealed that the adding of OMMT imparts benefits to the system also from this point of view. The results are reported in the summary table Tab.11. All samples containing OMMT showed higher Breakdown strength $E_b$ respect pristine materials, excepted for SY2MT, for which the values was comparable with SY0. $E_B$ for B4MT appeared higher than B0 but slightly lower than B2MT. We interpreted this results with the order of dispersion of the nanoclays, as the previous analysis suggested, and similarly to whom reported from Yi et.al. Moreover, it is well known that a decrease of the cross-link density produces a less electromechanical stability. Thus, a contemporary reduction of chemical cross-links of the matrix, an increase of physical cross-links due to clay, and different order of dispersion of the latter, gives the possibility of an enhancement of electro-mechanical integrity.
5.2.5  **Thermo-gravimetric Analysis**

The TGA analysis was carried out in the Polymer Materials Laboratory of DCCI of UniGe. TGA is a method of thermal analysis in which the mass of a sample is measured over time as the temperature changes. This measurement typically provides information about physical phenomena, such as phase transitions, absorption and desorption, as well as chemical phenomena including chemisorptions, thermal decomposition, and solid-gas reactions (e.g. oxidation or reduction).

5.2.5.1  **Methods and Protocols**

Thermo-gravimetric analysis (TGA) was performed by a TGA/DSC 1 STAR® System from Mettler Toledo in a nitrogen atmosphere with a heating rate of 10 [°C/min] from 25 to 900 [°C] to evaluate the filler concentrations in the materials. By taking into account the previous results, TGA were performed exclusively on LSR samples (B0, B2MT, B4MT), the more promising for our goals.

5.2.5.2  **Results**

Delebecq et al.\(^{126}\) carried out an exhaustive TGA study on silica-PDMS model samples, mimicking high-temperature vulcanizing (HTV) silicone and LSR formulations. They found out that without platinum in the recipe all the PDMS chains de-polymerized into volatile cyclosiloxanes, leading to no residue. With platinum, the residue content depended on the silica content and silica surface modification because an induced ceramization occurred.

Figure 89 reports the degradation curves under N2 of B0 and each part of formulation of B0. The two parts, named B0\(_A\) and B0\(_B\), are prepared from the same base, i.e., silica and a vinyl-terminated poly-dimethyl-siloxane; B0\(_A\) additionally contains the platinum catalyst, whereas B0\(_B\) adds on a few percent of hydrido-functionalized silicone chains as a cross-linker. The latter degraded at lower \(T\) with respect B0 (see Fig. 89), and same effect was observed for B2MT and B4MT (Fig. 90), coherently with the decrease of cross-link expected.
All formulations behaved similarly, with no significant degradation taking place before 250[°C], and a slow degradation up to around 500[°C], where weight losses of about 10% were detected. In Figure 90 it can be noted that B2MT and B4MT showed a degradation between 300 and 350[°C], which was not showed in B0. An increase of percentage residue at 900 [°C] by increasing the OMMT continent was observed; on the other hand, the values obtained seemed higher with respect the filler contents estimated.

Confirming the observations reported from Delebecq et al.\textsuperscript{126}, B0\textsubscript{B} showed a 19.6% of residue, thus attributed to the silica content, while B0\textsubscript{A} revealed a 26.9% of residue, 7% more than B0\textsubscript{B}. The final residue measured for B0 was higher than B0\textsubscript{A}, indicating a further ceramization occurred in presence of highly constrained cross-linked chains around silica\textsuperscript{126}. Similarly, when OMMT was added to the system an induced ceramization residue was observed: the difference in residues was coherent with the OMMT amount (see Tab.12). The OMMT ceramization is a sought-after fire protection property deeply exploited in industry\textsuperscript{111}.

| Formulation | OMMT [wt%] | $T_{\text{229}}$ [°C] | $T_{\text{max}}$ [°C] | Inorganic Residue [wt%] | Estimated silica [wt%] | Ceramization residue [wt%] |
|-------------|------------|----------------|----------------|-----------------|-----------------|----------------|---|
| LSR 3410 (part A) | - | 264 | 595 | 26.9 | - | 7.0 |
| LSR 3410 (part B) | - | 258 | 593 | 19.6 | 19.6 | 0 |
| LSR 3410 (B0) | 0 | 302 | 622 | 32.4 | 19.6 | 12.8 |
| B2MT | 2 | 297 | 601 | 36.5 | 19.6 | 14.9 |
| B4MT | 4 | 297 | 609 | 38.9 | 19.6 | 15.3 |

Table 12 - Results obtained by TGA on LSRs (Silbione-based samples).
Figure 90 - TGA of Silbione samples: B0, B2MT and B4MT.

A summary of the data obtained from TGA is shown in Table 12. Key values are defined here as temperatures at 2% weight loss ($T_{d2\%}$) and the temperature ($T_{\text{max}}$) at which the fastest derivative weight ($^\circ\text{C}^{-1}\%$) was observed, as well as the final inorganic residue at 900[$^\circ\text{C}$]. The temperature of $T_{d2\%}$ is especially important with respect to production scenarios. One serious drawback with a low $T_{d2\%}$ would be bubble formation during production or the elastomer curing weakening the produced materials. The amount of the silica content and the ceramization residue was thus estimated through the results of TGA and observations reported in literature.

