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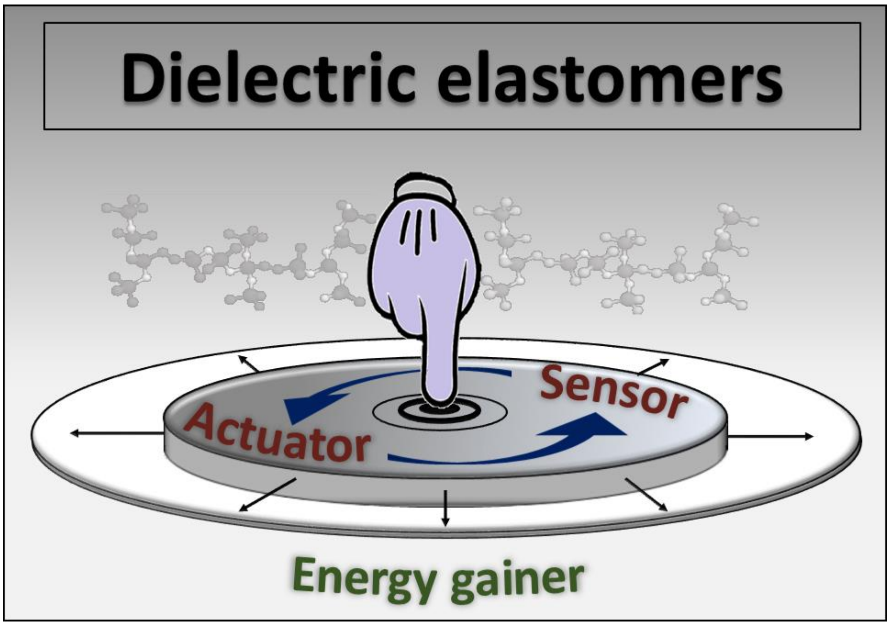
Electroactive silicone and organic-silicone networks

-PhD Thesis summary-



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Iasi - 2017

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To
Mr/Mrs____

We would like to inform you that on July 21st, 2017, at 12:00 p.m., in the library of the "Petru Poni" Institute of Macromolecular Chemistry Iasi, will be the public presentation of the doctoral thesis entitled **"Electroactive silicone and silicone-organic networks"** elaborated by **Ing. Codrin Tugui**, in order to confer the scientific title of doctor.

The doctoral commission has the following members:

PRESIDENT: **PhD Anton Airinei**, senior researcher (CSI)
Director of "Petru Poni" Institute of Macromolecular Chemistry, Iasi

SCIENTIFIC LEADER: **PhD Maria Cazacu**, senior researcher (CSI)
"Petru Poni" Institute of Macromolecular Chemistry, Iasi

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PhD. Valeria Harabagiu, senior researcher (CSI)
"Petru Poni" Institute of Macromolecular Chemistry of Iasi

Prof. PhD. Felicia Iacomi
"Alexandru Ioan Cuza" University of Iasi

In accordance with the Regulation regarding the organizing and conducting the doctorate for awarding the scientific titles within the Romanian Academy, we are sending you the summary of the doctoral thesis with the request to communicate in writing your appreciations and remarks.

On this occasion, we invite you to participate to the public defense of the doctoral thesis.

DIRECTOR,

Dr Anton Airinei

INSTITUTUL DE CHIMIE MACROMOLECULARĂ "PETRU PONI" IAȘI

Acknowledgments

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Thanks to my girlfriend Alina Buburuzanu who made my life more beautiful

I dedicate this thesis to my brother Mihai Tugui who have sustained me unconditionally regardless of the circumstances.

Content

Keywords: polymeric networks, dielectric elastomers, silicones, electroactive polymers, energy conversion.

In the early 1990s, R. Kornbluh, R. Pelrine, S. Chiba and others discovered the dielectric elastomers,[8-10] in fact developing an older concept that has been observed since 1880 by W. Röntgen.[11-13] Dielectric elastomers represents a subgroup of electroactive polymers (EAP)[14], having the ability to change their shape and/or size under the action of an external electric stimulus[15-17]. Unlike other types of existing transducers, those based on dielectric elastomers have several advantages such as greater elongations, higher energy density, or higher response speed.[18] Dielectric elastomers are used in a wide range of applications such as robotics[19-23], optical systems[24-26], sensors[27-33] or energy harvesting.[34-38]

Following the electromechanical evaluation of the available dielectric elastomers, siloxane-based polymers have been identified as the most suitable elastomers for their use as "active" elements in electromechanical transducers. Besides its outstanding properties such as high flexibility over a wide range of temperatures, stability over a wide range of frequencies and humidity, biocompatibility, silicone elastomers have some drawbacks (low dielectric permittivity, low breakdown voltage and breaking stress) that limits the conversion efficiency of the transducer. In this context, several methods have been identified regarding the improvement of the silicone elastomers features, based mainly on chemical modification (by the introduction of polar groups) or incorporation of suitable fillers. Combining the properties of silicone elastomers with the properties of other inorganic or organic networks to obtain a dielectric elastomer with cumulative properties is one of the most effective approaches but also a great challenge considering the incompatibility of silicone networks with almost any organic component.

In this sense, **the aim of the thesis** was to identify appropriate partners (inorganic and/or organic networks) and to modify and combine them with the siloxane network, in order to obtain new dielectric elastomers with superior electromechanical properties compared with existing materials.

Considering the main deficiencies of the electroactive polymers used in dielectric elastomer transducers and their applicative limitations, the doctoral thesis titled "**ELECTROACTIVE SILICONE AND ORGANIC-SILICONE NETWORKS**" had the following **objectives**:

- ✓ establishing the main factors that influence the performance of the dielectric elastomers transducers;
- ✓ Elaborating new strategies for development of new dielectric elastomers based on silicone and organic-silicone networks that exhibit superior properties as compared to those already known;
- ✓ Evaluating the prepared materials using several specific techniques to highlight the properties of interest;
- ✓ Assessing the applicability of the prepared materials by conducting breakdown, actuation and energy harvesting measurements.

As the structure, the thesis contains six chapters and an annex, with a total of 204 pages. The thesis also includes 114 figures, 30 schemes, 22 tables and 407 bibliographic references.

Chapter 1 presents the conceptual framework, purpose and objectives of the research.

In **Chapter 2** are presented the materials that can be used as dielectric elastomers and their optimization strategies.

Chapter 3 is the most comprehensive and contains personal contributions to the field of dielectric elastomers (Figure 1.1). Here are presented four strategies for optimizing the dielectric elastomers by combining the polydimethylsiloxane network with the appropriate partners (polydimethylsiloxane, poly(methyl-R)siloxane, polyurethane and polyimides) in order to obtain improved electromechanical properties. Also in this chapter are presented two strategies for obtaining compliant electrodes and their testing in actuation and energy harvesting systems. There are also presented three setups designed in laboratory for electromechanical testing of dielectric elastomers and electrodes.

