

Thermoelectric Polymer Aerogels for Pressure-Temperature Sensing Applications

Shaobo Han, Fei Jiao, Zia Ullah Khan, Jesper Edberg, Simone Fabiano and Xavier Crispin

The self-archived postprint version of this journal article is available at Linköping University Institutional Repository (DiVA):

<http://urn.kb.se/resolve?urn=urn:nbn:se:liu:diva-143632>

N.B.: When citing this work, cite the original publication.

Han, S., Jiao, F., Ullah Khan, Z., Edberg, J., Fabiano, S., Crispin, X., (2017), Thermoelectric Polymer Aerogels for Pressure-Temperature Sensing Applications, *Advanced Functional Materials*, 27(44), 1703549. <https://doi.org/10.1002/adfm.201703549>

Original publication available at:

<https://doi.org/10.1002/adfm.201703549>

Copyright: Wiley (12 months)

<http://eu.wiley.com/WileyCDA/>



DOI: 10.1002/1

Article type:**Thermoelectric Polymer Aerogels for Pressure-Temperature Sensing Applications***Shaobo Han, Fei Jiao, Zia Ullah Khan, Jesper Edberg, Simone Fabiano, and Xavier Crispin**

Mr. S. Han, Dr. F. Jiao, Dr. Z. U. Khan, Mr. J. Edberg, Dr. S. Fabiano, Prof. X. Crispin
Laboratory of Organic Electronics
Department of Science and Technology
Linköping University
S-60174 Norrköping, Sweden
E-mail: xavier.crispin@liu.se

The evolution of our society is characterized by an increasing flow of information from things to the internet. Sensors will become the corner stone of internet-of-everything as they track various parameters in our society and sent them to the cloud for analysis, forecast or learning. With the many parameters to sense, sensors are becoming complex and difficult to manufacture. To reduce the complexity of manufacturing, one can instead create advanced functional materials that react to multiple stimuli. To this end, conducting polymer aerogels are promising materials as they combine elasticity and sensitivity to pressure and temperature and they could eventually be functionalized further with chemical and biosensing molecules. However, the challenge is to read independently pressure and temperature output signal without cross-talk of these parameters. Here, we demonstrate a strategy to fully decouple temperature and pressure reading in a dual-parameter sensor based on thermoelectric polymer aerogels. We found that aerogels made of poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) can display either semiconducting or close to semimetallic properties upon exposure to high boiling point polar solvents, such as dimethylsulfoxide (DMSO). As DMSO acts as a secondary dopant, the conductivity of the PNG aerogel can be increased by more than two orders of magnitude, resulting into greatly enhanced pressure sensitivity. Importantly, because of the temperature-independent charge transport observed for DMSO-treated PEDOT-based aerogel, a decoupled

pressure and temperature sensing can be achieved without cross-talk in the dual-parameter sensor devices.

Introduction

Sensors perform a vital function in our daily life as they respond to physical, biological or chemical stimuli from the environment and transform them into communicable data. This provides us with an interface to the real world, and for this reason sensors have become the key components of sophisticated emerging technologies, such as robotics^[1], electronic skin^[2], artificial nose^[3], or electronic tongue^[4]. Their usage is expected to boom even further with the rise of internet-of-everything^[5], as sensors can be used for packaging (e.g. food, drugs) and medical analysis (e.g. point-of care diagnostics), as well as for mapping key environmental parameters (e.g. water purity^[6]), etc. In a near future, sensors will provide huge amount of data to the cloud, enabling machine learning for improving processes in transport, communication, security, healthcare, relationship, economy, pollution, energy and many other areas of our society. Depending on the application, the sensor reading can be extremely complex, involving many different types of sensors. Thus, there is a need for simplifying the manufacturing of sensors and one interesting approach is to create advanced functional materials that react to multiple stimuli in order to create multi-parameter sensors with a single material.

Researchers have started to investigate multi-parameter sensors for basic physical stimuli, such as temperature and pressure. One of the driving motivation is to create electronic-skin(e-skin). For robots or people with amputations or skin damage, artificial devices are needed to mimic the function of skin and restore similar sensing capabilities.^[7, 8] Without covering all the strategies that have been proposed to sense temperature and pressure, we will go through the main ones. A first approach is to create small wireless sensors for humidity and strain sensing. They can be constructed based on LC resonators of a stretchable antenna and humidity sensitive dielectric capacitor^[9]. Although it provides good sensitivity and stability, the readout signal depends on many simultaneous parameters that cannot be decoupled (position of the antenna, temperature, humidity, strain). A second approach is to use an organic transistor

composed of a piezoelectric gate dielectric^[10]. The current in the transistor channel is sensitive to both temperature, due to the temperature activated hopping transport in the organic semiconductor channel, and strain, due to the gate induced piezoelectric voltage. Since the temperature dependence of the current is smaller than the change in current due to strain, it is possible to estimate change in pressure but it is not possible to fully decouple temperature and pressure through the reading of a single parameter, that is, the current. A third approach is to measure the voltage across a droplet with a redox shuttle sandwiched between a hydrophobic electrode and a hydrophilic one^[11]. Temperature and pressure can be measured by the reverse electrowetting effect and the thermogalvanic effect in the liquid droplet. However, there is one signal (i.e. voltage) to read two parameters, which hampers the possibility to read them both simultaneously without cross-talk. A successful approach has been recently proposed to decouple temperature and pressure sensing by reading both current and voltage^[12]. It is based on a commercial polyurethane porous microstructure that is coated with a conducting polymer layer. The conducting polymer provides two functions to the device: resistance change upon pressure and thermoelectric voltage when subjected to a temperature gradient. Hence, in current-voltage characteristics, the voltage shift provides the temperature while the slope of the resistive behavior is related to the pressure.