**5.2.6 Swelling measurements**

Also the Swelling measurements were carried out in the Polymer Materials Laboratory of DCCI of UniGe. Extraction and swelling measurements can be performed on cross-linked materials to estimate the cross-link degree and the average molar mass between cross-links ($M_c$) via the Flory–Rehner equation 127.

When immerged in an opportune solvent, elastomer samples tend to swell more or less depending on their cross-link density. Such cross-links encompass the chemical links between PDMS chains and the physical interactions between the PDMS chains and the filler network.

**5.2.6.1 Methods and Protocols**

Thin films of initial dry weight $W_i$ of about 0.2 [g] are plunged into 100 [ml] of cyclohexane in a Petri plate. The sample are extracted, gently wiped on each side to remove liquid
solvent at the sample surface and immediately weighted to measure the equilibrium swollen weight $W_s$ along the time. The samples are then dried overnight at 70 [$^\circ$C] under vacuum and reweighted ($W_f$). The extractable material and polymer volume fractions in the swollen sample, respectively $E$ and $V$, can be calculated as follows:

$$E = \frac{W_i - W_f}{W_i(1 - c)} \times 100$$  \hspace{1cm} (93)

$$V = \frac{W_i - W_f}{W_f - W_i c + (W_s - W_f) \frac{\rho_p}{\rho_s}}$$  \hspace{1cm} (94)

where $c$ represents the filler weight fraction, $\rho_s$ and $\rho_p$ the density of the solvent and polymer respectively. From this volume fraction it is possible to estimate the average molar mass between cross-links ($M_c$) via $M_c$ via the Flory–Rehner equation 127:

$$M_c = \frac{-M_s \frac{\rho_p}{\rho_s} (V^{\frac{1}{2}} - \frac{V}{2})}{\ln(1 - V) + V + \chi V^2}$$  \hspace{1cm} (95)

with $M_s$ the molar mass of solvent, and $\chi$ the Flory–Huggins interaction parameter which in case of PDMS-methylcyclohexane equals 0.45 88. The value of the averaged molar mass between cross-links ($M_c$) gives an idea of the global effect of the adding of OMMT; $M_c$ is one of the main parameter driving the elastomer mechanical properties (due to entropic elasticity), and differs greatly from a system to another. The cross-link density $\nu$ is a measure of the degree of cross-linking in a polymer and is defined by

$$\nu = \frac{\rho_p}{M_c}$$  \hspace{1cm} (96)

This level of detail of the formulation of the silicones is proprietary and is not disclosed by the suppliers; however, it is useful because, in principle, it can be related to their physical properties 128.

5.2.6.2 Results

The LSR samples showed similar behaviour, finding after a pre-extraction process a maximum in percentage of swelling, quite high, as in can be noted in Tab.13, at similar measurement times.

The trends reported in Figure 91a, in which the swelling percentage showed different phases, were explained as the solvent molecules gradually solvates the siloxane chains, permitting the movement and the eventual expulsion of small molecules or small un-cross-linked chains. This process is called extraction, and led to a higher distension of the chains between the cross-links.
Figure 91 -  Results obtained by swelling measurements of LSR samples (B0, B2MT, B4MT)

By the use of the filler weight fraction obtained from TGA, the cross-link density and the extracted material (related to the gel fraction) of the LSR samples were calculated. Increasing the amount of OMMT, the cross-link degree density decreased, while the averaged molar mass between cross-links increased, as expected because the partial inhibition of the cross-link catalyst.

The evaluated molecular mass $M_e$ appeared moderate if compared to the values reported in literature\(^{117}\), meanwhile the extracted material was only around 2%. The latter also slightly increase with the amount of OMMT. Swelling measurements confirmed the observation from the previous analysis.

<table>
<thead>
<tr>
<th>Material (Silbione 4310)</th>
<th>OMMT [%wt]</th>
<th>Max Swelling [%]</th>
<th>Equilibrium Swelling [%]</th>
<th>Extracted material $E$ [%]</th>
<th>$M_e$ [g/mol]</th>
<th>Cross-link density $\nu$ [mol/m$^3$]</th>
</tr>
</thead>
<tbody>
<tr>
<td>B0</td>
<td>0</td>
<td>481</td>
<td>462</td>
<td>1.66</td>
<td>6000</td>
<td>160</td>
</tr>
<tr>
<td>B2MT</td>
<td>2</td>
<td>458</td>
<td>418</td>
<td>1.82</td>
<td>8500</td>
<td>117</td>
</tr>
<tr>
<td>B4MT</td>
<td>4</td>
<td>452</td>
<td>365</td>
<td>1.99</td>
<td>11000</td>
<td>88</td>
</tr>
</tbody>
</table>

Table 13 -  Summary of results obtained by swelling measurements of LSR in cyclohexane.

5.2.7  TEM analysis

TEM analysis was provided by IIT of Genova thanks to collaboration. The morphology and dispersion of the filler in the polymeric matrix was investigated using an integrated compact Transmission Electron Microscope (TEM) Jeol JEM 1011, operating at 100 [kV], equipped with a Gatan Orius SC-1000 CCD camera. Ultra-thin samples were prepared by Leica EM UC6 Ultramicrotome, previously cooled at -160[°C] using a cryo-system Leica EM FC6.
and a specific diamond knife for low temperature cut. Slices were collected and laid down onto a 200 mesh formvar/carbon-coated copper TEM grids, and then analyzed.