Chapter 4 presents in detail the materials and techniques used in this study.

Chapter 5 summarizes the general conclusions, the originality of the thesis and the outlooks for further research.

In Chapter 6 are presented the bibliography of the thesis.

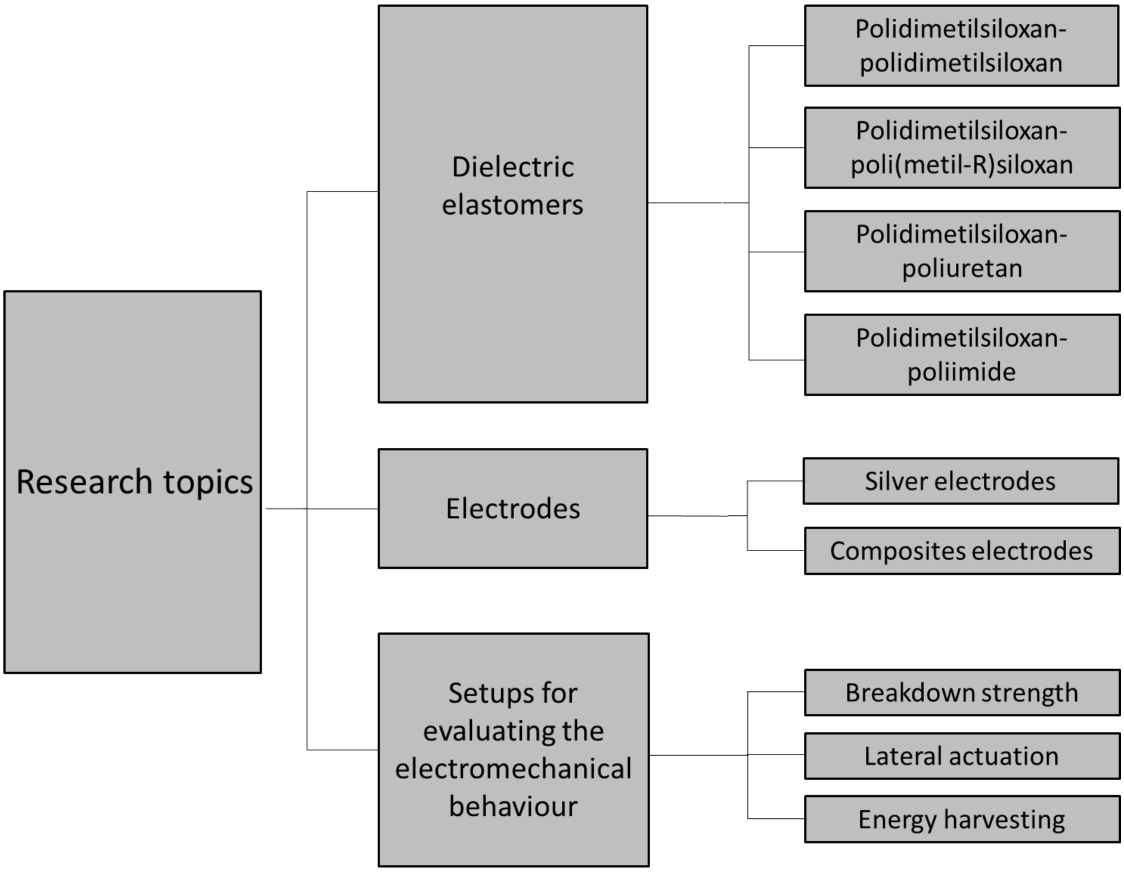


Figure 1.1. The main issues addressed in the thesis.

In order to improve the electromechanical performances of the dielectric elastomers, were prepared a series of bimodal polydimethylsiloxane-polydimethylsiloxane networks using a procedure reported by Q. Pei et al. The electrotechnical improvements were obtained by blocking the elastomeric films in a pre-stretched state.[304]

The preparation of bimodal networks was carried out sequentially, in two stages, the crosslinking of polymers took place trough different mechanisms, first the condensation reaction of polydimethylsiloxane-α,ω-diol (PDMS) with tetraethylorthosilicate (TEOS) in the presence of dibutyltin dilaurate (DBTDL) at room temperature (Net1), followed by the hydrosilylation reaction of α,ω-bis(vinyl)polydimethylsiloxane with α,ω-bis(trimethylsiloxy)poly(dimethylmethyl-H-siloxane) at elevated temperature in the presence of Speier catalyst, Net2 (Figure 3.2.2).

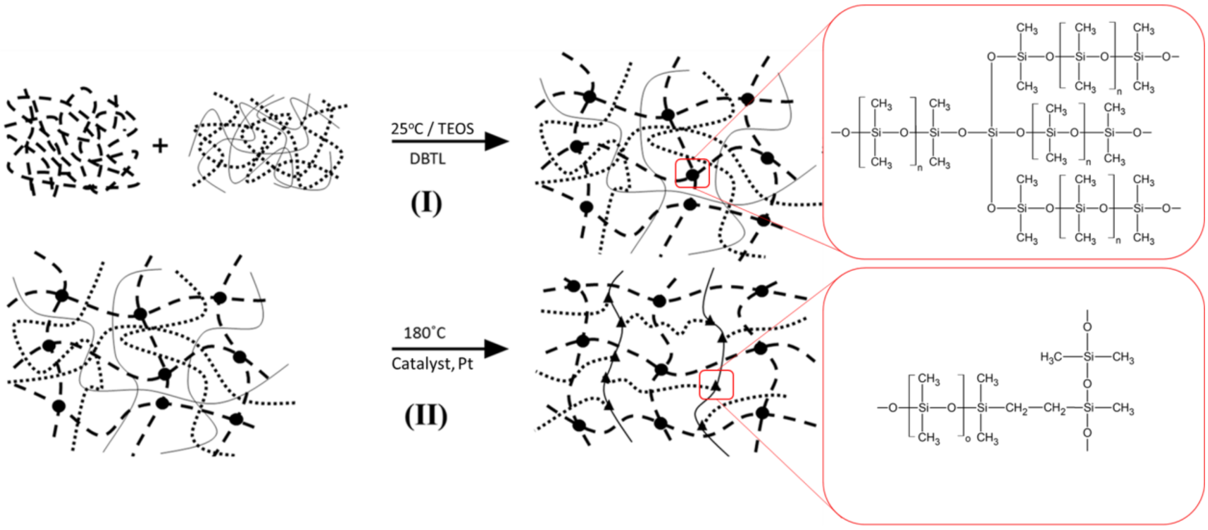


Figure 3.2.2. Sequential preparation method.

The pre-stretched films show higher breakdown strength values than unstretched ones (Table 3.1.4, Figure 3.1.9). Moreover, the breakdown voltage increases with the increasing amount of Net2. On the other side, the actuation strain in lateral direction (Sx), increases also with the increasing Net2 content. Similar behaviour was observed in the case of pre-stretched samples, but in this case the actuation values are slightly lower compared to those unstretched.

