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Effects of Solvents on the Material Properties of Screen-Printed Electrodes and a Polydimethylsiloxane Dielectric for Dielectric Elastomer Transducers

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Dielectric elastomer transducers (DETs), namely, highly stretchable dielectric membranes sandwiched between two compliant electrodes, allow to develop sensors, actuators, and generator systems. DET performance is strongly influenced by the material properties of dielectric and electrodes, as well as the type of manufacturing process. For instance, when manufacturing the electrodes via screen-printing, significant amounts of solvents are needed to adjust the printing materials' viscosity, but they potentially alter DET properties. The investigation of this effect, however, is largely unexplored. Addressing this issue, this article investigates the influence of various solvents on a DET consisting of a polydimethylsiloxane (PDMS) membrane with PDMS/carbon black electrodes. Physical and chemical interactions between dielectric and solvents are characterized by infrared-spectroscopy (IR), optical investigations, and light microscopy. Different solvents used for screen-printing are analyzed in terms of their effects on the electrode electrical resistance, screen-print image, and breakdown strength. Even though the solvent strongly affects the electrodes' electrical resistance (ranging from ≈22 kΩ to >MΩ), it does not modify the breakdown strength of the pure dielectric (113 V µm^{-1}). In the IR, no chemical aging is found. The obtained results provide systematic guidelines for the choice of solvents in DET electrode manufacturing.

1. Introduction

In recent years, dielectric elastomer transducers (DET) have attracted the interest of many researchers and industries. DETs consist of a dielectric elastomer membrane and two compliant flexible electrodes, which together form a capacitor. Because of their high stretchability, high energy density and efficiency, lightweight, and low power consumption, DETs can be used in applications as soft sensors, actuators, or generators.[1] Examples of DET-based systems which have been presented in the literature include pumps,[2] valves,[3] force and pressure sensors,[4] soft robots,[5,6] smart textiles,[7] and loudspeakers,[8] to name a few. In DET applications, the dielectric should commonly be thin (≈50 µm) to reduce the needed actuation voltage. Additionally, the dielectric thickness needs to be homogeneous, and the material must exhibit as low mechanical hysteresis as possible. Commonly adopted dielectric materials used in DET applications are silicone-based membranes, e.g., Wacker Elastosil 2030,[6,9] or an acrylic polymer, e.g., VHBs ("very high bond" materials are available by 3M),[10,11] which are both commercially available. The compliant electrodes, in contrast, need to be thin in order not to alter the DET mechanical properties, must have a low electrical resistance to allow fast and efficient actuation, and need to maintain their characteristics even when subjected to large strains on the order of 50–100% or higher. Common DET electrode materials vary from conductive polymers such as poly(3,4-ethylenedioxythiophene)-poly(styrene sulfonate) (PEDOT:PSS),[12,13] to nonconductive polymer matrices filled with carbon nanotubes or carbon nanofibers,[14–17] carbon black (CB),[9,10,18–21] or graphene,[22] up to thin metallic films.[23,24]

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The characteristics of a DET, e.g., the electrodes' electrical resistance,[25] the actuation strain,[26,27] the breakdown strength,[28] or the mechanical behavior,[1,27] can vary significantly when changing the composition of dielectric and/or electrode materials. In addition, the adopted manufacturing process and its parameters for the manufacturing of the electrodes can influence DET characteristics, such as the electrode thickness, the homogeneity, or the electrical resistance.[29,30] Examples of reproducible and up-scalable manufacturing processes using filled polymer-based electrodes for DETs (like the one used in this work) are pad-printing,[21,31] airbrushing,[32] inkjet-printing,[33,34] or screen-printing.[20,30] Even though those manufacturing processes are based on different principles, a common parameter among all of them is the amount of solvent used to adjust the viscosity of the printing material, whose amount can in some cases exceed 70 weight percent (wt%) of the electrode material.[20,30,33] While these solvent amounts are common, some approaches do not use solvent in their carbon-based electrode inks for DETs, e.g., when manufacturing them with screen-printing.[22] This may have benefits like manufacturing the electrodes more eco-friendly,[22] but since it is common to use solvents for DET electrode manufacturing, and the scope of the article is the investigation of the influence of various solvents on the properties of DET, when their electrodes are manufactured with CB/PDMS ink diluted by solvents, formulations without solvents are here not further discussed.