In the search for new materials that could display similar properties as the functionalized plastic microstructure, we will consider conducting aerogels. Pyrolysis of bacterial cellulose aerogel gives flexible and compressible microporous aerogel with a resistance change of about 10% [80%] for a strain of 10% [70%]. The conductivity is about 0.3 S/cm to 20 S/cm^[13, 14] depending on the synthesis parameters. Another type of conducting aerogels are based on pure graphene. Depending on the manufacturing techniques their mechanical and electrical properties can be completely different. For instance, graphene microporous foam obtained by pyrolysis of polyurethane foam coated with graphene oxide^[15] (conductivity 1 S/cm) displays sensitive pressure-induced change in resistance as high as 100% [800%] for a compression

strain of 10% [30%]; while graphene nanoporous foam has higher conductivity^[16] (40 S/cm) but is almost incompressible and fragile. Conducting polymers in water dispersion constitute another class of conductor that can form aerogels upon freeze drying. Poly(3,4-ethylenedioxythiophene)-polystyrene sulfonate (PEDOT:PSS) is an excellent organic thermoelectric material with high conductivity as well as Seebeck coefficient.^[17-26] Although aerogels of PEDOT:PSS are fragile, they can be reinforced by adding elastomeric cross-linkers such as glycidoxypopyl trimethoxysilane (GOPS)^[27] and mechanical strengthener like nanofibrillated cellulose (NFC)^[28]. Dual parameter sensors have been demonstrated with this type of polymer thermoelectric aerogel.^[21] However, the pressure/temperature sensing operation of such devices suffers from a significant cross-talk, which is likely related to the inherent charge transport mechanism in the conducting aerogels.^[29]

In this article, we demonstrate dual-parameter sensors based on thermoelectric polymer aerogels with fully decoupled temperature and pressure sensing capability. We achieve this by finely tuning the transport properties of the conducting aerogels with exposure to the vapor of high boiling point polar solvents, such as dimethylsulfoxide (DMSO). The thermoelectric polymer aerogels used in this work are prepared by freeze-drying the water dispersion of three organic materials: PEDOT:PSS, NFC and GOPS (PNG aerogels). The post-treatment with the DMSO vapor boosts the pressure sensitivity by two orders of magnitude and lead to an absence of cross-talk in the dual-parameter sensors. We found that this is due to a unique activated free transport regime close to the semimetallic regime of the polymer aerogel.

Results and discussion

PEDOT:PSS-NFC aerogel

The chemical structures of PEDOT:PSS, NFC and GOPS are shown in **Figure 1a**. Conducting polymer aerogels can be prepared by freeze drying a PEDOT:PSS water emulsion. However, the resulting aerogels from pure PEDOT:PSS are brittle and not flexible. In contrast, NFC aerogels are mechanically robust. NFC^[30] has been used with promising results as reinforcement material in composites because of its remarkable mechanical properties. It also exhibits low density, high aspect ratio, high surface area and, more important, it is non-toxic and biodegradable. Furthermore, by substitution of its abundant hydroxyl groups, modified NFC^[31] could have increased application versatility and benefits, and different NFC nanocomposites in the form of thin films^[32], aerogels^[33] and fibers^[34] have been reported. Although PEDOT:PSS itself can also form aerogels, the addition of NFC could further increase the strength, flexibility and thermal stability of the porous scaffold. By combining PEDOT:PSS and NFC, we obtained mechanically robust thermoelectric aerogel, although still not elastic enough. GOPS has an epoxy group which is active to bound with the hydroxyl groups present in PSS, thus reticulating PEDOT:PSS.^[35] The extra epoxy groups are also expected to react with hydroxyl groups of NFC. This crosslinking reaction reticulated the PNG aerogels. So, PEDOT:PSS will sense temperature variation while the combination of NFC and GOPS will ensure enhanced mechanical strength and elasticity of the aerogel to sense pressure changes. To prepare the aerogels, PEDOT:PSS, GOPS and NFC were mixed in solution form by an ULTRA-TURRAX T 10 disperser, with a solid content weight ratio of 1:0.2:1 respectively. To fabricate aerogels, water molecules in this solution should be removed without collapsing the NFC frame. To achieve this goal, the mixed solution was first loaded into an aluminum alloy mold and frozen by liquid nitrogen, followed by freeze drying (Benchtop Pro) under -60 °C and 50 μ bar for 12h. The aerogels were then put into an oven under 140 °C for 30 min with

atmosphere of air for the GOPS to crosslink with PEDOT:PSS and NFC. The shape and size of the aerogel was controlled by the mold used during the freeze drying step (**Figure 1b**). Scanning electron microscopy (SEM) images (**Figure 1c**) reveal that the PNG aerogel has porous structure composed of thin flakes and fibers which are connected to each other. Because of the crosslinking reaction between GOPS and PEDOT:PSS as well as NFC, the aerogel could completely recover to its initial form after being compressed to about 1/5 of its original thickness under pressure as shown in **Figure 1d**. Meanwhile, this uniform porous structure provides the aerogel the possibility that its resistance will change with different thickness caused by different pressure.