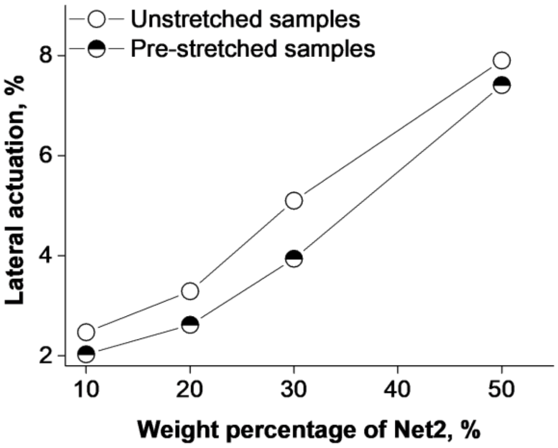


Figure 3.1.11. Lateral actuation strain of prepared samples (determined at an electric field E=40 V·µm^{-1}) depending on the Net2 content.

Like the bimodal network preparation method, a series of polydimethylsiloxane-poly(methyl-R-siloxane) (R-polar groups) were prepared to improve the dielectric permittivity by attaching different polar groups (phenyl, trifluoropropyl, 3-cyanopropyl) to the silicon atoms. For this, were used two chemical paths: condensing reaction of -OH groups from the ends of the siloxane copolymers chains containing different percentages of polar groups along the backbone (poly(dimethylsiloxane-co-diphenylsiloxane)-α,ω-diol or poly(dimethylsiloxane-co-methyl(3,3,3-trifluoropropyl)siloxane]-α,ω-diol), and an addition reaction of α,ω- bis(trimethylsiloxyl)poly(dimethylsiloxane-co-methylhydrosiloxane) or α,ω-bis (trimethylsiloxi)poly(methylcyanopropylsiloxane-co-methylhexylsiloxane-co-methylhydrosiloxane) to α,ω-bis(vinyl)polydimethylsiloxane. This time the obtained materials were processed without pre-stretched.[303] The breakdown field values reveal a clear decrease with the introduction of polar content. The decreasing in breakdown field value as the polarity of the material increases is an expected, well-known behaviour due mainly to large dielectric losses. Using the linear-elasticity and boundary approximations, valid for small strains (<10%), the lateral actuation strain $S_{x,}$ was estimated.[305] The calculated lateral actuation strain values are in concordance with both electrostatic pressure and electromechanical sensitivity values, the highest value being recorded for the sample containing cyanopropyl groups, IPN-CN1 (Figure 3.2.15, Table 3.2. 6).

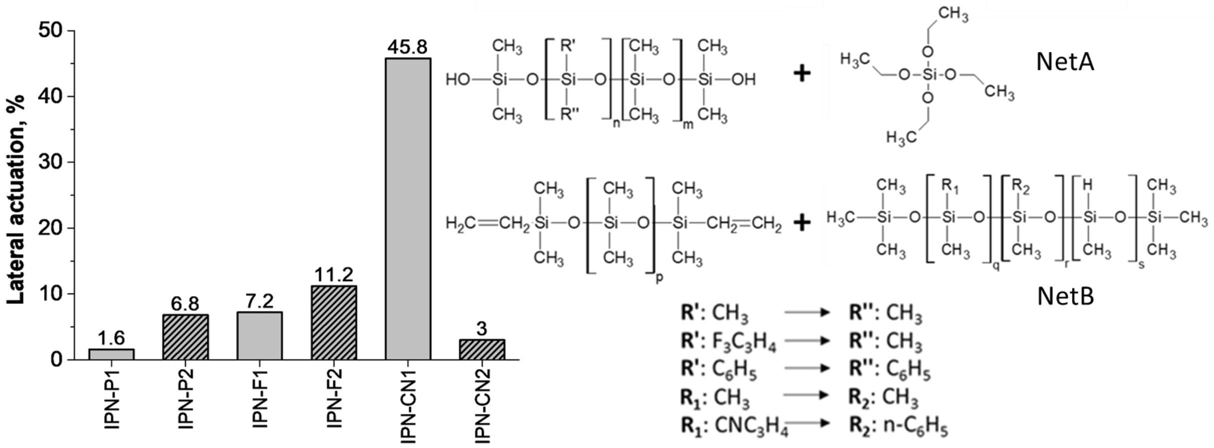


Figure 3.2.15. Comparative lateral actuation strain values estimated theoretically for prepared IPNs having different polar content.

Polyurethanes have high dielectric permittivity, which is based on the polar nature of the polyurethane fragments (roughly 7, compared to 3 for silicones[143]) allowing them to be actuated at lower electric fields but cannot develop large strains.[144] However, low concentration of chemical cross-links in the thermoplastic polyurethane (TPU), as well as in the acrylate, results in a high level of creep or permanent deformation of these materials under actuation strain.[143] Taking into account these aspects, three types of semi-interpenetrated (S-IPN) polydimethylsiloxane-polyurethane networks differing by the molecular weight of the polysiloxanes (X_{Mn}=70000 g·mol^{-1}; Y_{Mn}=270000 g·mol^{-1}; Z_{Mn}=370000 g·mol^{-1}) were prepared, in order to combine the properties of polyurethane with those of PDMS in a monolithic material that offers superior electromechanical properties (Figure 3.3.3).[305] For this, a polyurethane (PUUS) containing tetramethyldisiloxane fragments in the backbone was prepared, siloxane fragments favouring the compatibility with the siloxane network.

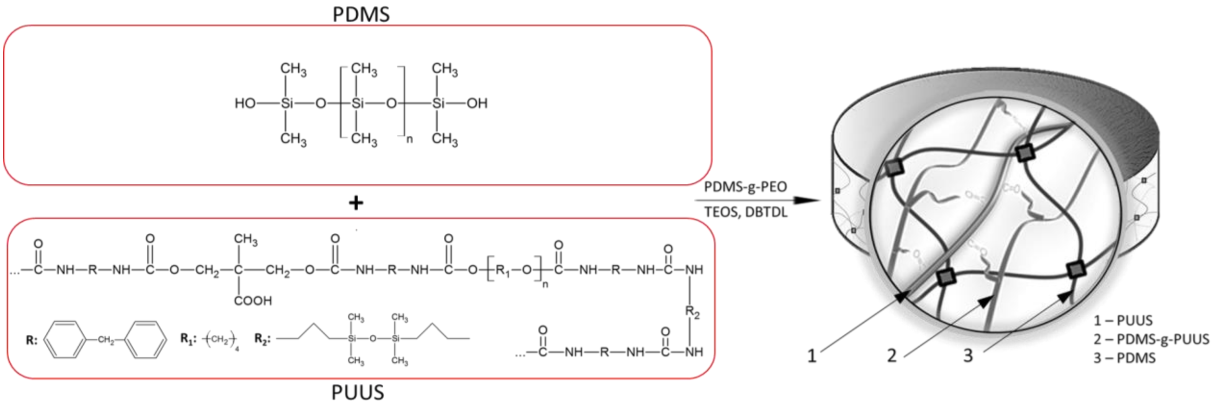


Figure 3.3.3. Graphic representation of S-IPN polysiloxane-polyurethane formation networks.