While the literature has mostly focused on the manufacturing processes or the used materials in the final DETs itself, the role of solvents and their influences on the resulting DET performance is often overlooked. The type of solvent may, for instance, change electrode features, diffuse in the dielectric matrix change the mechanical behavior and the dielectric constant, interact chemically with any of the DET components, or even have other influences. A relevant example occurs when considering matrices made of highly cross-linked polymers such as polydimethylsiloxane (PDMS), which do not dissolve in solvents but may exhibit swelling on contact. The degree of swelling is influenced by the cohesion energy and polarity of the two components.[35] The higher the similarity between solvent and polymer, the stronger the interactions. As PDMS has a hydrophobic surface, highly polar solvents such as water or ethanol are suitable for PDMS microfluidic systems due to their low solubility, while less polar solvents and amines such as hexamine or triethylamine are used to remove oligomers from PDMS.[36–38] As the dielectric membrane has direct contact with the solvent in the manufacturing processes of the DET electrodes, an interaction is expected. The nature of the solvent can thus have a significant impact on the type and intensity of this interaction. Moreover, the volatility of solvents is also an important factor to consider. High volatility and vaporization may be wanted during spray-painting, where solvents should vaporize while the electrode is sprayed from the nozzle to the substrate. Other processes like screen-printing require low volatility to prevent vaporization during the manufacturing process, as the increasing viscosity due to solvent evaporation from the printing material would cause differing electrode qualities. To date, no studies in the literature report the influence of different solvents on DET characteristics and manufacturing, to the best of our knowledge. This work aims at creating for the first time a basic understanding of the influences of various solvents on performance of screen-printed DETs. More specifically, we investigate the influence of six commercially available solvents (VD10 (VD = Verdünner; German for thinner), VD40, VD60, VZ40 (VZ = Verzögerer; German for retarder), Belsil DM 1 Plus – hereafter referred to simply as Belsil –, and Isopar M) on the electrical properties of DETs. The dielectric is chosen to be a commercially available PDMS membrane (Wacker Elastosil 2030 from Wacker Chemie AG, Munich, Germany), due to its low hysteresis in cyclic tensile tests,[39,40] fast actuation response,[40] good sensitivity and bandwidth in sensing applications,[41] as well as its commercial availability as large membranes with low but homogenous thicknesses ($\leq$50 µm). The electrode is a PDMS-based composite filled with CB to ensure electrical conductivity,[9,10,18-21] which is capable of high stretchability without losing its electrical conductivity or influencing the mechanical behavior to a too large extent,[20] and is based on readily available and low-cost ingredients. The manufacturing process chosen for the electrodes used in this work is screen-printing, since it is a fast method, allows to print all shapes and sizes of electrodes and allows fast prototyping, is mass-scalable, and provides good reproducibility. In this work, the above combination of DET material, electrodes, and screen-printing is used as a case study to showcase the importance and the influence of the solvents on the overall process. The characteristics of the solvents are evaluated with respect to their usability for up-scaled, as well as automated, processes for longer printing sessions. An extensive study is conducted using optical methods such as light microscopy, attenuated total reflection–Fourier transform infrared spectroscopy (ATR–FTIR), and direct observation. These tests are conducted on the pure membrane to obtain a reference, and additionally after the membranes have been treated with pure solvents. The obtained results provide initial insights into the physical and chemical interaction between solvents and dielectric membranes. Additionally, electrodes are applied to dielectric membranes by the screen-printing process, whereby the kind of solvent in the printing ink is varied. The electrodes are then characterized in terms of their screen-print image, their electrical resistance, and the breakdown strength of the dielectric membrane, including an observation of the influence of solvents on the manufacturing process. The results are discussed, and the solvents used in this work are rated according to their usability for the given manufacturing process of DETs. The results gathered in these investigations will allow the reader to comprehend the influence of solvents on PDMS dielectric membranes. Additionally, this work also provides the reader with guidelines on the type of solvent preferable for the manufacturing, and aspects that need to be considered during the process depending on the desired DET characteristics.

2. Materials and Sample Preparation

This section provides a brief overview of the used materials (Section 2.1), as well as the sample preparation with and without screen-printed electrodes (Sections 2.2 and 2.3).

2.1. Materials

Elastosil 2030 PDMS silicone film provided by Wacker Chemie AG is used in this work as dielectric material. The selected membranes have a thickness of 50 µm. The manufactured electrodes consist of PDMS (Silgel 612 A/B, Wacker Chemie AG, Munich, Germany) filled with CB, whereby the finally cured electrode consists of 16 wt% CB and 84 wt% silicone. The CBs used are Ketjenblack EC-600JD (Akzonobel, Amsterdam, Netherlands) and Ensaco 350G (Imerys, Paris, France), while their ratio to each other is 1:1 in the manufactured electrodes. The silicone dielectric membrane as well as the silicone and CB used for the electrodes are kept the same throughout all experiments, while only the solvents are varied. Information on the used solvents is listed in **Table 1**, based on the corresponding data sheets. Note that the displayed evaporation rate is measured against butyl acetate, while the evaporation rate of butyl acetate is set to 1 (without units). A lower evaporation rate means slower evaporation.

VD10, VD40, and VD60 are commercially available solvents suitable for pad-printing or screen-printing. The volatility of the thinner increases with lower number in the name. VZ40 is a retarding agent intended to increase processing time and is used in combination with a thinner rather than as a single solvent. For the investigation of the influence of the solvents with the dielectric membrane, VZ40 was used as a single solvent to separate influences of different solvents from each other. Additionally, Belsil from Wacker Chemie AG and Isopar M from ExxonMobil are investigated. Belsil was chosen as a solvent because the molecular structures of its components are like the structure of the PDMS used as matrix material for the electrode. Isopar M was used for its low volatility compared to the other solvents, thus allowing longer printing sessions without significant change in the viscosity of the printing material. This is best suited for long or automated processes.

2.2. Sample Preparation

In this section, the preparation of the samples is described. Section 2.2.1 contains the manufacturing and treatment of the silicone membranes with pure solvent, while Section 2.2.2 describes the manufacturing of the electrodes via screen-printing.

2.2.1. Pure Solvent on Silicone Membranes

The Wacker Elastosil 2030 silicone membrane was fixed with Kapton tape to metal frames without prestretch. Specimens with various frame sizes were used when performing different measurements. For optical measurements, the metal frames had a dimension of 110 × 110 mm with a free quadratic space in the middle of 70 × 70 mm. This geometry allows a large area for observation and investigations by light microscopy and ATR–FTIR. The second geometry of 140 × 60 mm and a free area of 130 × 30 mm was needed for the sample design used in the breakdown tester (see Section 6). Prepared samples with both geometries are shown in **Figure 1**.

After attaching the silicone membrane, 0.2 mL of solvent was rapidly distributed over the samples with a syringe and a plastic spatula. The samples rested for 1 min at room temperature (climatized room at 22 °C and 55% relative humidity) and were then placed in an oven for 60 min at 150 °C. The time durations were chosen to replicate typical operating times in the screen-printing process. In fact, when the electrodes were screen-printed, the ink had about 1 min time to interact with the membrane before they were put into the oven at 150 °C to cure for 60 min.

2.2.2. Screen-Printing Electrodes on Silicone Membrane

The DETs examined in this work were prepared as follows: 1) The silicone membrane was transferred unstretched to metal

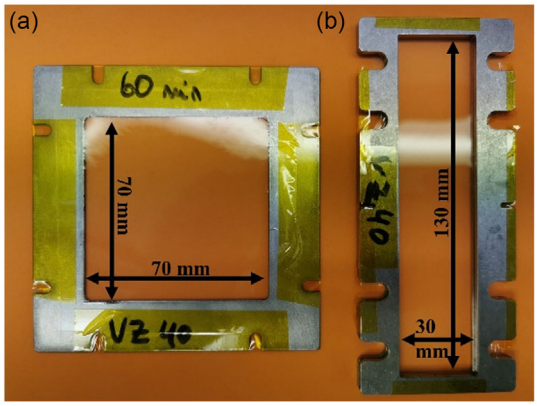

Figure 1. a) Sample geometries for Elastosil 2030 PDMS film treated with pure solvent for optical measurements, and b) breakdown tests.