Despite the difficulty of preparation of the samples through the microtomy process, mainly because of their very low thickness and high softness, two representative TEM images of the composites B2MT and B4MT are reported in Figures 92 and 93. In Figure 92 it can be noted that the clays are partially packed in macro regions, surrounded by the silicone matrix and silica particles (well displayed because of the transmission beam), and partially dispersed in single palettes (highlighted with circles). From the TEM analysis different levels of dispersion of the filler were confirmed. These are the responsible on the effects observed in our composites.

However, an estimation of the characteristic of the secondary network by the study of the composite morphology was not possible, because of the problems found in the preparation of the samples, which thus brought to low quality images, as previously stated.
5.2.8 Summary

The formation of the chemical (primary) network, formed by long and short chain fractions and low molecular mass molecules cross-linked by an hydrosilylation reaction carried out by Pt catalyst, was partially inhibited by the surface modifier of the fillers (quaternary ammonium salts). On the other hand, a secondary network composed by the filler loading (fumed silica and nanoclay) consisted in a physical network which provided stability to the primary one. The resulted bimodal network explained the improvement of all mechanical properties. Similar observations have been made by Madsen et al. and Bejenarius et al. and Romasanta et al. in their studies focused on heterogeneous bimodal networks, observing low viscous losses also in case of low moduli, thus not attributed to incomplete reactions and rather of network artifacts, but to a specific network formation\textsuperscript{129,130,131}. They found out those domains with the clusters of small polymers act as a reinforcing structure of the soft network from the long polymers, thus reducing the viscous losses. Also Shah et al. noted interesting mechanical performances with silicone hybrid bimodal networks though the use of functionalized titania providing bridging chains and creation of localized short spaced cross-links\textsuperscript{132}.

An increase of dielectric permittivity was obtained for all films containing OMMT, despite the effect of operational frequency clearly occurred, while dielectric losses remained at a very low level. Electrical breakdown strength also improved with OMMT respect pristine materials, and the effect of the amount and the order of dispersion of such filler were highlighted.

The synergistic effect of reducing elastic moduli, increasing dielectric permittivity and dielectric breakdown strengths thus resulted in globally improved performances.

In order to evaluate the performance of a DEs realized, a method proposed by Sommer-Larsen and Larsen\textsuperscript{133} was adopted, based on the calculation of the figure of merit, \( F_{om} \), defined as \( F_{om-SL} = 3 \varepsilon_0 \varepsilon_f E_B / E \). The latter was conceived for comparing different materials in condition of constant potential in a relaxed status. Despite without considering the mechanical and electrical losses, which for our materials appeared minimal, the figures of merit of the latter are calculated and reported in Tab. 14. Interestingly, it seems that the OMMT in our systems shows beneficial effects proportional with its amount in weight: in fact, if ones compare the normalized \( F_{M-SL}/F_{M-SL_{SY0}} \) and \( F_{M-SL}/F_{M-SL_{BO}} \), very similar trends are observed. A 2\%wt of OMMT in SY samples (RTV rubber material) brings to an enhancement of the performance of 220\%, while in B samples (LSR formulations) brings to a global improvement of 310\%. A 4\%wt of OMMT shows the same enhancement of the performance on both the matrixes, with a surprising value of 530\%. The Figure of merit is a very useful index for comparing the performance of dielectric elastomers; anyway, the one proposed by Sommer-Larsen et al. does not take into account important parameters such us mechanical and electrical losses. In order to taking into account also such parameters, a new figure of merit is introduced in this thesis. It is defined as
$F_{om} = 3\varepsilon_0\varepsilon_r\tan(\delta)\sigma_{\%\text{loss}}E_B^2/E$, where $\tan(\delta)$ denotes the dielectrical losses and $\sigma_{\%\text{loss}}$ the stress percentage losses. Using the figure of merit proposed in this work, our materials appeared very promising (see Table 14).

Such interesting performances could be exploited both for actuation and generation purposes\textsuperscript{84,124,134}.

Among the materials realized, following the observations from the experimental analysis taking into account the requirements of our system, and the composite material B4MT was chosen for the realization of DEGs.