As expected, the breakdown field values decrease with increasing molecular weight of PDMS and the content of PUUS. PUUS has the lowest value for the electrical breakdown field, Ebd, as compared with the other dielectric elastomers and the three PDMSs used as partners for S-IPNs. (Figure 3.3.13, Table 3.3.5). The lateral actuation strain values (Sx) are shown in Figure 3.3.16, where the actuation keeps the same trend in all three series. It can be seen that this parameter increases from the series X to series Z due to increasing siloxane chain length between the crosslinking nodes and, within any given series, it increases up to an addition of 10 % PUUS, the highest values naturally are obtained at higher applied voltages. The samples containing 20 wt % PUUS show poor actuation strain due to the high surface porosity that limits the compliance between electrode and dielectric. In general, the actuation values for the prepared samples are higher than those obtained for the three commercially available dielectric elastomers (Table 3.3.6).

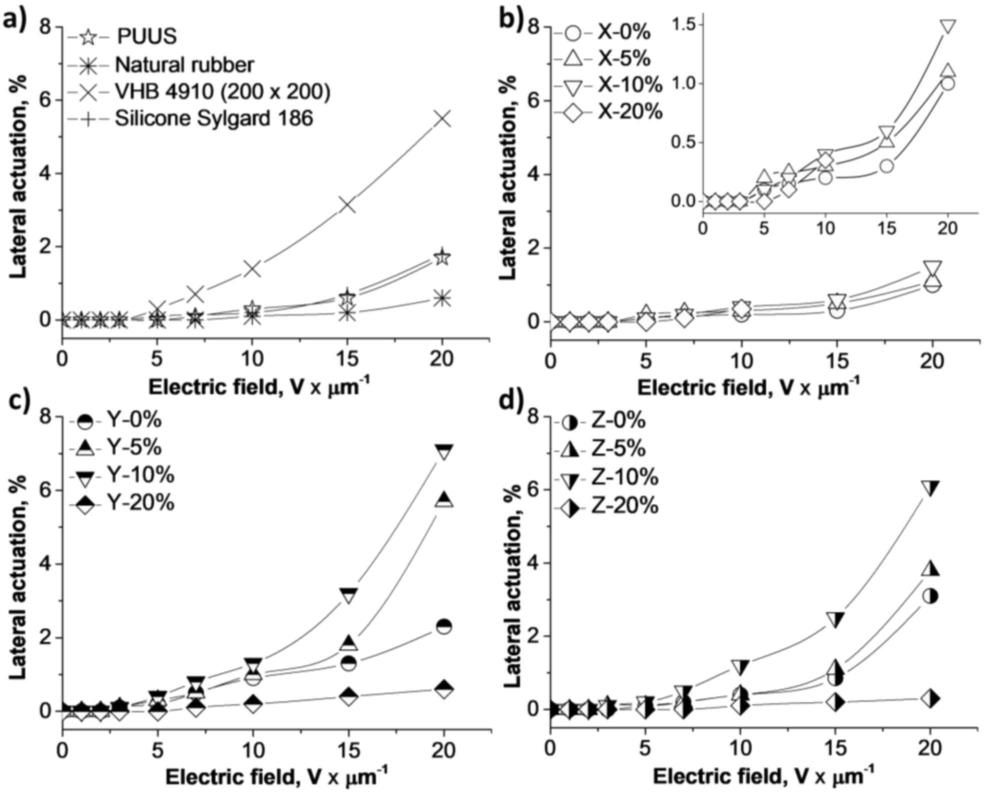


Figure 3.3.16. Lateral strain dependent on an applied electric field for: **a)** pure PUUS film, natural rubber, VHB 4910 (200% equibiaxially prestrained) and Sylgard 186; **b)**, **c)** and **d)** prepared PDMS-PUUS S-IPN films.

Due to their excellent thermal, mechanical and dielectric properties, aromatic polyimides attract particular interest in the development of high temperature piezoelectric and pyroelectric sensors and microelectromechanical devices (MEMS).[368,369] Their investigation was mainly motivated by NASA's interest for developing high performance piezoelectric polymers for a variety of aerospace applications..[370] Considering these perspectives, polydimethylsiloxane-polyimide networks were prepared in order to combine the properties of polydimethylsiloxane with those of polyimide (PI) in a material capable of generating both electrostatic and piezoelectric response (Figure 3.4.7).[306] There are few examples of PDMS–PI networks reported in the literature, but none of them addresses these materials from this perspective.[376,377]

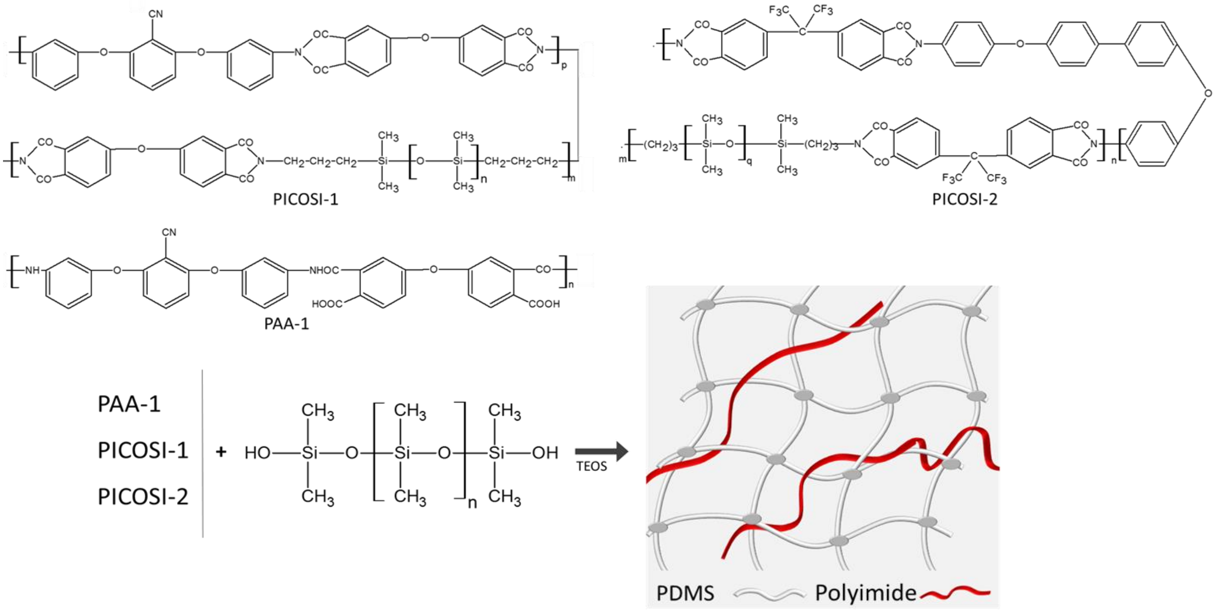


Figure 3.4.7. Graphic representation of S-IPN polysiloxane-polyimide networks formation.