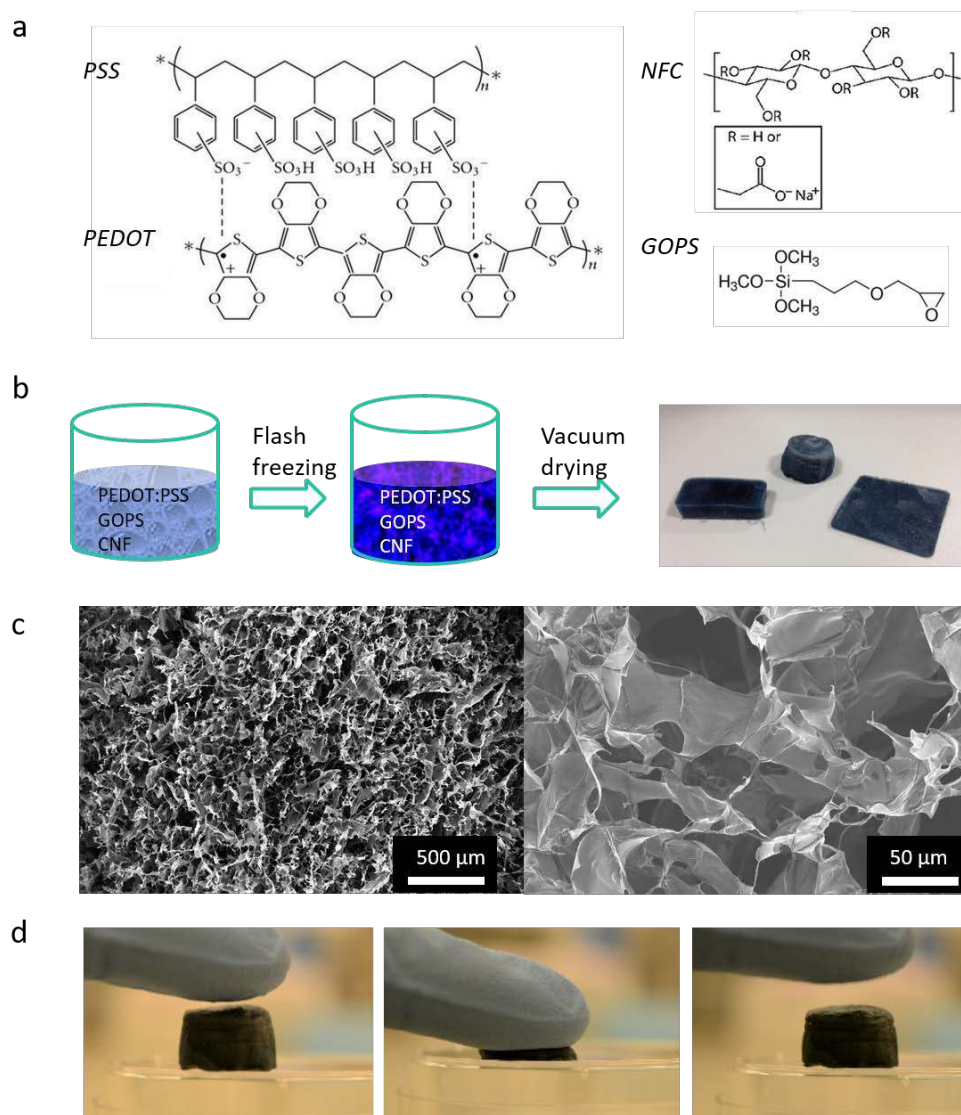


Figure 1. a) Chemical structures of PEDOT:PSS, GOPS and NFC; b) Production process of PNG aerogel and different samples with different shapes; c) SEM images of PNG aerogel and d), PNG aerogel states before (left), under (middle) and after (right) finger pressing.

Device fabrication and primary measurement

The components of a sophisticated application for example e-skin that need to be developed include sensors, signal encoding and a method to deliver the sensory information to a central control system^[36]. In the current study, we focus on the optimization of materials for sensing simultaneously temperature and pressure through an electrical readout; which is appropriate to integrate in a matrix of sensors for mapping/imaging pressure and temperature. **Figure 2a** shows the schematic of the experimental set-up used to characterize the PNG dual-parameter sensor. Peltier elements were used to control the temperature. The temperature is read on the top and bottom of the PNG aerogel by two gold thermistors patterned on silicon substrates. A Poly(methyl methacrylate) (PMMA) layer was then spin coated on the thermistor to electrically insulate it from PEDOT:PSS electrode. Flexible carbon fibers were used to connect the electrodes to the measurement system. Through the PEDOT:PSS electrodes, we applied voltage sweep and measured the current under different temperatures and pressures.