<table>
<thead>
<tr>
<th>OMMT [%wt]</th>
<th>SY0</th>
<th>SY2MT</th>
<th>SY4MT</th>
<th>B0</th>
<th>B2MT</th>
<th>B4MT</th>
</tr>
</thead>
<tbody>
<tr>
<td>Silica [%wt]\textsuperscript{+}</td>
<td>(30)\textsuperscript{−}</td>
<td>(30)\textsuperscript{−}</td>
<td>(30)\textsuperscript{−}</td>
<td>19.6</td>
<td>19.6</td>
<td>19.6</td>
</tr>
<tr>
<td>$n$ [mol/m\textsuperscript{3}]\textsuperscript{+}</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>160</td>
<td>117</td>
<td>84</td>
</tr>
<tr>
<td>$E$ [MPa]\textsuperscript{a}</td>
<td>0.87</td>
<td>0.48</td>
<td>0.24</td>
<td>0.52</td>
<td>0.35</td>
<td>0.17</td>
</tr>
<tr>
<td>$\sigma_{%\text{loss}}$\textsuperscript{b}</td>
<td>0.085</td>
<td>0.087</td>
<td>0.034</td>
<td>0.087</td>
<td>0.106</td>
<td>0.101</td>
</tr>
<tr>
<td>Rupture [%]\textsuperscript{a}</td>
<td>280</td>
<td>170</td>
<td>220</td>
<td>690</td>
<td>1050</td>
<td>1151</td>
</tr>
<tr>
<td>$\varepsilon$\textsuperscript{b}</td>
<td>3.73</td>
<td>3.54</td>
<td>3.14</td>
<td>2.75</td>
<td>2.75</td>
<td>3.23</td>
</tr>
<tr>
<td>$\delta$ [#]\textsuperscript{b}</td>
<td>0.0023</td>
<td>0.0118</td>
<td>0.0186</td>
<td>0.0038</td>
<td>0.0358</td>
<td>0.0404</td>
</tr>
<tr>
<td>$E_B$ [V/m]\textsuperscript{c}</td>
<td>19.1</td>
<td>18.3</td>
<td>21.3</td>
<td>24.8</td>
<td>35.7</td>
<td>30.2</td>
</tr>
<tr>
<td>$F_{M-SL}/F_{M-SL,SY0}$</td>
<td>1.0</td>
<td>2.2</td>
<td>5.3</td>
<td>2.8</td>
<td>8.9</td>
<td>14.9</td>
</tr>
<tr>
<td>$F_{M-SL}/F_{M-SL,SY0}$</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>1.0</td>
<td>3.1</td>
<td>5.3</td>
</tr>
<tr>
<td>$F_M/F_{M,SY0}$</td>
<td>1.0</td>
<td>10.8</td>
<td>104.8</td>
<td>4.6</td>
<td>109.2</td>
<td>223.8</td>
</tr>
<tr>
<td>$F_M/F_{M,B0}$</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>1.0</td>
<td>23.8</td>
<td>48.8</td>
</tr>
</tbody>
</table>

Table 14 - Summary of the characterization of the materials realized. \textsuperscript{1}TGA, \textsuperscript{2}Swelling (in cycloexane), \textsuperscript{3}Tensile tests: \textsuperscript{a} from Yeoh polynomial, \textsuperscript{b} from cyclic tests at 20[mm/min] to $\lambda=2$, \textsuperscript{c}Dielectric Spectroscopy (at 40 [Hz]), \textsuperscript{d}Electrical breakdown, \textsuperscript{e}Figure of merit, normalized to SY0 and B0, \textsuperscript{f}proposed in Sommer-Larsen et al.\textsuperscript{133} and ** in this work. \textsuperscript{g}From supplier.

5.3 Stretchable electrodes

The electrodes of dielectric elastomer actuators are one of the most important aspects on the device’s performance: they must be conductive, yet they must be soft; they must sustain large deformations while remaining conductive, yet they must be able to do so for millions of cycles. The life of a compliant electrode for DEAs is a tough one, and it comes as no surprise that the ideal electrode has yet to be developed. However, much progress has been made from the first reported devices whose electrodes were hand-painted carbon grease, and over the years, creative new methods and technologies have started to emerge.

5.3.1 State of art

Particulate and grease electrodes adhere poorly to the surface of the elastomer and are hence unstable over repeated actuations. For example, greases may dry up or get rubbed off, while
powders may dislodge; this results in a reduction, or even a loss, in electrical conductivity over cycles. These issues of electrode stability can be alleviated by impregnating the conductive particles in the elastomer matrix to prevent their ablation. However, such electrodes generally have high electrical resistances, typically above the range of [kΩ/cm]. A lower electrical resistance can be attained by impregnating more conductive particles, but this leads to an increase in electrode stiffness. In order to address these issues of stability and electrical resistance, metal thin-film electrodes can be considered for use in DEAs. Metal thin films on elastomeric substrates have been proven to make reliable stretchable electrodes with low electrical resistances. Although flat-metal films fracture easily upon stretching, they can readily flex without fracture.

They can be designed into a stretchable form that favours flexing and allows large deformations of the elastomeric substrate. Serpentine patterning can make metal thin films on elastomers stretchable in the in-plane direction, but it reduces the electrode area coverage, which would result in a decrease in the generated electrostatic force. Alternatively, the corrugation of metal thin films on elastomer substrates offers full-area coverage and uniaxial stretchability. For example, Danfoss PolyPower’s corrugated silver thin-film electrodes could be mechanically stretched by up to 80% linear strain, while its DEAs could actuate up to 15% linear strains. However, corrugated metal thin-film electrodes are generally stiffened in the transverse axial direction and are consequently almost inextensible in that direction.

Other strategies for obtaining transparent electrode by embedding an ultrathin silver nanowire network in the surface layer of an elastomeric matrix was also investigated. The composite electrodes retain high surface conductance at tensile strains up to 60%, and can be stretched repeatedly with minimal loss of electrical conductivity under both slow and fast strain rates. However the control of the morphology typically affects the final properties of the materials.

Conductive polymers (conjugated polymers) have been investigated in the field of stretchable electronics. Although a variety of conducting polymers, such as poly-acetylene (PA), poly-aniline (PANI), poly-pyrrole (PPY), and poly(3,4-ethylenedioxythiophene) (PEDOT) have been developed for diverse applications, their wide-spread use is limited by their poor mechanical properties. For example, a poly(3,4-ethylenedioxythiophene):poly(styrene sulfonic acid) (PEDOT:PSS) film, which is widely employed in plastic electronics and organics-based optoelectronic devices, showed high electrical conductivity up to 1000 S/cm, but its breaking strain was below 10%. This level of tolerance to strain is not acceptable for the above-mentioned applications.