The breakdown strength values of S-IPNs based on PDMS–PAA-1 decrease with the increasing amount of polyamic acid. By contrast, the electrical breakdown of S-IPN series based on PDMS–PICOSI-1 and PDMS–PICOSI-2 increases with the amount of polyimide (Figure 3.4.18, Table 3.4.3). Furthermore, the breakdown values of these S-IPNs are higher than of PDMS network and polyimide, suggesting that there is a synergistic effect between the IPN components, as was also found in the case of thermal behaviour.

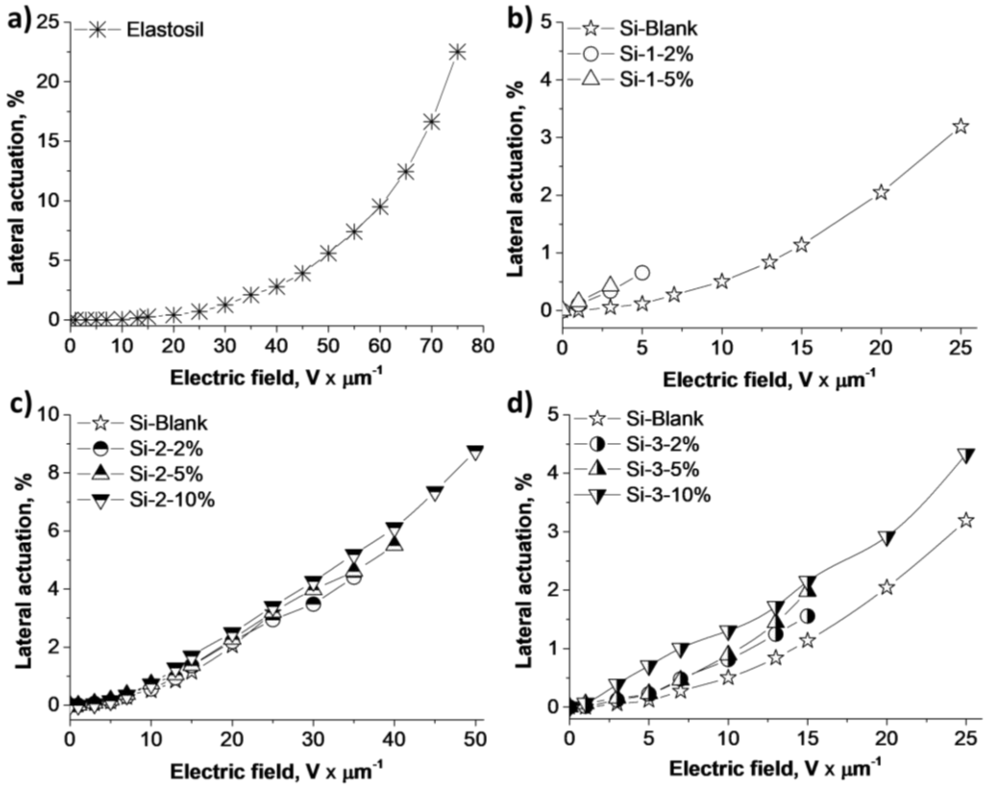
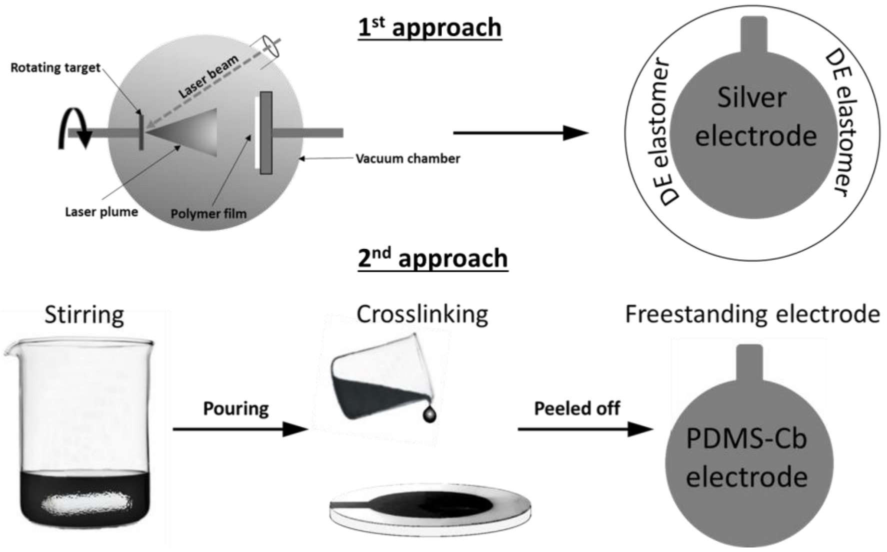


Figure 3.4.20. Lateral actuation strain of: **a)** Elastosil; **b)** PAA-1 series; **c)** PICOSI-1 series and **d)** PICOSI-2 series.

The commercial film Elastosil has the highest lateral strain, of about 22 %, but a very high electric field, nearly 80 $V\cdot\mu m^{-1}$, was required to reach this value (Figure 3.4.20a). Instead, although the actuation strain values of siloxane–polyimide semiinterpenetrating networks are lower, the electric field required to be actuated is much lower (Figure 3.4.20c and d). For instance, at an electric field of 15 $V\cdot\mu m^{-1}$, the lateral strain of the commercial film Elastosil is 0,3 %, while for all S-IPNs the lateral strain is higher than 1 % and even higher than 2 % for sample Si-3-10% (Table 3.4.3). Moreover, samples Si-2-10% and Si-3-10% having the highest polyimide content (10 wt%) were evaluated by piezoelectric force microscopy (PFM) measurements and compared with pure crosslinked silicone sample Si-Blank used as a reference (Figure 3.4.22). In the case of pure crosslinked silicone, no noticeable piezoresponse and phase contrast were found, while the magnitude and phase images of polysiloxane–polyimide films show a variety of contrasts implying widespread local d_{33} values and allow identifying structure domains with different orientations of polarization. The obtained values are lower than those reported for a highly pure piezoelectric such as $PbZr_xTi_{1-x}O_3$ (PZT) (360 to 500 $pm\cdot V^{-1}$), [395] similar to a PC/PVDF/PZT (portland cement/polyvinylidene fluoride/lead zirconate titanate) composite (up to 24 $pm\cdot V^{-1}$) [396] and much larger (about 7,5 times) than values found in the case of Parylene C films.[397]

Regardless of the DET's applications, a conductive and flexible electrode that survives at a large number of deformations is required. In this regard and taking into consideration the economic aspects, two types of electrodes were achieved, a highly conductive silver electrode and a highly stretchable freestanding PDMS-carbon black (PDMS-Cb) electrode using a versatile physical method and a straightforward chemical method (Scheme 3.5.1). In the first approach, a silver electrode of about 40 nm thickness was deposited directly on the dielectric surface by pulsed laser deposition (PLD). The PLD technique being one of the cheapest, simplest and most versatile method of depositing thin films. [406] The second strategy was focused to obtain flexible and soft freestanding electrodes based on PDMS-carbon black (PDMS-Cb), the carbon black powder being the cheapest conductive powder on the market.