Table 1. Used solvents and their vapor pressure, evaporation rate,[a)] and main components as described in the corresponding data sheets.

Company	Product name	Vapor pressure [hPa] @ 20 °C	Evaporation rate[a)] [–]	Main components
SunChemical Parsippany, NJ, USA	VD10	11	0.814	1-Methoxy-2-propanol
SunChemical Parsippany, NJ, USA	VD40	Not specified	0.3	CyclohexanoneAromatic hydrocarbons, C9
SunChemical Parsippany, NJ, USA	VD60	0.31	0.03	2 – Butoxyethyl acetate
SunChemical Parsippany, NJ, USA	VZ40	Not specified	<1	2-(2-Ethoxyethoxy)ethyl- acetate
Wacker Chemie AG Munich, Germany	Belsil DM 1 Plus	7.10	Not specified	OligodimethylsiloxanePolydimethylsiloxane
ExxonMobil Irving, TX, USA	Isopar M	0.1	0.01	Isoalkanes, C12–C16, cyclicIsoalkanes, C11–C13

[a)]In comparison to butyl acetate.

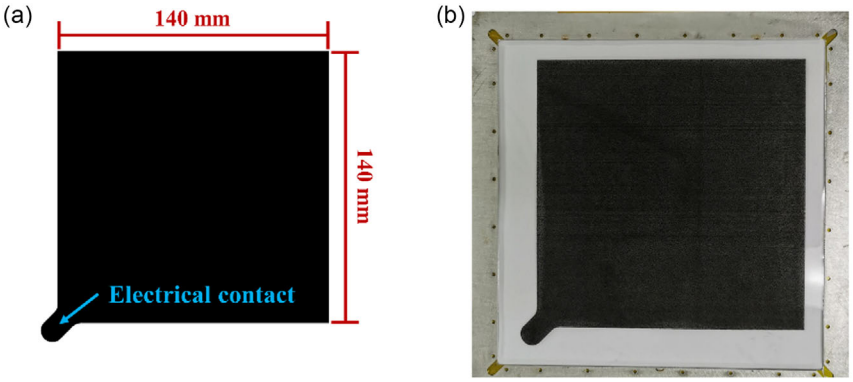

Figure 2. a) Schematic electrode geometry: a square with the edge length of 140 mm completed with a contact region in the lower-left corner; b) image of an actual screen-printed electrode with this design.

frames with a free area in the middle of 171 × 171 mm; 2) CB was briefly mixed into one of the solvents shown in Table 1. Afterward, a two-component silicone (Silgel 612 A/B, mixing ratio 1:1) was added. The mixture was homogenized by milling (three-roll mill from EXAKT, Norderstedt, Germany), and subsequently mixed using a Thinky planetary mixer (Laguna Hills, CA, USA; 2000 rpm for 1 min). The composition of the ingredients was 3.4 wt% CB, 19.2 wt% silicone, and 77.4 wt% solvent. This paste was used as the screen-printing ink; 3) A SEFAR (Heiden, Switzerland) 90/55 PET screen (90 polyethylene terephthalate threads/cm and 55 µm thread diameter) was used for screen-printing the electrodes. The choice of the screen parameters and the screen-print parameters were based on our prior research.[30] These parameters were kept constant for all manufactured electrode compositions. The electrode geometry used in the conducted investigations was a square with an edge length of 140 mm, completed with an ellipsoidal strip in the lower-left corner used for the electrical connection. A sketch of the electrode geometry and a picture with the printed electrode is shown in **Figure 2**a,b, respectively; and 4) After screen-printing, the electrodes were cured in an oven for 60 min at 150 °C.

3. Results

In this section, the results from the conducted measurements are presented and discussed. A detailed explanation of the conducted measurements can be found in Section 6. In Section 3.1, the results from the investigations on the membrane treated with pure solvent are displayed. These are followed by the discussion of the results for the samples with printed CB electrodes in Section 3.2.

3.1. Results for Pure Solvent on Silicone Membranes

This section displays the results from the experiments conducted on the pure film with and without solvent treatment.

3.1.1. Optical Analysis and Microscopy

Samples were prepared as described in Section 6. **Figure 3** shows the pictures that were taken at different states during the solvent and heat treatment. In the first row, the silicone membranes in the initial state are shown. In the second row, the samples are shown 1 min after the solvent was applied. In the third row, they are shown after a 60-min heat treatment at 150 °C. The fourth and final row displays the light microscope images of interesting spots (highlighted with light blue circles in row 3).

The interaction between solvent and silicone membrane differed significantly when different solvents were applied. After solvent application, VD10 and VZ40 formed droplets, showing no notable interaction between solvent and PDMS. Small droplets are segregated to bigger rounder ones at ambient conditions, thus minimizing their surface energy. In comparison, all other solvents showed swelling effects. VD40 and VD60 penetrated the film, inducing wrinkles. In these wrinkles, small droplets of solvent could be seen. Belsil and Isopar M induced swelling on a larger scale than VD40 and VD60. They even spread themselves over the sample. This indicates a higher level of interaction for these two solvents, allowing faster and easier penetration of the PDMS membrane.

The different behavior of the solvents on PDMS thin membranes can be explained by analyzing the chemical structure of the solvents. The swelling behavior of different organic solvents with PDMS bulk samples has already been studied by Whitesides et al.[35] In the present work, small amounts of commercially available solvents (which consist of a mixture of different chemicals) are put onto the PDMS membrane and then the samples are placed into the oven for solvent evaporation, while Whitesides et al. investigated bulk PDMS samples in different pure organic solvent immersions for 24 h until saturation is reached. Even though their swelling regime differs from the given one, their results help to explain the observed phenomena in this work. Whitesides et al. showed a correlation between swelling and Hildebrand solubility parameter δ_H, which depends on the cohesive energy density of different solvents. They established the relationship that solvents with a solvent parameter close to that of PDMS cause stronger swelling than those with a different δ_H. However, this relationship is not linear and differs for each polymer-solvent system. Thus, in addition to δ_H, polarity also influences the swelling behavior. For solvents with polar or hydrogen bonding properties, δ_H is not a suited parameter to derive the swelling tendency and the dipole moment must be

