Pre-strain in Dielectric Elastomer Actuator; Challenges towards Structure-Property Relationship

Dhananjay Sahu1, Om Prakash Prabhakar1, Raj Kumar Sahu1\*, Karali Patra2

*1Department of Mechanical Engineering, National Institute of Technology, Raipur, India.*

*2Department of Mechanical Engineering, Indian Institute of Technology, Patna, India.*

\*Corresponding author Email: raj.mit.mech@gmail.com

------------------------------------------------------------------------------------------------------------------

Pre-strain is one of the most preferred techniques to enhance electromechanical performance of dielectric elastomer actuators (DEA). However perfect strain regime and deformation conditions toward efficient mechanical output are yet to be recognized and the relation with micro/macromolecular parameter perceive relevant attention. This article collects recent studies on the influences of pre-strain on electroactive behaviour of dielectric elastomer. Strain regulated electromechanical properties are discussed in respect to modification in micro/ macromolecular structures. Molecular modification due to strain hardening in biaxially pre-strained VHB 4910 dielectric elastomer is evidenced. Hardening may lead crystallization which is a probable reason that control dielectric and elastic characteristics. The results address well the significance of requisite development towards structural-property relation, as a promising understating to improve actuation performance of elastomer based soft actuator.

***Keywords*:** strain-inducedcrystallization, micro/ macromolecular, VHB 4910, soft actuators

1. **Introduction**

Dielectric elastomers belong to the family of electroactive polymers and are recognized as futuristic material towards design and development of soft actuator of multidisciplinary applications. Dielectric elastomers are found as promising material owing to durable softness, high flexibility, ease in fabrication, high power to weight ratio, self-healing competence, biocompatibility, etc.[1]–[4]. Subsequently, numerous actuator configurations are proposed to be utilized as sensors, artificial muscles, tunable lenses, energy harvesting micro-electromechanical devices, etc. [5][6].

The basic configuration of dielectric elastomer actuators (DEAs) comprises a pre-strained elastomer film that sandwiched in a rigid frame and surfaces coated with compliant electrode. When a high voltage is applied across the electrodes, opposite charges attract each other results in elastic deformation known to be actuation strain [7]. The magnitude of actuation strain is dependent on electrostatic pressure owing to the principle of Maxwell stress as represented in eq.1 and 2. As revealed from eq.1, the electrostatic pressure can be improved by increasing dielectric permittivity and decreasing film thickness, while eq.2 depict the significance of low elastic modulus to achieve high actuation strain [8]. Further, it is noteworthy that the applied pre-strain, as utilized in the fabrication of actuator configuration, decreases dielectric permittivity due to the disturbed distribution of dipoles in expanded surfaces. On the other hand, decreases in elastomer film thickness lower down the voltage required to induce actuation strain. The strain-induced variation in electromechanical properties are well recognized to depends on pre-strain regimes such as uniaxial, equi/unequal-biaxial, pure deformation, radial deformation, etc. and the deformation parameter that includes deformation rate, stretch ratio, type of loading, etc. [9]. Moreover, it also depend on the type of material like silicon, rubber, and acrylic [10]. Therefore, depending on the potentiality of material for distinct application enormous investigation are devoted to characterize electromechanical properties of commercially available elastomers.

(1)

(2)

*Where, p is electrostatic stress, V is applied voltage, is dielectric permittivity of elastomer, is permittivity of free space, E is the electric field (volts per meter), d is film thickness, is field induced actuation strain in thickness direction, and Y is elastic modulus.*

Extensive literature study revealed that varieties of silicon based elastomers from different manufacturers and acrylic VHB tape from 3M Technology are frequently investigated material with reliable performance [11]. Thus, the adequate information on their characteristics for actuator design commences them as feasible materials in fascinating technology of soft electromechanical devices. In preliminary studies dielectric permittivity of VHB 4910 elastomer is observed to decrease from 4.4 to 2.4 with increasing equi-biaxial strain nearer to stretch ratio 5. This linear decrement is attributed to strain-induced chain entanglement, that decreases the polarizing competence of polar charge [12]. Similarly, pre-strain also influences the mechanical characteristics that include energy density, creep, hysteresis, etc., significant to achieve efficient actuation strain [13]–[15]. Although instance, the electromechanical performance is found independent of strain regimes (biaxial, uniaxial, pure shear etc.), and are related to final thickness obtain through pre-strain [9]. Further it is noteworthy that the effect of pre-strain is more prominent in acrylic VHB than the silicon based elastomers which depict the performance dependency on material content. Hence considering the fact that intrinsic structure plays an essential role in material properties [16], it is of utmost importance that effects of strain on basic structure, hence on performance of dielectric elastomer should be understood in a newer way.