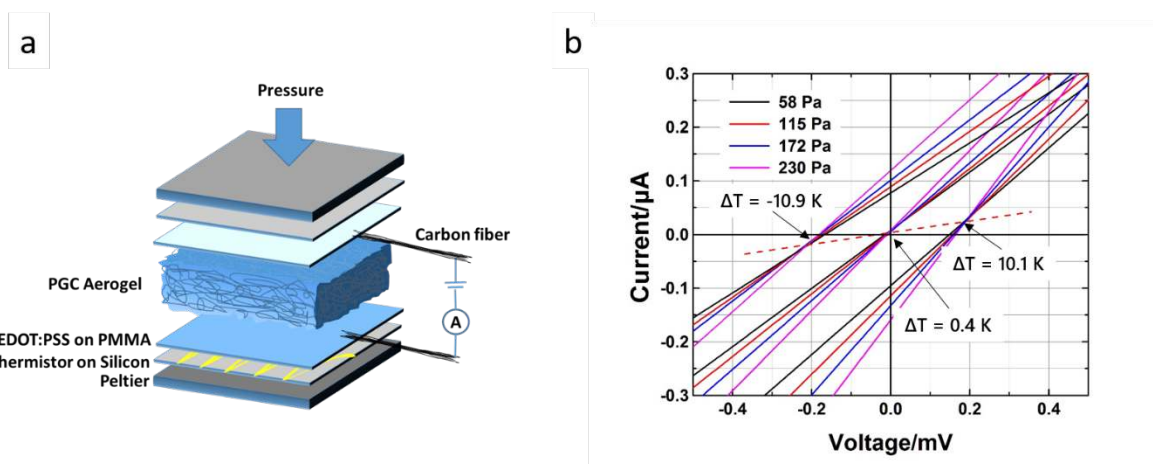


Figure 2. a) Schematic of a sensor made by PNG aerogel. A PNG aerogel is sandwiched between two electrodes. The electrodes are then connected with a measurement system,

Keithley 4200. Two thermistors are attached with the electrodes to read the temperature and two Peltier element are then attached with the thermistors to control the temperature. b) Measured I-V curves with different temperatures and pressures. The temperature of one Peltier element was constantly kept at 22 °C while varying the other one.

Figure 2b displays the response of a sensor made of the PNG aerogels. When the aerogel is compressed by applying a pressure on the top of the PNG aerogel, its resistance decreases, as revealed by an increase in the slope of the I-V linear curves with increasing the pressure (58, 115, 182 and 230 Pa). By controlling the temperature of the two electrodes with two Peltier elements, a temperature difference is added and a corresponding thermovoltage equals to $\alpha \Delta T$ (with α the Seebeck coefficient) is created, which shifts the I-V curves ($\Delta T = -10.9, 0.4, 10.1$ K). Hence, the PNG dual-parameter sensors allow decoupling the pressure and temperature signals by reading, respectively, slopes and voltage axis intercept of the I-V curves. However, because of the low conductivity of PNG aerogel, the read Ohmic current is very low leading to a consequent low pressure sensitivity of the sensor. At the same time, because of the high activation energy for charge transport in PEDOT:PSS, the conductivity of the aerogel changes not only with pressure but also with temperature. As a result, one cannot get the real pressure data from the slopes of the I-V characteristics because they are different under the same pressure at different temperatures. Hence, with this material composition, temperature gradient and pressure cannot be simultaneously decoupled in a simple way.

Optimization of the PEDOT:PSS-NFC aerogel sensor properties

As mentioned above, any changes in the slope of the I-V curve indicates a change in the applied pressure^[18]. A good sensor should have a high pressure sensitivity which is defined as $S_P = \Delta I / \Delta P$, where ΔI is the change in current under constant voltage caused by a change in pressure (ΔP). This requires the sensor to have high conductivity to achieve high S_P . As the

conductivity of the pristine aerogel is only about 1×10^{-4} S/cm, the pressure sensitivity is as small as 3×10^{-7} A/Pa. Such a low pressure sensitivity causes a big challenge for the signal encoding section of e-skin as it requires very accurate measurements to resolve such low variations. Hence, to solve the conductivity issue of the PNG aerogel and to increase its pressure sensitivity, we used DMSO vapor to treat the samples (**Figure 3a**). DMSO, as well as other high boiling polar solvents such as glycerol, sorbitol, and ethylene glycol, are in fact very well-known “secondary dopants” for PEDOT:PSS, and they are known to affect the morphology of PEDOT film rather than its oxidation level. As reported by Yeo et al.^[37], for instance, DMSO vapor induces significant phase separation between the excess of PSS and PEDOT chains and spontaneous formation of a highly enriched PSS layer on the top surface of the PEDOT:PSS polymer blend. This leads to an increase of the electrical conductivity by many orders of magnitude. We treated the PNG aerogel with a DMSO vapor, as sketched in Figure 3a. A glass petri-dish is placed on top of a hotplate with the PNG aerogel inside. Small amount of DMSO (a few drops) is placed around the sample but not in contact with it, to generate a saturated DMSO vapor environment in the chamber. The conductivity and Seebeck coefficient are measured for different DMSO treating time and the results are shown in **Figure 3b**. The conductivity increases of more than two orders of magnitude going from $\sim 1 \times 10^{-4}$ S/cm for the pristine samples to a saturated peak value of ~ 0.03 S/cm after 10-minute exposure to the DMSO vapor. The increased conductivity leads to a higher I under a constant pressure P , which further leads to a greater ΔI caused by the same change in pressure ΔP . Thus, the pressure sensitivity S_P increased from 3×10^{-7} A/Pa (measured under 100 mV) to 2×10^{-5} A/Pa after 5-minute exposure to DMSO vapor (see Figure 3c). This enhanced sensitivity will not only make the encoding easier but also provide more accurate pressure information.