Also some blends have been considered for allowing the conductivity of elastomers. For instance PEDOT:PSS blends have been deeply studied, but also in this case the morphology was a crucial issue for both controlling and understanding the charge carrier transport in the
PEDOT/PSS system, and the compatibility of the matrix revealed to be very important also for the
electro-mechanical stability. In fact Noh et al., to reduce the hydrophobicity of the PDMS and
make possible blending with the PEDOT:PSS, used poly(dimethyl-siloxane-b-ethylene oxide)
(PDMS-b-PEO), which is hydrophilic, as a third component. The PEDOT:PSS:PDMS:PDMS-b-
PEO blends were cross-linked using a unique curing agent.

5.3.2 Strategies explored

A first approach explored for obtaining stretchable electrodes was the use a mixture of a
silicone matrix filled with silver and copper nanofibers. The composite, in order to conduct
electrical current must have more than 40% wt/wt of metallic particles, with a Young modulus of
more than 2 [MPa] (about 60 shore A) and maximal stretching of less than 20%. Moreover, the
minimal thickness necessary to obtain a homogeneous surface was not less than 700 [μm].
Excepted for the problem related to the thickness, found for experimental limits, these are very
common problems in the design of stretchable electronics, as stated in the previous sub-section.

By the help of the geometry an alternative to the use of thick and high filled elastomers for
the electrodes was proposed: as the technological problem of carbon grease is related to its
degradation by drying and gradual removal, a method for encapsulating it inside a protected layer
of silicone is investigated. This method revealed the advantage of avoid the otherwise inevitable
 stiffening of the electrodes: having the electrodes higher Young modulus then dielectric
elastomer, the thickness of the layer of conductor must be minimized in order to minimize the
stiffness induced to the composite. In fact, the carbon grease consists in a viscous mixture, and in
terms of final material, at equal volume of elastomer and DEG, the composite appears softer.

5.4 Realization of Dielectric Elastomer Generators

5.4.1 Architecture of Dielectric Elastomer Generators

The composite B4MT, for which its realization and characterization was presented in the
previous sections, was chosen as the best performance material for our goal.

A simplified method was adopted for its realization in order to allow a cheaper and faster
method, since literature very often reports methods which involve expensive processes and
instruments; this brought the possibility of leading different attempts of DEGs realization
strategies, in complete autonomy, in Physic Department of Unige.

Bluestar Liquid Silicone rubber 4310 part B was thus mixed before with Cloisite 20A with
an accurate slow hand mixing, and after with part A, which contained the catalyst, with a filler
concentration of 4%wt. The mixture was placed on a substrate of rectified sheet of aluminium
covered by Teflon fibre tape, on the boards of which two layer of electrical tape were arranged.
With the use of a smooth cylindrical bar of brass of diameter 5 [mm] and length 25 [cm], the viscous mixture (of order of 200 000 [cP]) was spread on the surface created with a slow hand translational movement, repeated if necessary (see Fig. 94). The form was then placed in oven for a cure of 15 [min] at 180[°C]. After cooling, the film was gently peeled-out and placed on a millimetre sheet of paper. The layers obtained by this method reached thickness less than 50 [μm] with good isotropy, thanks to the curing temperature which imparted homogeneity on the surfaces of the viscous layers. Anyway, in order to properly handling the layers (differently to an industrial process), a limiting thickness of 150 [μm] was imposed in the manual process for avoiding induced defects.

Very similar results in terms of mechanical and electrical properties of the films have been obtained with the described cheaper and faster method, with respect the method of realization reported in the sub-section 5.2.1.2.

A mask having three rectangular windows was positioned on the Dielectric elastomer film, and graphite grease was gently deposited to forming a homogeneous layer.

Two contacts made by elastic silver texture (known as silver socks) were placed on two of the three electrodes realized after removing the frame. Then, the dielectric elastomer was peeled out from the millimetre sheet of paper, and folded in order to obtaining a sandwich-type structure.

The configuration must allow the insulation of the electrodes by the active dielectric part (the layer which remained encapsulated, see Fig. 95).

Exploiting the high electrostatic properties of these thin films, a simple pressing was sufficient to guarantee a sealant closure of the smart material. Therefore, a very robust waterproof DEG was obtained. A further step can be introduced in the presented method in order to cross-linking the borders of DEGs (with a RTV silicone).
Following the observations deducted from the first DEGs realization, and taking advantage of the information obtained from the aeroelastic study, an alternative procedure was developed.

Since the capacitance was inversely proportional to the thickness of the sample and proportional to the surfaces of the electrodes, and since a too thin DEG brought to very low forces also after stretching, bringing to low oscillation frequencies, the idea of using two different layers for the realization of DEGs was investigated.

As it can be observed from Fig. 96, adopting the previous procedure for the realization of two electrodes (instead of three) on a thicker dielectric elastomer substrate, and placing a thinner dielectric membrane between them, an improvement of robustness, performance and handleability was achieved.

In fact, also the practical limits found in the use of a very thin elastomer for the architecture of the DEGs, was thus overcome.

Figure 95 - Realization process of Water-proof DEGs.
5.4.2 Mechanical Characterization

Adopting DEGs realized with the procedure reported in Figure 96, tensile tests were carried out using a home-made traction setup, composed by a micrometric handler, a 5 [N] load cell, a digital oscilloscope (Picoscope) and a fast camera described in section 2.2.