Scheme 3.5.1. Schematic representation of the path of silver electrodes deposition (top), and for obtaining freestanding electrodes (bottom).

Both types of electrodes were tested from electrical point of view. The electrical conductivity of silver electrode decreases dramatically with the linear strain while the freestanding PDMS-Cb electrodes exhibit excellent electrical properties, the electrical conductivity being unaffected by the uniaxial strain (Figure 3.5.4). The PDMS-Cb electrodes were evaluated as electrode for dielectric elastomer generators (DEG). Depending on the number of DEG active layers, the U_{output}/U_{input} ratio increases as the inflating strain increase (Figura 3.5.14)

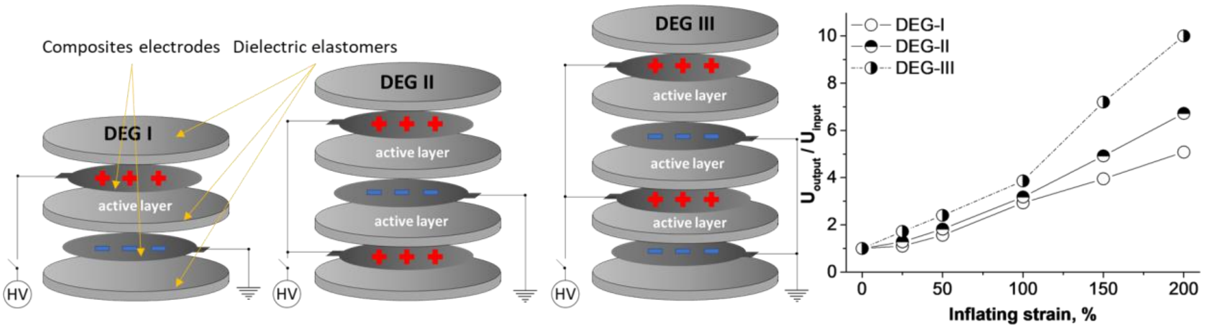


Figure 3.5.14. Measured U_{output}/U_{input} ratio as a function of inflating strain, U_{input}=100V.

The electromechanical evaluation represents the most important aspect that contributes to the identification and quantification of new materials suitable for electromechanical applications. In this regard, three types of installations were designed and built (electrical breakdown, lateral actuation and energy harvesting setup). All three installations were designed in accordance with the recent recommendations of European Society for Electromechanically Active Polymer, EuroEAP.[367]

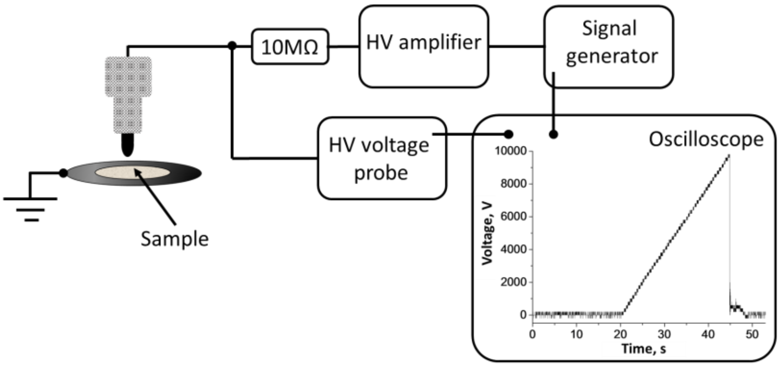


Figure 3.7.2. Electrical breakdown measurement setup.

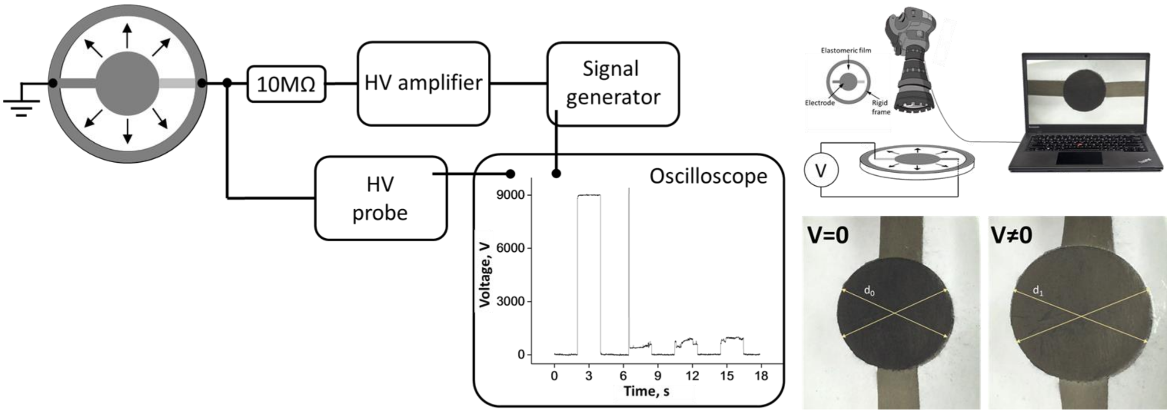


Figure 3.7.3. Schematic representation of lateral actuation setup.

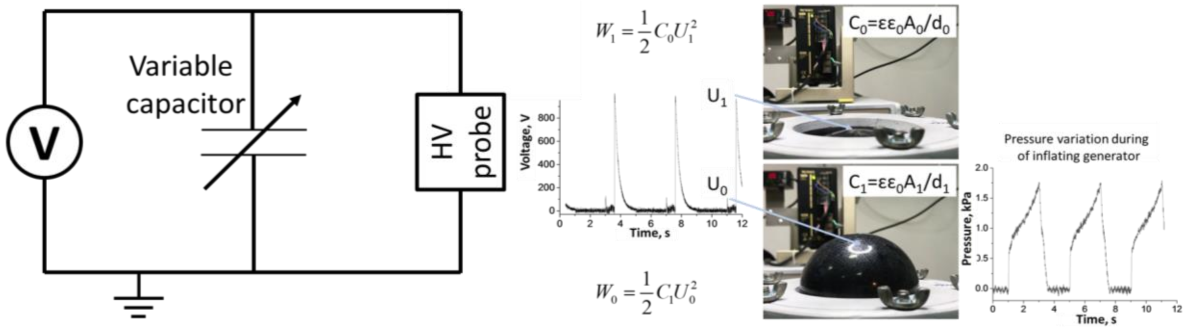


Figure 3.7.5. Energy harvesting evaluation setup.