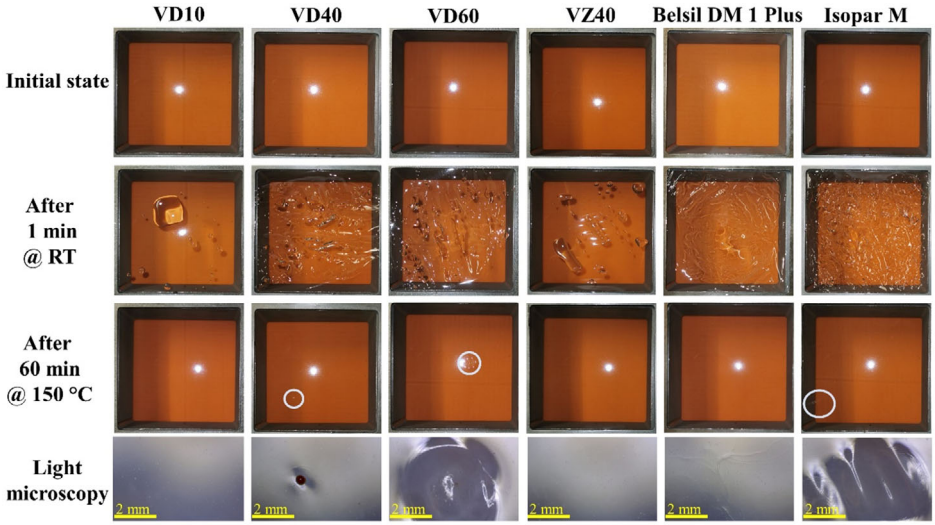

Figure 3. Images of samples at different times in the treatment procedure: row 1: silicone membranes in the initial state before applying the solvents; row 2: 1 min after applying the solvents and resting at ambient conditions (20 °C and 55% relative humidity); row 3: after 60 min in the oven at 150 °C. Interesting spots are highlighted with light blue circles; row 4: images of interesting spots taken by light microscopy after heat treatment.

included in the consideration. In this context, Whitesides et al. distinguished between low solubility, moderate solubility, high solubility, and extreme solubility.

As the δ_H and polarity values are not known for all solvents described here, they are classified based on structural similarity of their main components to the solvents studied by Whitesides et al. Most alcohols can be classified in the low solubility group. Thus, VD10 (with alcohol as the main component, see Table 1) showed no interaction with the silicone matrix. Alkanes, in contrast, are completely nonpolar and lead to good solubility. This was confirmed by the high swelling of Isopar M. Belsil shows the same structural units as PDMS and is therefore the solvent with the highest swelling capability. The main components of VZ40 and VD60 differ from each other only by one ether group. In general, ethyl acetates show mainly low solubility, while others exhibit good swelling. Consequently, VZ40 should show better solubility than VD60. However, the microscopy images indicate that VZ40 showed almost no penetration of the PDMS. This suggests that other additives in the solvent affect the interaction between solvent and matrix. VD40 also showed swelling, like VD60. Due to the increased polarity of the cyclohexanone, less swelling would initially be expected. However, solvent naphtha (aromatic hydrocarbon solvents) leads to increased penetration. The increased swelling of VD40 can be explained by the mixture of those compounds.

In general, using optical detection and comparison with the literature, the following order of increasing solvent swelling in the PDMS matrix can be determined

$$\text{VD10} < \text{VZ40} \ll \text{VD60} < \text{VD40} \ll \text{Isopar M} < \text{Belsil DM 1 Plus} \quad (1)$$

The solvents could be removed from the matrix after swelling of the PDMS. However, the process of swelling and deswelling may cause deformation due to nonuniform evaporation and the formation of stress in the polymers. The appearance of the matrix after treatment at 150 °C for 60 min is shown in row 3 in Figure 3. After heat treatment, the solvents which did not interact with the silicone membrane, i.e., VD10 and VZ40, showed no noticeable spots, and no changes to the membrane could be distinguished. Belsil also did not show any anomalies when looking at the whole film. However, when performing investigations with the light microscope, circle-like structures resembling water stains could be detected. Due to the similar chemical structure of Belsil and silicone, one would not expect a chemical change in the silicone film. The stains were most likely the aftermath of the swelling, but they were small and barely visible. Therefore, it is assumed that there was no significant harm done to the membrane. VD60 and Isopar M showed some spots distributed over the samples. Due to the images taken with the light microscope, it is assumed that in these spots solvent which penetrated the membrane could still have been present. Especially for Isopar M, these spots looked shiny, indicating remains were present in the membrane. One reason why these spots may be occurred for VD60, and Isopar M is that they have the lowest vapor pressure (compare Table 1), and therefore require more time to evaporate. In addition, molecules which diffuse in the silicone film also need to diffuse to the surface again before vaporizing. Therefore, it is reasonable that such spots could be seen for these two solvents in comparison to the other ones. Lastly, on the films treated with VD40, one could see orange/red spots. Those resembled little droplets on the surface of the film. When slightly touching them with a spatula, they could be taken off the film. Smaller spots seemed to be hard, while bigger spots had a hard shell and the inner of these droplets was highly viscous. As these droplets could be easily removed without leaving residues on the film, the dielectric film seemed not to be harmed by the process, leading to these orange/red solvent residues which must be a consequence of the thermal treatment of the solvent. However, these droplets could lead to imperfections in the

electrode and affect the actuation properties. Therefore, VD40 is not suitable for the applied manufacturing process.

In summary, it can be concluded that the contact of the membrane with the solvents induces swelling, depending on the polarity and chemical structure of the solvent. In the following section, ATR–FTIR spectroscopy is used to verify the optical results to analyze whether there is still solvent trapped in some samples, and to evaluate if chemical aging occurred in the PDMS matrix due to interaction with the solvents.