The DEG examined was had the following external dimension: width of 50 [mm], length of 127 [mm], thickness of 0.88 [mm].
As it can be noted in Figure 97, two curves are reported: they differ by the initial section of the sample considered. Considering $A_0$ the section of all composite material, composed by 3 layers of elastomers and two layer of carbon grease, a storage modulus of 20 [kPa] was obtained. On the other hand, if $A_0$ was considered the section of only the 3 layers of elastomers, to which the stress imposed was effectively applied, a storage modulus of 23 [kPa] was obtained.

5.4.3 **Electrical Characterization**

An electrical characterization was carried out by measuring the capacitance and the resistance (in series) between the two electrodes and at the extremes of the electrodes (in series), respectively, through a digital multimeter varying the stretching.

The active part of the DEG had width of 30 [mm], length of 127 [mm], thickness of 0.22 [mm] and dielectric constant of 3.4. The resulted capacitance in the relaxed status was therefore 0.52 [nF], a value relatively high. By applying a stretching to DEG the capacitance grew following the curve in Figure 98a. The series resistance was quite high, around 10 [kΩ], grew following a quasi-linear curve (see Fig. 98b), and reached very high values, around 70 [kΩ]. The layers of graphite grease were very thin in this case, and an improvement of such parameter was easily obtainable with thicker ones. Anyway, for our goals this was a satisfactory result, since the electrodes of DEG at stretching of 1.6 were still conductor.

![Figure 98 - Effect of stretching on Capacitance (a) and Resistance (b) vs. Stretching of DEG realized.](image)

5.5 **Self Priming Circuit**

After reporting the operational cycles of the DEGs in a boots step up converter mode (CASE M in the sub-section 5.1.2.2), and a simplified equivalent circuit (sub-section 5.1.3), this section aims at presenting an electronic circuit solution individuated for efficiently increase the input voltage provided by the EMc.
As already introduced in section 5.1, the complete technology of DEGs is composed by the DEG materials, a priming circuit (PC), which transfers high potential charge onto/off the DE electrodes, and a power extraction circuit that harvests the generated power. To generate power, the PC must charge and discharge the DEGs in synchronization with the DEGs capacitance change. A simple circuit to do this exists: the self-priming circuit (SPC). The SPC consists of diodes and capacitors that passively switch between charge delivery and charge receiving states in synchronization with the DE’s capacitance change. Until now, there has been no understanding of how to design an SPC in order to maximize harvested energy from the DE. A new mathematical model for the design and optimization of SPC has been proposed by Illenberger et al.\textsuperscript{156}. 

A typical DEG system is depicted in Fig. 99 and consists of dedicated priming, charge extraction, and control circuitry. The SPC replaces the priming circuitry and portion of the control circuitry, as indicated by the dashed area. Energy extraction and the control associated with it must still be performed by another circuit. Typically, a flyback or buck converter powered by a lithium-ion battery is used. This allows the lithium-ion battery to power the extraction circuitry and accept the energy generated by the DEG system. When extracting energy from a DEG/SPC system, the electrode resistance of the DEG plays a significant role in determining where the extracted energy is from. A high electrode resistance will result in more energy being extracted from the SPC than the DEG. The energy generated can be used to power various off-the-shelf electronic devices (e.g., wireless sensor nodes, personal fitness trackers).

A first-order SPC is shown in Fig. 100(a). If the SPC capacitors $C_{a1}$ and $C_{b1}$ are of equal value, charge will be shared evenly during circuit operation, resulting in the best performance.
Higher order SPCs are realized by paralleling and modifying the first-order circuit, as depicted by the $n^{th}$-order SPC in Fig. 100(b). For circuits higher than first order, capacitor values are referenced to the highest capacitance present in the circuit, $C$. For an $n^{th}$-order SPC, the capacitance values are given by $C_{ax} = C_{bx} = C/x$.

The model proposed by Illenberger et al. allows the optimization of the circuit for voltage boosting in terms of SPC capacitance and order. The optimums were found to depend on DEG capacitance and swing.

With optimum voltage boosting per cycle, it is possible to boost voltage rapidly to working levels for harvesting. This would enable low-voltage start-up and eliminates the need for an external high-voltage source.

### 5.6 Prototypes

With the help of the information provided by the multi-disciplinary study, a proof of concept prototype exploiting both the electrical extraction strategies, EMc and DEGs, is proposed, and reported in Fig.101.

The frame of the device shown in Figure had external dimensions of 10x12x24 [cm$^3$], while the wing adopted had a chord $C$ of 40 [mm] a span $S$ of 100 [mm] and Pivot Point $PP$ coinciding with the Leading Edge $LE$. The electro-magnetic coupling was realized using electro-mechanical parameters similar to those reported in Chapter 4, with two coils of 10 [mH] and 150 [ohm] placed on the rotational axis of the wing, connected with four spring-shaped copper wire (in order to avoid fatigue failure).

![Figure 101 - A FLEHAP prototype exploiting EMc and DEGs at 5 m/s](image-url)
Two series of magnets of 10x10x10 [mm$^3$] and 38 [MOe] in alternated polarity in front of their predicted trajectory were collocated. The DEGs were pre-stretched with a value of $\lambda_s=1.3$ and fixed by a screw system, and connected in parallel in order to enhance the capacitance of the system. The capacitance $C_0$ obtained showed a value of 1.8 [nF].