General conclusions

Based on the literature review, the main requirements of dielectric elastomer were established and thus the most suitable dielectric elastomers and the appropriate optimization method have been identified.

Four types of IPN/S-IPN networks were prepared based on high molecular weight polydimethylsiloxane crosslinked through a condensation mechanism used as the main component and:

- lower molecular weight polydimethylsiloxane crosslinked through a hydrosilylation mechanics (bimodal networks);
- polysiloxane with polar groups attached to the silicon atom (polydimethylsiloxane-poly(methyl-R)siloxane networks, where R represents the polar group);
- polyurethane (polysiloxane-polyurethane networks);
- polyimide (polysiloxane-polyimide networks).

Depending on the functional groups, various crosslinking mechanisms have been used, mainly consisting of condensation and hydrosilylation reactions in the case of silicones and physical crosslinks in the case of organic components.

All prepared materials were processed as freestanding thin films with uniform thickness.

All IPN/S-IPN films were investigated from the mechanical (elongation at break, Young's modulus and plastic deformation), dielectric (dielectric permittivity and dielectric loss) and electromechanical points of view by measuring the breakdown voltage and lateral actuation strain (excepting polydimethylsiloxane-poly(methyl-R)siloxane networks where the lateral actuation strain was estimated theoretically).

✓ Bimodal polydimethylsiloxane-polydimethylsiloxane networks were prepared and it was revealed that the electromechanical properties of the achieved IPNs depend on both the network composition and the pre-stretched or unstretched state. Due to the higher crosslinking density, the network obtained by hydrosilylation reaction induces an increasing of the Young's modulus and a decreasing of elongation at break of IPNs. On the other hand, the rigid network leads to an improvement of both breakdown strength and actuation strain. IPNs networks based on polydimethylsiloxane-poly(methyl-R)siloxane having different polar groups (trifluoropropyl, phenyl or 3-cyanopropyl) along the siloxane chain were prepared, in order to increase the dielectric permittivity and other electromechanical parameters. All the prepared films showed a low mechanical hysteresis. The dielectric results point out that the dielectric permittivity shows a significant increase only in the low frequency range where the dielectric losses are also high as compared with the blank samples but decreasing as frequency increases. At higher frequencies, the effect of the polar groups on the dielectric permittivity is less spectacular. The breakdown field values reveal a clear decrease with the introduction of polar groups and their content, the most significant decreasing being recorded in the case of samples that contain phenyl and trifluoropropyl groups. However, the lateral actuation strain estimated theoretically reveals a very good electromechanical behavior, especially for IPNs networks that contain trifluoropropyl and cyanopropyl groups.

✓ Polysiloxane-polyurethane S-IPN networks designed to improve the electromechanical properties by introducing a polar component, in this case polyurethane, and by ranging the molecular weight of the polysiloxane were prepared. The tensile test results revealed that, in all cases, with the increasing amount of polyurethane, the films become stiffer. The plastic deformation also increases with increasing amount of polyurethane (which itself has the highest plastic deformation) and also with the PDMS molecular weight, as cyclic stress–strain tests showed. Increasing the polyurethane content within S-IPN networks leads to an increasing of dielectric permittivity and a decreasing of breakdown strength. The actuation measurements revealed that S-IPN networks shows higher actuation strain than commercial elastomers. Moreover, the actuation strain values determined experimentally are in concordance with those calculated theoretically.

✓ Polysiloxane-polyimide networks designed to obtain a multifunctional material able to operate as both actuator and sensor were prepared. The mechanical results, showed that the imidic chains increase the Young's modulus of S-IPNs. The breakdown values of the S-IPNs are much higher than both the PDMS network and polyimides, suggesting that there is a synergetic effect between the S-IPN components, as was also found in the case of thermal behaviour. Besides the large deformation, up to 8.7 %, mainly given by a flexible dielectric silicone component, the presence of the imide component confers the piezoelectric effect, as has been revealed and measured by piezoelectric force microscopy, the average values obtained (17÷26 pm·V^{-1}) being of high interest.

✓ Two types of electrodes were obtained, a very conductive silver electrode and a highly elastic PDMS-carbon black electrode. Both types of electrodes were used in a “sandwich” configuration with a common commercial dielectric elastomer (Elastosil) in order to evaluate their efficiency. The electrical measurements revealed that the electrical conductivity of the metallic electrode decreases significantly with elongation, while the electrical conductivity of PDMS-Cb electrodes remained almost unchanged up to 300 % elongation. Lateral strain values achieved as a result of applied electric field showed a large difference between the two electrodes, the maximum lateral strain of PDMS-Cb being several times larger than in the case of using silver electrode. Furthermore, by using a simple and efficient method, three "sandwiches" made from PDMS-Cb electrodes and commercial elastomer (Elastosil) were built and tested in an energy harvesting setup. The obtained results showed that the achieved electrodes work perfectly under large strains and after hundreds of repeated harvesting cycles.

✓ Three laboratory facilities were designed and built to test the electrical breakdown, lateral actuation strain and energy harvesting capabilities. The three installations allow a complete electromechanical evaluation of dielectric elastomers.

Scientific activity

The scientific contributions from this thesis are contained in five scientific articles, four of which were published and one under review, and nine presentations (five oral and four posters) at different scientific manifestations.

I) Works published in ISI-rated journals reported to the PhD thesis:

1) **C. Tugui**, M. Cazacu, L. Sacarescu, A. Bele, G. Stiubianu, C. Ursu, C. Racles; Full silicone interpenetrating bi-networks with different organic groups attached to the silicon atoms, Polymer **2015**, 77, 312-322 **(FI: 3.684; SRI: 2.527)**;

2) **C. Tugui**, G. Stiubianu, M. Iacob, C. Ursu, A. Bele, S. Vlad, M. Cazacu; Bimodal silicone interpenetrating networks sequentially built as electroactive dielectric elastomers, J. Mater. Chem. C **2015**, 3, 8963- 8969 **(IF: 5.256; SRI: 2.756)**;

3) **C. Tugui**, S. Vlad, M. Iacob, C. D. Varganici, L. Pricop and M. Cazacu; Interpenetrating poly(urethane-urea)–polydimethylsiloxane networks designed as active elements in electromechanical transducers, Polym. Chem. **2016**, 7, 2709-2719 **(IF: 5.375; SRI: 3.775)**;

4) **C. Tugui**, A. Bele, V. Tiron, E. Hamciuc, C. D. Varganici, M Cazacu; Dielectric elastomers with dual piezo-electrostatic response optimized through chemical design for electromechanical transducers, J. Mater. Chem. C **2017**, 5, 824-834 **(IF: 5.256; SRI: 2.756) selected as HOT Paper**;

5) **C. Tugui**, C. Ursu, L. Sacarescu, M. Asandulesa, G. Stoian, G. Ababei, M. Cazacu, Highly stretchable silicone nanocomposite vs. highly conductive silver thin layer as electrodes for dielectric elastomers transducers, "***under review***".