As for temperature detection, the level of generated thermovoltage (due to Seebeck coefficient of the PNG aerogel) is an indication of the temperature sensitivity ($\alpha = \Delta V_{\text{therm}}/\Delta T$). As shown in **Figure 3b**, the Seebeck coefficient of the PEDOT-based aerogel ($\alpha = \sim 20$ μ V/K) remains

constant regardless of the DMSO vapor treatment which means the temperature sensitivity of PNG aerogel is not affected by DMSO. Similar findings were reported for PEDOT thin films with a constant Seebeck coefficient versus conductivity below 500 S/cm together with a temperature activated transport^[38].

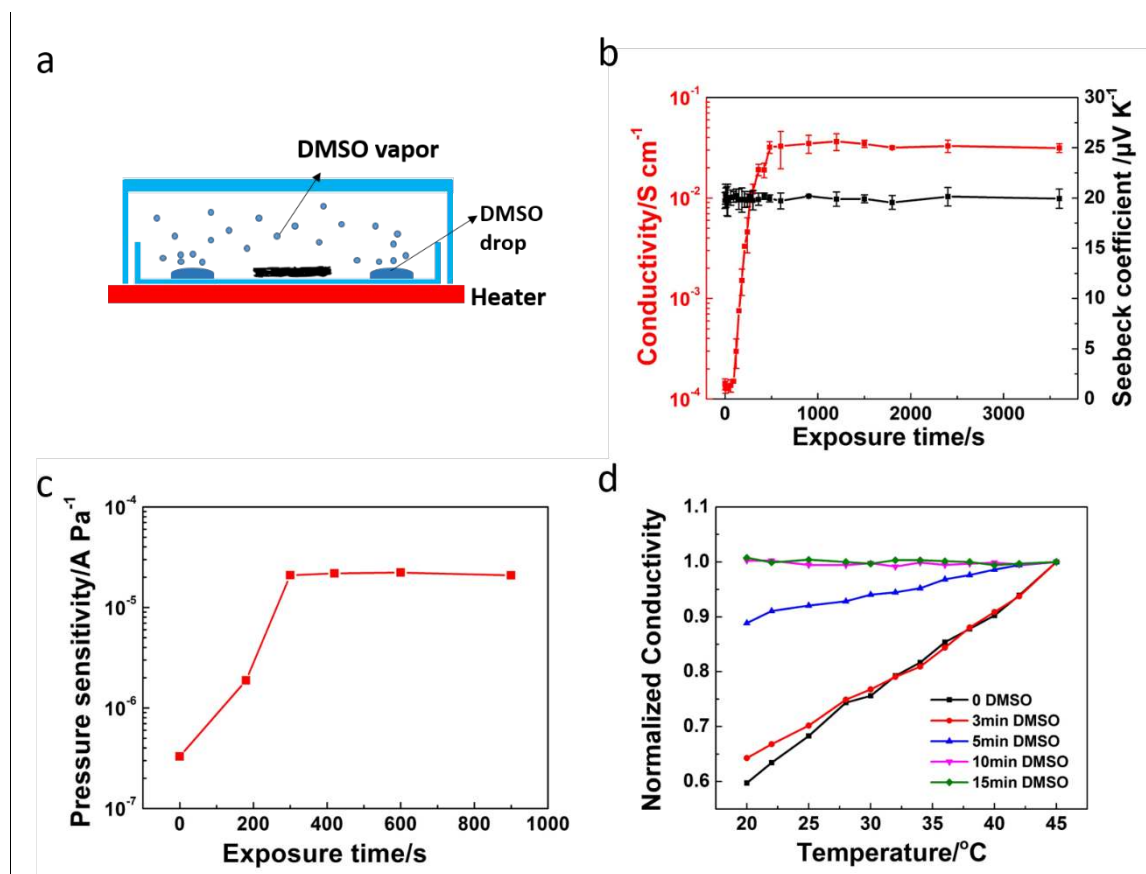


Figure 3 a) Schematic of DMSO vapor treatment. b) Conductivity and Seebeck coefficient as functions of exposure time in DMSO vapor. These two curves show that DMSO vapor increases the conductivity but does not affect the Seebeck coefficient of the PNG aerogel. c) Pressure sensitivity versus DMSO vapor exposure time. The current was measured under 100 mV. It shows a dramatic increase of pressure sensitivity after about 5 min treatment. d) Normalized conductivity as a function of temperature after DMSO vapor treatment in different time. After about 10 min treatment, the conductivity of PNG aerogel kept constant with temperature