The electro-mechanical parameters chosen allowed considerable self-sustained oscillations in a wind speed range interesting for our applications, between 3 and 6 [m/s], producing with the EMc an output power between 1 and 6 [mW] with the use of commercial electronics reported in the sub-section 4.3.3 (EH300 Energy Harvesting Module).

The DEGs showed a relative deformation of about 11%, obtained from the maximal deformation $\lambda=1.45$ respect the pre-stretching deformation $\lambda_s=1.3$, producing a proportional change in capacitance from 1.8 to 2 [nF].

With our calculation, a Self Priming Circuit similar to what has been proposed by Illenberger et al.$^{156}$ (see Fig.100a), could be composed by a cascade of 44 capacitors (22$^{nd}$-order SPC) for ensuring the step-up conversion mode. A dedicated SPC is currently on development with the collaboration of DITEN of UniGe.
6 Conclusions

A multi-disciplinary study permitted the development of a novel aeroelastic-based energy harvesting device. The innovative outline of the mechanical system appears very simple, cheap and without rotational parts. It is composed by a passive rotating airfoil bonded to two elastic elements parallel to a fluid flow.

The system developed and investigated showed very interesting aeroelastic effects that can be exploited for the generation of electrical energy. In particular, the self-sustained limit cycle oscillations exhibited by the system (LCOs) are characterized by large amplitude and high frequency of oscillation, as well as large pitch angles, in a wide range of wind speeds, included the low ones. Thanks to the large amount of parameters with which is possible to act, the system appeared very versatile. Indeed, moving in the space of the explored parameters, relevant LCOs can be always obtained in very different external conditions. The latter include the density of the fluid, since FLEHAP demonstrated its application also in water. The aeroelastic study revealed that specific LCOs correspond to the most efficient conditions for the energy extraction purpose. Differently from what literature reports, for which the phase between the two degrees of freedom (pitch and plunge) should be 90° in order to efficiently convert electrical energy (always positive work), the experimental evidences suggested that the latter in our system must be always less than 90°, typically between 20- and 60°. Despite the system is still lacking predictive models, some methods aimed at maximizing the parameters of our interest have been identified, mainly through phenomenological models and experiments, thus demonstrating its technological applications.

Several prototypes have been investigated in terms of energy extraction.

The first strategy for extract electrical energy is the electromagnetic coupling (EMc), using moving coils placed on the airfoil Pivot Point axis and fixed permanent magnets. The system exploiting the EMc is now a very mature device, with a Technology Readiness Level (TRL) between 4 and 5. The latter demonstrated to generate powers in the order of several [mW] also by the use dedicated electronics, able to store the produced electrical energy in a supercapacitor. Such power is more than enough to supply sensor nodes in applications such as Internet of Things (IoT) and Wireless Sensors Networks (WSN). The know-how provided by many experiments permitted to found a valid approach aimed at the optimization of the performance of the devices.

The second strategy investigated is the use of Dielectric Elastomer Generators (DEGs) as soft elastic elements of the aeroelastic system, for extracting energy through their deformation. A new soft dielectric elastomer with improved performances has been prepared through the use of commercially available silicone rubbers and organo-modified nanoclay. The competition between a partial inhibition of the cross-linking, caused by the nanoclay surface modifier, and an increase of physical cross-links imparted by nanoclays and their related stiffness, brought a decreases in
Young's moduli, without decrease mechanical stability, and thereby the lifetime of the dielectric elastomers. Conversely, a specific bimodal network formed by long siloxane chains, silica and a particular dispersion of the nanoclays decreased the viscoelasticity of the material. An enhancement of about 500% on the electro-active performance was obtained by the use of the method proposed. DEGs designed for our scopes have been realized with the most promising material obtained, demonstrating the manufactory of such smart materials even in the absence of sophisticate processes and instruments.

The DEGs realized have been integrated in prototypes using EMc, thus constituting hybrid devices. Despite the DEG electronics is currently in development, the preliminary experiments produced promising results. The adoption of both strategies in the same device provides an appreciated synergy, since the EMc is able to supply the DEGs (DEG technology needs an external source of charging), and DEGs increase the output powers when proper electronics are utilized. In addition to ensuring the autonomy of these materials, a tunable variable stiffness of the aeroelastic system can further introduced in particular conditions of the charge process: this could also extend the operational flow range of FLEHAP, already versatile because of its peculiar mechanical configuration.
7 Perspectives

Despite the wide spectrum of the research focused on the system proposed in this thesis, some efforts still have to be attempted in different areas of the knowledge in order to completely optimize the system.

Future works include a further exploration of relevant aeroelastic parameters, with the support of experiments, phenomenological models and numerical simulations (CFD). The latter are currently in development by our collaborators of DICCA, since a clear comprehension of the mechanism involved in the system is still missing. For example, the estimation of the energy exchanges between the degree of freedom and between the fluid and the moving wing in the different LCOs could provide explanations related to the unusual effects observed during the experiments. Furthermore, the study of the effect of the electrical extraction on the evolution of the LCOs is also very important for the technological application of the device.

The study related to the synthesis and realization of DEGs could be developed: the adding of fillers as complex BaTiO$_3$ nanoparticles to the organo-modified nanoclays in the liquid silicone rubber could be explored for further increasing the performance of DE. Other important characterizations of the materials realizes should be carried out: these include aging, fatigue and the electro-mechanical response of DEGs. Also new techniques for the realization of stretchable electrodes could be prospected.