IF total = 19,571 / RIS total = 11,814

II) Works published in ISI-rated journals related to the subject of the PhD thesis:

1) M. Iacob, G. Stiubianu, **C. Tugui**, L. Ursu, M. Ignat, C. Turta, M. Cazacu; Goethite nanorods as a cheap and effective filler for siloxane nanocomposite elastomers RSC Adv. **2015**, 5, 45439-45445 **(IF: 3.108; SRI: 1.826)**;

2) Carmen Racles, V. E. Musteata, A. Bele, M. Dascalu, **C. Tugui**, A-L Matriarcala; Highly stretchable composites from PDSM and polyazomethine fine particles, RCS Adv. **2015**, 5, 102599-102609 **(IF: 3.108; SRI: 1.826)**;

3) G. Stiubianu, A.M.C. Dumitriu, C.D. Varganici, **C. Tugui**, M. Iacob, A. Bele, M. Cazacu; Changes induced in the properties of dielectric silicone elastomers by the incorporation of transition metal complexes, High Perform. Polym. **2016**, 28, 8, 915-926 (**IF: 1.142; SRI:0.629**);

4) A. Bele, M. Dascalu, **C. Tugui**, M. Iacob, C. Racles, L. Sacarescu, M. Cazacu; Dielectric silicone elastomers filled with in situ generated polar silsesquioxanes: Preparation, characterization and evaluation of electromechanical performance, Mater. Design. **2016**, 106, 454-462 **(IF: 4.364; SRI: 2.065)**;

5) G. Ştiubianu, A. Soroceanu, C-D. Varganici, **C. Tugui**, M. Cazacu; Dielectric elastomers based on silicones filled with transitional metal complexes, Composites Part B Engineering **2016**, 93, 236-243 **(IF: 4.727; SRI: 3.229)**;

6) A. Bele, G. Stiubianu, S. Vlad, **C. Tugui**, C.D. Varganici, L. Matricala, D. Ionita, D. Timpu, M. Cazacu; Aging behavior of the silicone dielectric elastomers in simulated marine environment, RSC Adv. **2016**, 6, 8941-8955 **(IF: 3.108; SRI: 1.826)**.

7) C. Racles, M. Dascalu, A. Bele, V. Tiron, M. Asandulesa, C. Tugui, A. L. Vasiliu, M. Cazacu; All-silicone elastic composites with counter-intuitive piezoelectric response, design for electromechanical applications, J. Mater. Chem. **2017**, doi: 10.1039/C7TC02201H **(IF: 5.256; SRI: 2.756)**.

IF total = 24,813 / RIS total = 14,157

III) Oral communications at national/international events

1) **Codrin Tugui**, Cristian Ursu, Magdalena Aflori, Maria Cazacu; Metallic compliant electrodes for dielectric silicone elastomers, EAP Workshop **2014**, Electromechanically Active Polymer (EAP) transducers & artificial muscle, Queen Mary University of London, 25-26 November, London, Uk;

2) **Codrin Tugui**, Cristian Ursu, Magdalena Aflori, George Stiubianu, Mihail Iacob, Adrian Bele, Xenia Patras, Maria Cazacu; Dielectric elastomers transducers as medical devices, Congres International "Pregatim Viitorul Promovand Excelenta", editia XXV-a, sectiunea B – Repere in medicina moderna, 26 februarie - 1 martie **2015**, Universitatea Apolonia, Iasi, Romania;

3) **Codrin Tugui**, George Stiubianu, Mihail Iacob, Cristian Ursu, Maria Cazacu; Elastomeri dielectrici pe baza de retele siliconice bimodale, A XXV-a Sesiune de Comunicari Stiintifice a Institutului de Chimie Macromoleculara „Petru Poni" (Zilele Academice Iesene), 24-26 septembrie **2015**, Iasi, Romania;

4) **Codrin Tugui**; Elastomeri dielectrici cu aplicatii in conversia de energie, Conferinta Stiinta Moderna si Energia, editia XXXIII-a, 14-15 Mai **2015**, Cluj-Napoca, Romania;

5) **Codrin Tugui**, Adrian Bele, Stelian Vlad, Carmen Racles, Maria Cazacu; Dielectric elastomers optimized through interpenetration strategies, Eighth Cristofor I. Simionescu Symposium – Frontiers in Macromolecular and Supramolecular Science, 29 May – 3 June **2016**, Iasi, Romania.

IV) Posters at national/international events

1) **Codrin Tugui**, Maria Cazacu, Stelian Vlad, Cristian Ursu; Full silicone interpenetrating bi-networks with different organic groups to the silicon atoms, EuroEAP 2014: 4th International conference on Electromechanically Active Polymer (EAP) transducers & artificial muscles, 10-11 June **2014**, Linköping, Sweden;

2) **Codrin Tugui**, George Stiubianu, Mihail Iacob, Cristian Ursu, Cristian-Dragos Varganici, Maria Cazacu; Bimodal silicone networks as dielectric elastomers, EuroEAP

2015: 5th International conference on Electromechanically Active Polymer (EAP) transducers & artificial muscles, 9-10 June **2015**, Tallinn, Estonia;

3) **Codrin Tugui**, Cristian Ursu, Adrian Bele, Mihail Iacob, Maria Cazacu; Ultra-thin stretchable silver electrodes as compared with PDMS-based electrodes designed for energy harvesting; EuroEAP 2016: 6th International conference on Electromechanically Active Polymer (EAP) transducers & artificial muscles, 14-15 June **2016**, Helsingor, Danemark;

4) **Codrin Tugui**, Maria Cazacu; Comparative approaches to high performance stretchable electrodes, EuroEAP 2017: 7th International conference on Electromechanically Active Polymer (EAP) transducers & artificial muscles, 6-7 June **2017**, Cartagena, Spain.

V) Member of research projects

1) Project 7th Framework Programme FET (www.polywec.org, prj. ref. 309139) and co-financed by CNCS- UEFISCDI (Contract 205EU); New mechanisms and concepts for exploiting electroactive Polymers for Wave Energy Conversion, Coordinator: Scuola Superiore Sant'Anna, January 2014 – December 2016.

2) Project PN-III-P2-2.1-PED-2016-0188; Silicon-based conversion units obtained by "green" chemistry, Coordinator: "Petru Poni" Institute of Macromolecular Chemistry, January 2017 – present.

VI) Internship abroad

1) COST Action, ESNAM Training School on Dielectric Elastomers, 25-27 March 2014, Darmstadt, Germany;

2) COST Action, European Network on smart inorganic polymers, COST-TS-ECOST-TRAINING SCHOOL CM1302-250914-049000, 25-30 September 2014, Graz - Austria, Trieste - Italy.

VII) Other activity

1) Dissemination & outreach committee member of European Society for Electromechanically Active Polymer, EuroEAP.

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