The electromechanical performance of dielectric elastomer based devices significantly depend on the dielectric characteristics as dielectric constant, loss tangent, electric breakdown strength, etc. and the mechanical properties that include elasticity, energy dissipation due to viscoelastic behaviour, tensile strength, and durability. Also, the electromechanical properties are known to depends macromolecular parameters like types of monomers, covalent bond, molecular weight, crosslink density other than physical parameters that includes dimensions, types of compliant electrode, applied voltage, environmental conditions, etc. Unfortunately, the correlation between structures and properties of dielectric elastomer is limited in available literature and need significant attention of the research community.

This article highlights the overall understanding towards pre-strain-induced structural changes responsible for properties enhancement in dielectric elastomer. Initiated with outline of the effect of pre-strain on electromechanical properties and its effectiveness in actuation performance. The respective change in properties are discussed in terms of the modification in micro/macromolecular structure. A comparative infrared absorbance analysis of VHB elastomer is accomplish to understand molecular modification behind strain hardening considerate. The evidence to strain induced modification in structure with its consequences in material characteristics is small but significant contribution towards the state-of-art of structure-property relation in dielectric elastomers.

1. **How pre-strain regulate electromechanical properties of dielectric elastomer**

A simple and easy way to understand how applied pre-strain efficiently regulate the electromechanical properties and so the electromechanical performance of dielectric elastomer is to identify the molecular and macromolecular parameters that respectively embed the elasticity and the dielectric characteristics. As in macromolecular parameters, a long chain and low crosslink density structure of silicon elastomer is shown to acquire low elastic modulus, while elastomer with low molecular weight is found to have high permittivity [17]. Apart, consideration of molecular weight is an important parameter for analysing strain-induced crystallization in natural rubber [18]. Recently, self-healing competence of silicon dielectric elastomers shown to depend on dynamic non-covalent bonds [19][20]. Consequently, pre-strain influenced properties alteration in dielectric elastomer is expected as a result of modification in macromolecular parameter that includes chemical structure, material composition, lattice structure, molecular weight, crosslink density, etc.

For instance, attribution to the strain effects on permittivity is related to a decrease in electron polarization as result of distressed dipoles in chain structure of VHB 4910 elastomer [21][22]. A schematic depend on the basis of chain orientation and dipole distraction is as shown in **fig.1**. However, it can also be attributed to strain-induced; crystallization, change in glass transition phase, reduction of dipole interaction, material morphology, homogeneity, etc. [21]. However, it is good to highlight that the investigations in relevance to strain-induced crystallization limited in literatures of dielectric elastomer although the co-relation between crystallization and mechanical properties are well established for natural rubber, polyethylene terephthalate, poly (trimethylene terephthalate) polylactides, etc. [23]. Recently, stiffen macromolecular chains of nano-composite are shown to acquire effective segmental movement resulting in low glass transition temperature and high permittivity [24]. Interestingly consideration of chain entanglement theories in the practical investigation is both inadequate and significant to control dielectric and mechanical losses.



Fig.1: Schematic illustration for macromolecular chain with (a) aligned dipoles in unstrained and (b) distressed dipoles under pre-strained condition



Fig 2: Characteristic stress-strain plot of vulcanized natural rubber, to illustrate strain induces elastomer softening, hardening, and the difference in uniaxial and biaxial strain[25].