Notwithstanding a dedicated electronic for the management of the electrical charging of DEGs has been individuated for ensuring the technological applications of DEGs in FLEHAP, a self priming circuit has to be tested in the real operational conditions of the system.

Some of the prototypes realized can now be tested in real scenario applications, providing the demonstration of its use in environment operational conditions: specifically, the developmental test and evaluation (DT&E) of the system in its intended weapon system to determine if it meets design specifications.

The positioning of different devices and its effect on the EH performance has to be deepened, since preliminary experiments suggested promising results.
APPENDIX A  

*Design of a dedicated integrated circuit for EMc*

In order to solve poor adaptation of the load and internal inductance of the commercial circuits, we aimed at improving the efficiency in the energy transfer from the wind to the sensor node through a dedicated AC/DC converter. This work have been done with the collaboration of DITEN of University of Genova, and it has been published in \(^{157}\).

It mainly consists in the cascade of two buck-boost converters: one AC/DC and one DC/DC.

![Figure 103 - Details of the input buck-boost stage](image_url)

The former is equipped with a MOSFET bridge rectifier \(^{158}\) able to present approximately the absolute value of the voltage attainable at the input capacitor \(C_{IN}\). The effective value of such a voltage is determined by the impedance adaptation realized by the circuit of Fig. 103.

The proposed block diagram of the autonomous sensor node is reported in Fig.104.

![Figure 104 - Proposed block diagram of the autonomous sensor node](image_url)
Figure 105 - Results of the circuit simulation in the case of a sinusoidal input. (a) Value of the input capacitor voltage (green line) compared with the ideal half of the source voltage (violet line); (b) Supercap voltage $V_{sc}$ (red line) and its associated storage energy (blue line).

It implements the technique described in $^{157,158}$. The main clock CK is generated as a Pulse Width Modulation (PWM) signal by the MCU (a low power 8 bit microcontroller from Microchip), and drives the four low impedance switches of the converter (ADG821 from Analog Devices). In particular, one key point for the sake of efficiency is to use an ideal diode for the supercap charging process. It is realized by the parallel of a low leakage diode and one of such switches, driven by a current direction detector implemented with a low-power high-speed comparator (TLV3201 from Texas Instruments) and some logics.

Adapting the notation reported in $^{157}$, the equivalent resistance can be written as:

$$R_{EQ} = \frac{2L_H}{D^2t_{CK}}$$

being $L_H$ the inductance in the input buck-boost stage, $D$ the duty cycle, and $t_{CK}$ the clock period. Detailed circuit simulations have been performed with LTspice $^{159}$ to validate the approach.

Fig. 105 reports the case where the harvester is modelled by a sinusoidal voltage source at 10 [Hz], with a peak voltage of 3 [V], and an impedance of 300 [Ω] in series with 20 [mH]. In the top chart (Fig. 8a) the value of the voltage across $C_{IN}$ is compared with half the source voltage (which corresponds to the case of electrical maximum power transfer). In this experiment, we used $C_{IN} = 10$ [$\mu$F], $L_H = 470$ [$\mu$], $t_{CK} = 200$ [$\mu$s], and $D = 12.5$ %. It can be seen that after the very first half cycle, a good adaptation level is achieved. The values of the voltage and energy stored in the supercap are shown in Fig. 105b: the actual efficiency varies cycle by cycle with the output voltage, and reaches 80% after half a second.

The second buck-boost stage features a LTC3106 from Linear Technology, monitored by the MCU; it supplies the 3.3 [V] voltage needed by the radio link. It is worth noting that the MCU is powered by an auxiliary circuit which takes its input either from $V_{C_{IN}}$ and the output voltage.
As the optimal $R_{EQ}$ value strongly depends on the wind speed, it is necessary to employ a MPPT technique, and we choose a Hill-Climb Search Algorithm derived from $^{160}$, implemented in firmware on the MPU. Summarizing, the input voltage $V_{CIN}$ is sampled at about 1 [kHz], the extracted energy is then computed over the duration of each flattering cycle, taking into account the $R_{EQ}$ value. Then, the duty cycle $D$ and the switching frequency $f_{CK} = 1/t_{CK}$ are adapted to track the maximum power point.

The algorithm behaviour has been simulated in C language in an ideal case where the wind speed has been stepped from 4 [m/s] to 6 [m/s]. Considering a typical real case provided by the experiments, for which, as example, the maximum power available is 6.95 [mW] at 4 [m/s] and 22.27 [mW] at 6 [m/s], the tracking ability is demonstrated in Fig. 106, where the horizontal axis represents the simulation steps.

The proposed approach, based on a specifically designed, MCU driven, two-stage AC/DC converter features improved performances and maximum power point capabilities. Simulations demonstrate the possibility to reach more than 80% of the overall efficiency in a realistic case.
References


84. Roy D. Kornbluh; Ron Pelrine; Qibing Pei; Richard Heydt; Scott Stanford; Seajin Oh; Joseph Eckerle. Electroelastomers: applications of dielectric elastomer transducers for actuation, generation, and smart structures. in Smart Structures and Materials 254 (Proc. SPIE, 4698, 2012).
89. Søgaard, Emil; Taboryski, Rafael J.; Smistrup, K. Injection Molded Plastics. DTU Nanotech print (2014).


120. Rueda, M. M. *et al.* Rheology and applications of highly filled polymers: A review of


**Personal Published Scientific Publications:**


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