One could in principle extract both temperature and pressure information from the I-V curves reported in **Figure 2b**. However, untreated (pristine) PNG aerogel cannot achieve this goal with precision because, under the same pressure but different temperature, the I-V curves are not parallel to each other (that is, they have different resistances). The reason is that the conductivity of the aerogel is governed by a temperature activated hopping transport as it is typically observed for untreated PEDOT:PSS thin films.^[39] **Figure 3d** shows normalized conductivity values (with respect to conductivity at 45 °C) as a function of temperature. For 0, 3 and 5 min exposure time to the DMSO vapor, the PNG aerogel retains a positive correlation of conductivity *vs.* temperature. As a result, under a fixed pressure, a change of temperature leads to a change in resistance which further causes a change of the I-V curve's slope as shown in **Figure 4a**. The change in the I-V slope caused by the temperature dependent conductivity will interfere with the change in the I-V slope caused by pressure. Thus, to achieve a neat decoupled read-out, the correlation between temperature and conductivity of PEDOT:PSS has to be removed. **Figure 3d** clearly shows that the conductivity of PNG aerogel tends to become constant with temperature after 10 min of DMSO vapor treatment. This activated-free transport of the charge carriers is indicative of a transition from semiconductor to semimetallic character^[38]. Our findings are in agreement with other works showing the effect of DMSO treatment on PEDOT:PSS films where even negative slopes in conductivity *vs.* temperature, that is metallic-like behavior is observed^[39]. It is remarkable that we succeed to obtain similar properties in an aerogel for the first time. As a result, the I-V curves under same pressure and different temperature tend to be parallel with each other, as shown in **Figure 4b**. Consequently, the problems of PNG aerogel sensor of (i) low pressure sensitivity and (ii) resistance changing with both pressure and temperature are solved by simple treatment with DMSO vapor.

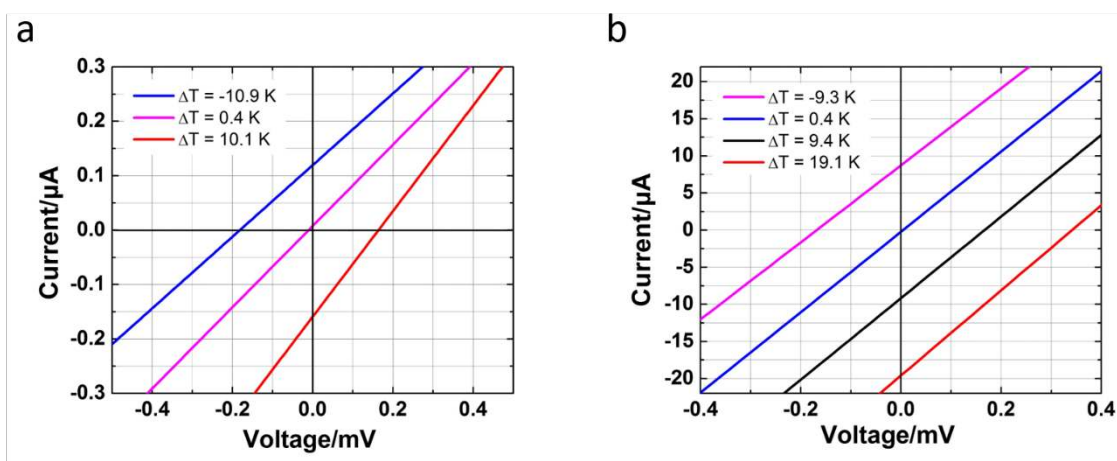


Figure 4 a) I-V curves with same pressure (230Pa) and different temperature before DMSO treatment. The curves are not parallel with each other because the conductivity of PNG aerogel is changing with temperature. b) I-V curves with same pressure (230Pa) and different temperature after DMSO treatment. The curves become parallel with each other because the conductivity of PNG aerogel is no longer changing with temperature.

The combined pressure and temperature effects after DMSO treatment are shown in **Figure 5**. At a fixed temperature value and varying pressures, different slopes of I-V curves are observed, whereas under a fixed pressure and changing temperatures, we get different intersection position of I-V curves perfectly aligned on voltage axis. Hence, the activated-free transport leads to a true decoupling of the output reading of the sensor: the slope is a direct measurement of the pressure and the intercept with the V-axis is a reading for the temperature gradient.

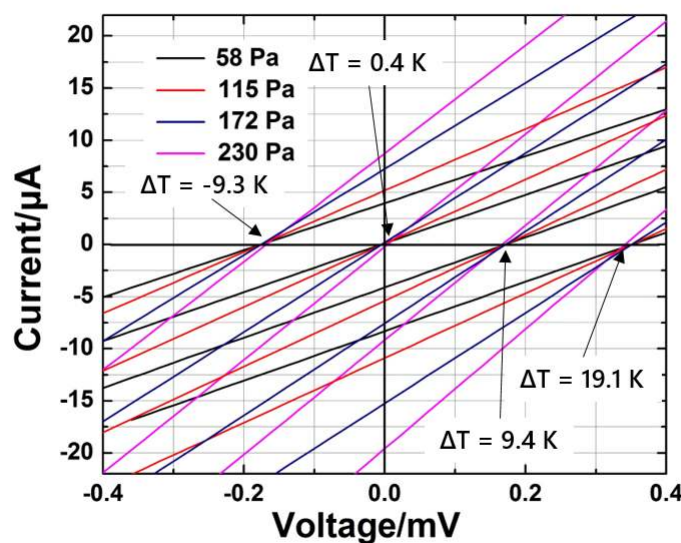


Figure 5. The measured I-V curves of PNG aerogel after 10 min DMSO vapor treatment. The T_0 is 22 °C. Under a temperature difference and pressure, by measuring two points in the I-V curves, one can achieve a line in which the temperature information could be read from the intersecting point position on voltage axis and the pressure information could be read from the slope.