3.1.2. ATR–FTIR

To evaluate the chemical structure of the samples before and after treatment, ATR–FTIR spectroscopy measurements are carried out. **Figure 4** shows the results of these measurements. As a reference, the Wacker Elastosil 2030 membrane was measured in the untreated state. The spectrum of this reference is shown in green in all IR spectra. It should be noted that, in most cases, the Elastosil reference spectra (green) nearly fully overlap with other spectra and are therefore not always easily visible. In addition to the Elastosil reference, the pure solvents were measured, and their spectra were put into the corresponding graph as a blue spectrum. Due to the similar chemical structures in Belsil and the PDMS film, the spectra of Belsil and Elastosil overlay each other. As a result of the two references (solvent and untreated PDMS membrane), it can be directly distinguished if the bands in the spectra of the treated samples belong to the solvent, belong to the untreated/unaged PDMS membrane, or if they are different bands, which occur, e.g., because of a chemical reaction or altering effects. For the membranes treated with solvent, two measurement spots are chosen and displayed. Specifically, a random spot treated with solvent was measured and is shown as a black dashed line. Additionally, spots which showed deformation or optical conspicuities in the silicone matrix in the light microscope measurements ("interesting spots" as shown in Figure 3), were specifically measured to verify the assumption that solvent residuals were still left at these spots and to review if chemical changes occurred in the PDMS matrix. These spectra are shown in red in Figure 4. Since samples treated with VD10 or VZ40 showed no interesting spots (compare Figure 3), additional random spots were measured to verify if solvent residuals can be detected. For all sample types, the spectra of the random measured spots (black dashed lines) overlay with the spectrum of the silicone reference, thus no chemical aging is observed. For Belsil and VD60, at interesting spots, no additional bands in comparison to the Elastosil spectrum occur, thus indicating that neither solvent was present in the silicone matrix nor that chemical aging occurred even at the deformations seen in the light microscopy images. For Isopar M, at these optical conspicuities, differences between its spectra and the spectra of the untreated silicone membrane can be seen. As all these bands can be assigned to the spectra of the pure solvent, at these spots, it can be concluded that solvent residuals were present in the matrix. Nevertheless, no bands occur, which cannot be assigned to either the membrane or the solvent. Therefore, also for Isopar M, no chemical aging of the silicone membrane can be observed. For VD40, at a random measured spot, the spectrum shows only bands, which can be assigned to the PDMS membrane. Therefore, these random spots show no chemical aging or solvent residuals. At interesting spots of VD40 treated samples (orange/red spots, compare Figure 3), bands were observed, which cannot be assigned to the PDMS or the pure solvent. Since these spots could be removed with a spatula without residuals and additionally random spots showed no chemical aging, it can be assumed that the PDMS was not involved in the chemical reactions happening during the thermal decomposition of VD40 and remained unaffected. The orange/red spots were decomposition products of VD40, which seem to not be stable at 150 °C.

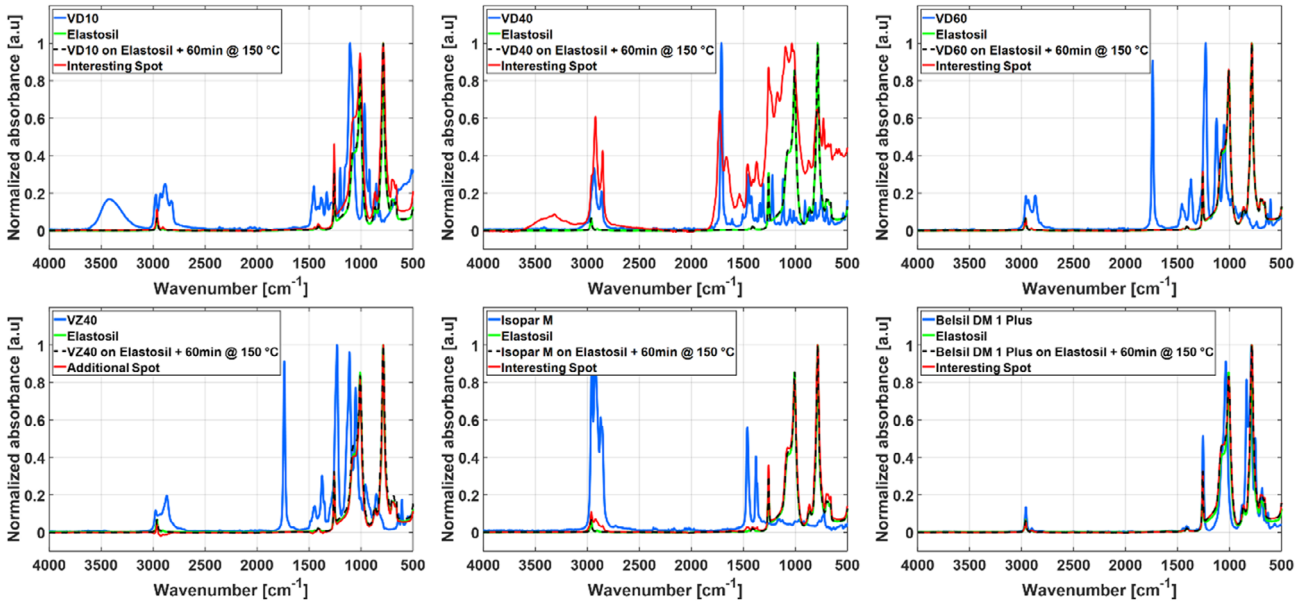

Figure 4. ATR–FTIR spectra for solvents (blue), untreated silicone membrane (green), random spot of treated samples (dashed black line), and interesting spots seen under light microscopy (red) for the different solvents used to treat the silicone membrane.

As the composition of the solvents is not fully known, no precise statement can be made about the decomposition products and the decomposition mechanism. However, VD40 should not be used for this specific manufacturing process due to its thermal instability.

Using ATR–FTIR spectroscopy, it could be shown that the matrix did not undergo any chemical changes due to contact with solvents and subsequent heat treatment. Nevertheless, solvent residues were detected in samples treated with Isopar M and deformations of the PDMS membranes could be seen under light microscopy for samples treated with Isopar M, Belsil, and VD60. For VD40, after heat treatment, orange/red spots can be found, which are shown to be decomposition products of the solvent. These residues, deformations, and decomposition products may influence the properties of the final DETs. This influence is investigated in the following sections by characterizing the breakdown strength of the treated silicone membranes and subsequently by characterizing printed electrodes with different solvents.

3.1.3. Breakdown Strength of Silicone Membranes Treated with Pure Solvents

To evaluate if the solvents influence the breakdown behavior of the silicone membrane, the breakdown strength of the silicone membranes treated with solvents was investigated. A total number of 66 measurement points were tested for each configuration, by considering constant environmental conditions of 20 °C and 55 % relative humidity, as described in Section 6. **Figure 5** displays the results of the breakdown tests.