Viscoelastic behaviour is one of the undesired behaviour of rigorously investigated VHB series of dielectric elastomer. Equi-biaxial strain is realized to be more reliable than un-equi biaxial strain, owing stabilize position in actuation state and reduced viscoelastic effects at high strain ratio [26]. In biaxial straining of VHB 4910, the hysteresis curve for relatively smaller elongation (λ= ~1.8) mismatch the expected exponential behaviour at high strain ratios, reasoned as insufficient deformation for strain-hardening due to viscoelastic effect. Also, it is proved that the concept of strain-induced stress-softening and hardening are the function of temperature and cyclic loading [27]. Simultaneous heating of VHB 4905 samples is shown to stabilize the hysteresis thought it found to decreases with increasing temperature [28]. Therefore, in order to have a better understanding of strain softening and strain hardening phenomenon under biaxial and uniaxial strain, the stress–strain curve of vulcanized natural rubber can be referred to **fig.2.** This may able to justify strain-induced chain hardening and softening of VHB 4910 owing similar energy conversion competence as of natural rubber [28][29]. It can be observed that the strain hardening occurs much before in biaxial than the uniaxially strained elastomer so the influence on mechanical properties will probably be different. Low elastic modulus in dielectric elastomer actuator is important parameter to achieve high energy density which is till date lower than the theoretical prediction of 1700 MJ/ kg based on acrylic elastomers [29]. Therefore, reduction in elastic modulus due to strain-induced hardening can be a reason to enhanced electromechanical performance. However independent of analytical evidence this attribution to macromolecular structure is a key drawback in the majority of studies. So the next we demonstrate evaluation of hard segments in terms of strain-induced molecular modification of VHB 4910 dielectric elastomer.

1. **Comparative FTIR of biaxially pre-strained VHB 4910**

The comparative spectra of an unstrained and 300 % biaxially pre-strained VHB 4910 dielectric elastomer is shown in **fig. 3**. In standard observation, the absorbance of infrared light is appeared to decrease with reduced thickness due to applied strain. The absorbance peak at 1747 cm-1 that appeared for strongest C=O bond is found to be unaffected by the action of deformation and a new band emerges for the same at 3448 cm-1 [30]. Further, the absorbance peak between the wavenumber of 630 to 690 cm-1 is observed to emerge as a function of strain. This belongs to an aliphatic molecular structure indicating hard segments in elastomers chains that allow stronger chain mobility and weak molecular interaction, thus significantly decrease the elastic modulus [30]–[32]. This absorbance peak for hard segment signifies that the strain hardening phenomenon as an important parameter in terms of strain induced crystallization and further indicate that the classical theory of chain entanglement and respective variation in mechanical properties [33]. Similarly, the chalcogen bonds that regulate dielectric properties in VHB 4910 can be referred to results of Raman spectroscopy [34]. So the fundamental of chain entanglement is evidenced in terms of molecular modification through spectral analysis and scope to investigate the same for a wide range of pre-strain regimes and deformation parameters.

****

Fig.3 Comparative infrared absorbance spectra of 300 % biaxially strained VHB 4910 dielectric elastomer

1. **Summary**

This article presents a perception towards the development of structure-property relation to understand the effects of pre-strain on actuation performance of dielectric elastomer actuator. The dependency of dielectric and mechanical properties on micro/macromolecular parameter chemical structure, crosslink density, molecular weight is discussed. Chain entanglement is considered as potential parameter to control change in the dielectric permittivity and the elastic modulus. Emergence of hard segment in biaxially pre-strained VHB 4910 is evidence against modification in molecular structure. Analysis on molecular mechanism underlines the prospect of future research towards electromechanical characterization in terms of strain-induced crystallization.

**Acknowledgements**

Authors acknowledge the help rendered by Department of Science and Technology, Govt. of India, New Delhi, for funding this research through grant file no. ECR/2016/000585 dated 27-09-2016.

**References**

[1] R. Shankar, T. K. Ghosh, and R. J. Spontak, Soft Matter, vol. 3, pp. 1116–1129, 2007.

[2] C. Keplinger, M. Kaltenbrunner, N. Arnold, and S. Bauer, Proc. Natl. Acad. Sci., vol. 107, no. 10, pp. 4505–4510, 2010.

[3] L. Yu, F. B. Madsen, and A. L. Skov, Int. J. Smart Nano Mater., vol. 00, no. 00, pp. 1–16, 2017.

[4] L. Jiang, Y. Zhou, S. Chen, J. Ma, A. Betts, and S. Jerrams, J. Appl. Polym. Sci., vol. 45733, pp. 6–11, 2018.

[5] S. Michel, X. Q. Zhang, M. Wissler, C. Löwe, and G. Kovacs, Polym. Int., vol. 59, no. 3, pp. 391–399, 2010.

[6] F. Wu et al., Small, vol. 1702170, pp. 1–7, 2018.

[7] L. Deng, Z. He, E. Li, and S. Chen, J. Appl. Polym. Sci., no. 45850, pp. 1–16, 2018.