Conclusion

We have optimized the pressure sensitivity of temperature-pressure dual-parameter sensor made by PNG aerogel via a treatment with DMSO vapor. As DMSO acts as a secondary dopant, the conductivity of the PNG aerogel increases from 1×10^{-4} S/cm to 3×10^{-2} S/cm, which results into a great enhancement of the pressure sensitivity from 3×10^{-7} A/Pa to about 2×10^{-5} A/Pa (measured at 100 mV). Importantly, the DMSO treatment leads to a change in the mechanism of charge carrier transport in the aerogel, going from a thermally activated hopping to a temperature-independent transport typically observed for PEDOT at the semiconductor to semimetal transition. This has only been observed for thin film so far, so it is remarkable reach a semimetallic behavior in a conducting polymer aerogel. The impact of the transport mechanism is very valuable for the dual sensor. Indeed, as the conductivity does not change

with temperature, there is a clear decoupling between temperature and pressure reading with the same material sensor. We foresee that arrays of these optimized dual-parameter sensors can be utilized in various e-skin applications.

Acknowledgements

S. Han and F. Jiao contributed equally to this work. The authors acknowledge the European Research Council (ERC-starting grant 307596), the Swedish Foundation for Strategic Research, the China Scholarship Council (CSC), the Knut and Alice Wallenberg Foundation, The Swedish Energy Agency, the Advanced Functional Materials Center at Linköping University and the Research Institute of Sweden (RISE).

Received: ((will be filled in by the editorial staff))

Revised: ((will be filled in by the editorial staff))

Published online: ((will be filled in by the editorial staff))

- [1] G. Indiveri, F. Douglas, *Science* **2000**, 288, 1189.
- [2] Q. Sun, W. Seung, B. J. Kim, S. Seo, S. W. Kim, J. H. Cho, *Adv. Mater.* **2015**, 27, 3411.
- [3] T. A. Dickinson, K. L. Michael, J. S. Kauer, D. R. Walt, *Anal. Chem.* **1999**, 71, 2192.
- [4] J. J. Lavigne, S. Savoy, M. B. Clevenger, J. E. Ritchie, B. McDoniel, S. J. Yoo, E. V. Anslyn, J. T. McDevitt, J. B. Shear, D. Neikirk, *Journal of the American Chemical Society* **1998**, 120, 6429.
- [5] S. C. Li, L. D. Xu, X. H. Wang, *Ieee Transactions on Industrial Informatics* **2013**, 9, 2177.
- [6] E. Mitraka, L. Kergoat, Z. U. Khan, S. Fabiano, O. Douheret, P. Leclere, M. Nilsson, P. A. Ersman, G. Gustafsson, R. Lazzaroni, M. Berggren, X. Crispin, *J Mater Chem C* **2015**, 3, 7604.
- [7] A. Chortos, J. Liu, Z. Bao, *Nat Mater* **2016**, 15, 937.
- [8] P. D. Marasco, K. Kim, J. E. Colgate, M. A. Peshkin, T. A. Kuiken, *Brain* **2011**, 134, 747.
- [9] X. Huang, Y. H. Liu, H. Y. Cheng, W. J. Shin, J. A. Fan, Z. J. Liu, C. J. Lu, G. W. Kong, K. Chen, D. Patnaik, S. H. Lee, S. Hage-Ali, Y. G. Huang, J. A. Rogers, *Adv. Funct. Mater.* **2014**, 24, 3846.
- [10] N. T. Tien, S. Jeon, D. I. Kim, T. Q. Trung, M. Jang, B. U. Hwang, K. E. Byun, J. Bae, E. Lee, J. B. H. Tok, Z. N. Bao, N. E. Lee, J. J. Park, *Adv. Mater.* **2014**, 26, 796.
- [11] K. Liu, Y. S. Zhou, F. Yuan, X. B. Mo, P. H. Yang, Q. Chen, J. Li, T. P. Ding, J. Zhou, *Angew. Chem. Int. Ed.* **2016**, 55, 15864.
- [12] F. J. Zhang, Y. P. Zang, D. Z. Huang, C. A. Di, D. B. Zhu, *Nat Commun* **2015**, 6.
- [13] Z. Y. Wu, C. Li, H. W. Liang, J. F. Chen, S. H. Yu, *Angew. Chem. Int. Ed.* **2013**, 52, 2925.
- [14] H. W. Liang, Q. F. Guan, Z. Zhu, L. T. Song, H. B. Yao, X. Lei, S. H. Yu, *Npg Asia Mater* **2012**, 4.
- [15] Y. A. Samad, Y. Q. Li, A. Schiffer, S. M. Alhassan, K. Liao, *Small* **2015**, 11, 2380.
- [16] Z. Xu, Y. Zhang, P. G. Li, C. Gao, *Acs Nano* **2012**, 6, 7103.
- [17] F. Zhang, Y. Zang, D. Huang, C. A. Di, D. Zhu, *Nat Commun* **2015**, 6, 8356.
- [18] Z. U. Khan, J. Edberg, M. M. Hamedi, R. Gabrielsson, H. Granberg, L. Wagberg, I. Engquist, M. Berggren, X. Crispin, *Adv Mater* **2016**, 28, 4556.
- [19] F. L. E. Jakobsson, X. Crispin, L. Lindell, A. Kanciurowska, M. Fahlman, W. R. Salaneck, M. Berggren, *Chem Phys Lett* **2006**, 433, 110.
- [20] M. Vosgueritchian, D. J. Lipomi, Z. Bao, *Adv. Funct. Mater.* **2012**, 22, 421.
- [21] Y. H. Kim, C. Sachse, M. L. Machala, C. May, L. Müller-Meskamp, K. Leo, *Adv. Funct. Mater.* **2011**, 21, 1076.
- [22] J. Y. Kim, J. H. Jung, D. E. Lee, J. Joo, *Synth. Met.* **2002**, 126, 311.
- [23] J.-G. Chen, H.-Y. Wei, K.-C. Ho, *Sol Energ Mat Sol C* **2007**, 91, 1472.
- [24] J. Luo, D. Billep, T. Waechtler, T. Otto, M. Toader, O. Gordan, E. Sheremet, J. Martin, M. Hietschold, D. R. T. Zahn, T. Gessner, *J. Mater. Chem. A* **2013**, 1, 7576.
- [25] M. Scholdt, H. Do, J. Lang, A. Gall, A. Colsmann, U. Lemmer, J. D. Koenig, M. Winkler, H. Boettner, *Journal of Electronic Materials* **2010**, 39, 1589.
- [26] H. Park, S. H. Lee, F. S. Kim, H. H. Choi, I. W. Cheong, J. H. Kim, *J. Mater. Chem. A* **2014**, 2, 6532.
- [27] J. Lin, J. A. Siddiqui, R. M. Ottenbrite, *Polym Advan Technol* **2001**, 12, 285.