The untreated silicone membrane was also characterized by its breakdown strength as a reference. For all sample types, the median breakdown field was between 112 and 114 V μm^{-1}. These breakdown fields fit to results of breakdown measurements done on the same dielectric material membrane conducted in other works,[45] and fit to the breakdown strength specified by the manufacturer. As the deviations between sample types are in the range of standard deviation, and as they are in the same order to the breakdown field of the untreated reference, the pure solvents used in this study did not influence the breakdown behavior of the silicone membrane. In this respect, it can be concluded that all solvents investigated are suitable for manufacturing DETs. Possible behavior changes due to the addition of CB in the electrodes will be investigated in Section 3.2.

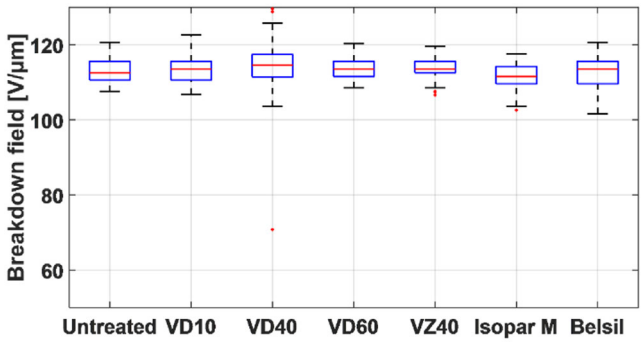

Figure 5. Breakdown fields of the untreated silicone membrane and of the solvent-treated silicone membranes.

3.2. Results for DETs with Screen-Printed Electrodes

The following sections focus on the characterization of the printed electrodes containing different solvents, manufactured as described in Section 2.2.2. In the first step, their screen-print image is visually analyzed and the influence of the solvents on the visual image is discussed. Afterward, the electrical resistance of the electrodes is measured and its influence on the breakdown strength of the silicone membrane is investigated.

3.2.1. Screen-Print Image

Figure 6 shows one sample per sample type after curing. Printing images differ significantly. Using VD10 and VZ40 resulted in a very thin layer of electrode material, allowing light to shine through the electrode and giving a gray image. VD60 samples showed a denser layer of electrode material and therefore darker electrodes, while the print image was inhomogeneous. VD40, Belsil, and Isopar M also showed a much denser layer of electrode material, but here the electrodes were homogeneous and even darker than for VD60. It is found that solvents that penetrate the silicone film and cause swelling/wrinkling result in a better printing image. This can be explained by the interaction of the solvents with the silicone film, as the greatest amount of the printing material was solvent (as stated in Section 2.2.2, the printing material consists of 7.4 wt% solvents). The influence of the compatibility between solvent and dielectric film while screen-printing is schematically shown in **Figure 7**.

A higher compatibility leads to a higher swelling of the dielectric material and thus better adhesion of the electrode material to the PDMS membrane. If this compatibility either decreases or is absent (e.g., for VD10 and VZ40), the adhesion of the electrode ink to the PDMS membrane decreases. The adhesion of the ink to the screen then exceeds that of the adhesion to the membrane, which results in the ink remaining in the screen rather than on the dielectric membrane. This leads to a thinner printed electrode containing more imperfections.

This phenomenon is reflected in the thickness measurements. The mean thicknesses and standard deviations are shown in **Table 2**.

The average thicknesses are in the same order of magnitude ranging from 4.5 to 5.6 μm, but the standard deviations differ significantly. The enlarged microscopic images of VD10 and VZ40 in Figure 6 illustrate a very inhomogeneous print pattern with areas not covered with CB at all. Since these solvents do not interact with the silicone membrane, most of the printing material remains on the screen. The ink staying on the silicone membrane forms small droplets, similar to shown in Figure 3. After curing, this leads to an inhomogeneous electrode distribution with areas covered with electrode material (black) and areas without any electrode material (transparent). Thus, the standard deviation of the electrodes with the solvents VD10 and VZ40 exceeds the standard deviations of the other four electrodes. The enlarged pictures of the other electrodes in Figure 6 show a much more homogenous distribution of the electrodes, while resembling the

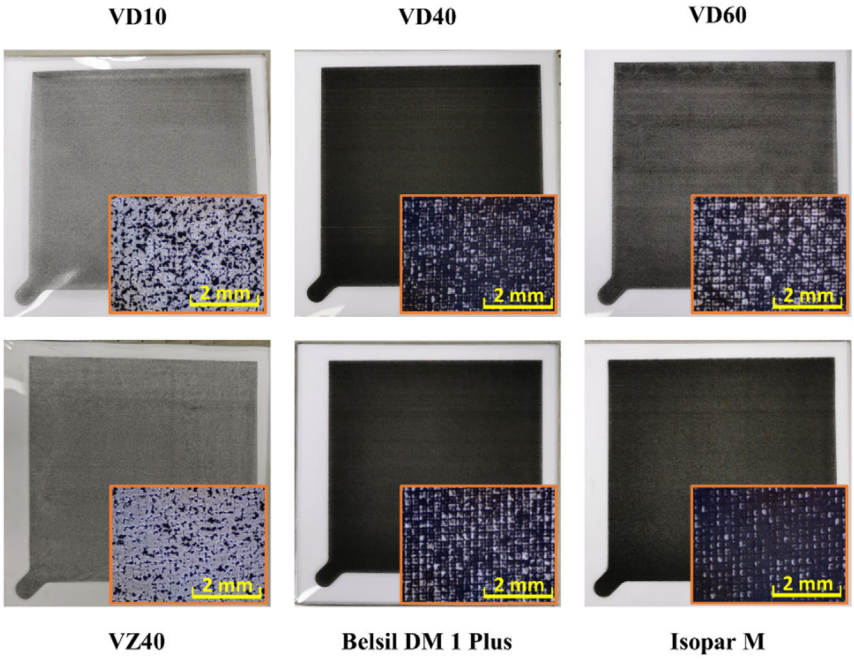

Figure 6. Photos of screen-printed electrodes and enlarged microscopic images of the electrodes printed with different solvents.

mesh structure of the screen, which has already been seen in prior research.[30] These electrodes are on average the same thickness as the ones printed with VD10 and VZ40, but their standard deviation is far lower due to the homogeneous printing image.