[8] R. Pelrine, R. Kornbluh, Q. Pei, and J. Joseph, (80), vol. 287, pp. 836–840, 2000.

[9] A. Kumar, D. Ahmad, and K. Patra, IOP Conf. Ser. Mater. Sci. Eng., vol. 310, no. 012104, pp. 1–9, 2018.

[10] R. K. Sahu, A. Saini, D. Ahmad, K. Patra, and J. Szpunar, J. Mech. Sci. Technol., vol. 30, no. 1, pp. 1–9, 2016.

[11] M. Hossain and P. Steinmann, Smart Mater. Struct., vol. 27, no. 025010, pp. 1–19, 2018.

[12] J. Qiang, H. Chen, and B. Li, Smart Mater. Struct., vol. 21, no. 025006, pp. 1–9, 2012.

[13] E. Biddiss and T. Chau, Med. Eng. Phys., vol. 30, no. 4, pp. 403–418, 2008.

[14] R. K. Sahu and K. Patra, Mech. Adv. Mater. Struct., vol. 23, no. 2, pp. 170–179, 2016.

[15] R. K. Sahu, K. Patra, and J. Szpunar, Strain, vol. 51, pp. 43–54, 2015.

[16] B. Kussmaul et al., Adv. Funct. Mater., vol. 21, no. 23, pp. 4589–4594, 2011.

[17] Q. P. Zhang, J. H. Liu, H. D. Liu, F. Jia, Y. L. Zhou, and J. Zheng, Appl. Phys. Lett., vol. 111, no. 15, pp. 1–4, 2017.

[18] S. Cheng et al., Phys. Rev. Lett., vol. 116, no. 3, 2016.

[19] Y. Nie, Z. Gu, Y. Wei, T. Hao, and Z. Zhou, Polym. J., vol. 49, no. 3, pp. 309–317, 2017.

[20] H. Khatoon and S. Ahmad, J. Ind. Eng. Chem., vol. 53, pp. 1–22, 2017.

[21] T. Vu-cong, N. Ngyen-Thi, and A. Sylvestre, in Electroactive Polymer Actuators and Devices (EAPAD), SPIE, 2014, no. 9056, pp. 1–9.

[22] C. Jean-Mistral, A. Sylvestre, S. Basrour, and J. J. Chaillout, Smart Mater. Struct., vol. 19, no. 075019, pp. 1–9, 2010.

[23] E. Lizundia, A. Larran, A. Larrañaga, and E. Lizundia, Strain-Induced Crystallization. 2018.

[24] S. Javadi, M. Panahi-Sarmad, and M. Razzaghi-Kashani, Polym. (United Kingdom), vol. 145, pp. 31–40, 2018.

[25] D. Q. Tran, J. Li, F. Xuan, and T. Xiao, Mater. Res. Express, vol. 5, no. 065303, pp. 1–12, 2018.

[26] A. Helal, M. Doumit, and R. Shaheen, Appl. Phys. A, vol. 124, no. 1, pp. 1–11, 2018.

[27] G. Berselli, R. Vertechy, M. Fontana, and M. Pellicciari, in Proceedings of the ASME 2014 Conference on Smart Materials, Adaptive Structures and Intelligent Systems, 2014, pp. SMASIS2014-7604, 1–8.

[28] S. J. A. Koh, C. Keplinger, T. Li, S. Bauer, and Z. Suo, IEEE/ASME Trans. Mechatronics, vol. 16, no. 1, pp. 33–41, 2011.

[29] J. D. Davidson and N. C. Goulbourne, J. Mech. Phys. Solids, vol. 61, no. 8, pp. 1784–1797, 2013.

[30] Horiba Jobin Yvon, Raman Application Note, 2017. [Online].

[31] A. Mishra, V. K. Aswal, and P. Maiti, J. Phys. Chem. B, vol. 114, no. 16, pp. 5292–5300, 2010.

[32] L. Jiang, Z. Ren, W. Zhao, and W. Liu, R. Soc. Open Sci., vol. 5, no. 180536, pp. 1–11, 2018.

[33] A. Gros, E. Verron, and B. Huneau, *J. Mech. Phys. Solids*, vol. 125, pp. 255–275, 2019.

[34] D. Sahu, R. K. Sahu, and K. Patra, in Advances in Polymer Composites: Mechanics, Characterization and Applications, 2019, vol. 020032, pp. 1–7.