- [28] A. Malti, J. Edberg, H. Granberg, Z. U. Khan, J. W. Andreasen, X. Liu, D. Zhao, H. Zhang, Y. Yao, J. W. Brill, I. Engquist, M. Fahlman, L. Wagberg, X. Crispin, M. Berggren, *Adv. Sci.* **2016**, 3, 1500305.
- [29] O. Bubnova, Z. U. Khan, H. Wang, S. Braun, D. R. Evans, M. Fabretto, P. Hojati-Talemi, D. Dagnelund, J. B. Arlin, Y. H. Geerts, S. Desbief, D. W. Breiby, J. W. Andreasen, R. Lazzaroni, W. M. Chen, I. Zozoulenko, M. Fahlman, P. J. Murphy, M. Berggren, X. Crispin, *Nat Mater* **2014**, 13, 190.
- [30] J. Leitner, B. Hinterstoisser, M. Wastyn, J. Keckes, W. Gindl, *Cellulose* **2007**, 14, 419.
- [31] S. Fujisawa, Y. Okita, T. Saito, A. Isogai, *Abstracts of Papers of the American Chemical Society* **2012**, 243.
- [32] Y. Qing, R. Sabo, Y. Q. Wu, Z. Y. Cai, *Bioresources* **2012**, 7, 3064.
- [33] F. Jiang, Y. L. Hsieh, *J. Mater. Chem. A* **2014**, 2, 6337.
- [34] R. Endo, T. Saito, A. Isogai, *Polymer* **2013**, 54, 935.
- [35] A. Hakansson, S. B. Han, S. H. Wang, J. Lu, S. Braun, M. Fahlman, M. Berggren, X. Crispin, S. Fabiano, *J Polym Sci Pol Phys* **2017**, 55, 814.
- [36] C. Antfolk, M. D'Alonzo, B. Rosen, G. Lundborg, F. Sebelius, C. Cipriani, *Expert Rev Med Devices* **2013**, 10, 45.
- [37] J. S. Yeo, J. M. Yun, D. Y. Kim, S. Park, S. S. Kim, M. H. Yoon, T. W. Kim, S. I. Na, *Acs Applied Materials & Interfaces* **2012**, 4, 2551.
- [38] O. Bubnova, Z. U. Khan, H. Wang, S. Braun, D. R. Evans, M. Fabretto, P. Hojati-Talemi, D. Dagnelund, J. B. Arlin, Y. H. Geerts, S. Desbief, D. W. Breiby, J. W. Andreasen, R. Lazzaroni, W. M. M. Chen, I. Zozoulenko, M. Fahlman, P. J. Murphy, M. Berggren, X. Crispin, *Nat. Mater.* **2014**, 13, 190.
- [39] S. K. Yee, N. E. Coates, A. Majumdar, J. J. Urban, R. A. Segalman, *Physical Chemistry Chemical Physics* **2013**, 15, 4024.