It is clearly shown that the screen-print image as well as the homogeneity of the electrodes significantly depend on the used solvent. The different screen-print images are most likely to influence the characteristics of the DET. Therefore, in the following sections, the printed electrodes are investigated regarding their electrical resistance and their influence on the breakdown strength.

3.2.2. Electrical Resistance of the Screen-Printed CB-Electrodes

One important aspect of electrodes for DETs is their electrical resistance, as stated in Section 1. The results of the resistance measurements (as described in Section 2.2.3) are listed in **Table 3**.

The electrical resistances vary significantly between sample types, reflecting the correlation between resistance and quality of the printing image. The worse the printing image, the higher the electrical resistance. The printing image for the solvents VD10 and VZ40 was inhomogeneous, and no sufficient electrical conductivity was achieved. The resistance was in the range of megaohms, thus it is not suitable for DET applications. Looking at the microscopic images in Figure 6, it can clearly be seen that due to the agglomeration of the electrode material while screen-printing, the number of conductive paths decreased significantly in the final electrode as the areas with electrode material are in most cases not connected to each other, thus leading to a bad conductivity. In comparison, VD60 was better screen-printable, and the electrical resistance was with an average of 133 kΩ lower than for VD10 and VZ40. The inhomogeneous printing image seen in Figure 6 can also be seen in the electrical resistance for VD60. In the printing direction, the resistance was lower than perpendicular to it, which resulted in the higher deviation in the electrical resistance for VD60. VD40, Belsil, and Isopar M, which all showed a thick and homogenous printing image, the electrical resistances were significantly lower than for the other sample types and were not dependent on the printing direction. On average, the electrical resistance values equal 32, 22, and 20 kΩ for VD40, Belsil, and Isopar M, respectively. The slightly higher electrical resistance of VD40 can be explained by the weaker interaction between VD40 and PDMS film, in comparison to Belsil and Isopar M, as shown in Figure 3. In summary, the electrical resistance reflects the results from the screen-print image, demonstrating that the choice of the right solvent is fundamental for achieving a desirable electrode resistance characteristic.

3.2.3. Breakdown Strength of Dielectric Films with Screen-Printed CB-Electrodes

Besides the electrodes electrical resistance, the influence of the printed electrodes on the breakdown strength of the dielectric membrane was characterized as well. The measurement method differed from the one used for the dielectric film without electrodes, as explained in Section 6. For the determination of the breakdown strength of the silicone membrane after electrodes were applied, 30 measurement points were measured per sample type. The results of the breakdown strength are shown in **Figure 8**.

The median breakdown fields for the different samples vary only in a range of 2 V μm^{-1} and therefore in the standard deviation range. No significant difference between the solvents can be

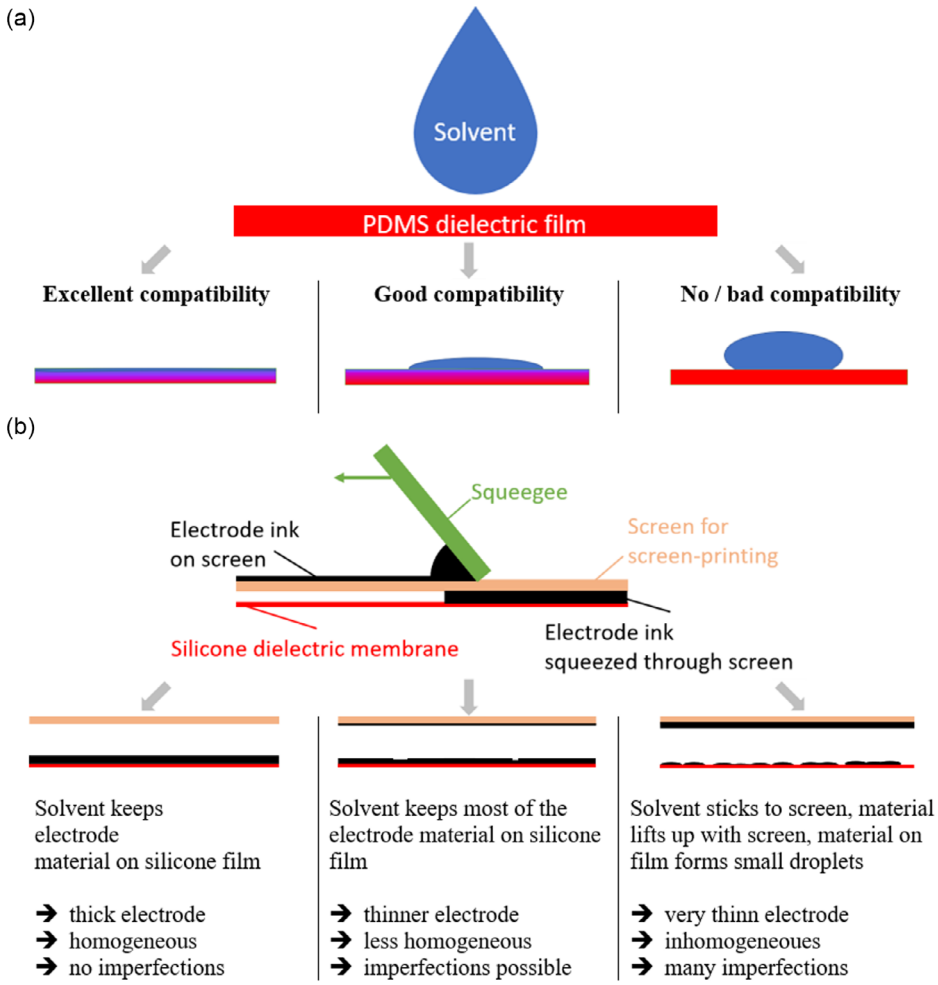

Figure 7. a) Influence of compatibility between solvent and PDMS dielectric film when pure solvent is put on the film, and b) when using the solvent in the printing material for screen-printing. It is schematically shown how an excellent, a good and a bad compatibility between solvent/printing material and dielectric film influences the behavior while screen-printing.

Table 2. Electrode thicknesses and standard deviations of electrodes printed with various solvents.

Used solvent	Electrode thickness [µm]
VD10	4.5 ± 7.0
VD40	5.6 ± 3.1
VD60	5.5 ± 2.8
VZ40	4.7 ± 4.8
Isopar M	5.1 ± 2.5
Belsil DM 1 Plus	4.9 ± 3.0

Table 3. Average of 12 electrical resistance measurements (6 samples with each 2 measurements) of printed electrodes with different solvents.

Used solvent	Electrical resistance [kΩ]
VD10	>1000
VD40	32 ± 14
VD60	133 ± 29
VZ40	>1000
Isopar M	20 ± 4
Belsil DM 1 Plus	22 ± 3

determined. When comparing the breakdown strength of the samples with printed electrodes to the membranes without electrodes (see Figure 5), it can be noted that the breakdown strength dropped significantly when an electrode was applied (from a median value of 113 V µm^{-1} to around 90 V µm^{-1}). The fact that the application of an electrode decreases the breakdown strength was already documented in earlier literature.[45] From the results shown in Figure 8, it can be concluded that different solvents do not influence the breakdown strength in a different way. Combining this result with the ones in Figure 5, where it is shown that the pure solvents do not decrease the breakdown strength of the silicone membrane, it can be concluded that

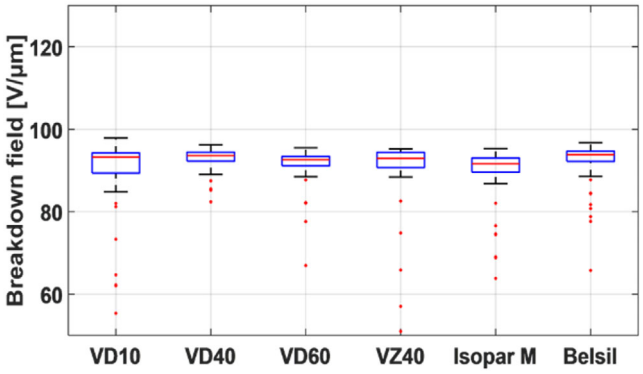

Figure 8. Breakdown fields of the silicone membrane after applying screen-printed CB-electrodes manufactured with different solvents.

the mechanism leading to the drop of breakdown strength after applying an electrode is independent of the used solvent. The mechanisms which lead to the drop in the breakdown strength after applying an electrode require further investigations. However, since this work focuses on the influence of different solvents, and in this case, no difference for varying the type of solvent can be observed, this aspect is not further discussed at this point. Regarding only the breakdown strength measurements, all investigated solvents would be suitable for manufacturing DETs with the screen-printing process.

4. Summary

The results from Section 3show that the physical interaction between the silicone membrane and solvent is very important for the characteristics of the DETs, especially considering the electrical resistance and the screen-print image. When using solvents for the manufacturing process for DETs, many parameters must be considered. In **Table 4**, characteristic parameters – discussed in the previous sections – are listed, assessed for their importance, and then rated with a point system which is explained in the supporting information Section S3. This rating provides a fast overview of the suitability of these six solvents for the screen-printing manufacturing process and application of a DET.

The investigation shows that VD10 and VZ40 are not suitable for the screen-printing manufacturing process on the given PDMS membrane. Their total score is way below the ones of the others, as they show insufficient results in different categories. Concerning VD40, as it is not chemically stable in the investigated process, and since this is an essential criterion, the use of this solvent is not recommended for the DET manufacturing process. VD60 scores higher than the already discussed solvents, and the results are good enough for DET applications. Therefore, for the given system, VD60 could be used for DET manufacturing. However, the best results achieved Belsil and Isopar M. Due to the high vapor pressure of Belsil, the usage time is limited. Isopar M showed some solvent residuals in the membranes treated with pure solvents. Although no influence on the breakdown strength could be seen, these residuals could potentially alter the performance of other DET characteristics, which were not further investigated in this work. In summary, Belsil and Isopar M exhibited the best results in the conducted tests and are therefore recommended for screen-printing electrodes on a PDMS membrane for DET manufacturing, whereas Belsil is recommended for shorter and Isopar M for longer printing sessions.

Table 4. Rating of the investigated properties for different solvents.

Weighting	Characteristic	VD10	VD40	VD60	VZ40	Belsil	Isopar M
6	Chemical stability	54	6	54	54	54	54
5	Screen-printability	5	45	25	5	45	45
4	Breakdown strength	36	36	36	36	36	36
3	Electrical resistance	3	27	15	3	27	27
2	Vapor pressure	2	2	10	18	2	18
1	Optical conspicuities	9	1	5	9	5	1
Points		109	117	145	125	169	181

5. Conclusion

This article investigates the influence of six different commercially available solvents on the material behavior of dielectric elastomer sensors and actuators. The used dielectric elastomer is based on a PDMS membrane, while the electrodes consist of PDMS as matrix material which is filled with CB and applied to the dielectric by screen-printing. The influences of the solvents are discussed in regard to the dielectric, the electrodes, and the manufacturing process itself. As described in this article, different solvents result in different intensities of physical penetration on the PDMS dielectric. While treating the PDMS membrane with pure solvent, some solvents penetrate the membrane deeply, while others do not penetrate at all. The order of physical interaction between solvent and dielectric film is VD10 $<$ VZ40 $\ll$ VD60 $<$ VD40 $\ll$ Isopar M $<$ Belsil DM 1 Plus. Solvents with lower polarity show a deeper interaction with the PDMS membrane. However, even for solvents which show deep penetration of the membrane, no chemical aging of any of the membranes occurs. Especially in combination with the screen-printing process, better physical compatibility between solvents and membrane leads to a better screen-printing image and a higher conductivity of the electrodes. Additionally, neither the breakdown strength of the dielectric treated with pure solvent nor the breakdown strength of the dielectric with screen-printed CB electrodes is influenced by the variation of the used solvent. It is shown that the solvents in the manufacturing process and their compatibility with the dielectric material influence the characteristics of the DETs significantly, even though the solvents are no longer present in the DETs after curing. From the results gathered in this study, the following solvent properties are required: 1) A good physical compatibility with the dielectric membrane leads to a better adhesion of the electrode material to the dielectric in the screen-printing process; 2) A low vapor pressure for long manufacturing times; 3) No chemical altering of the dielectric film or the components of the electrodes; and 4) Fitting solvent properties to the manufacturing conditions so that, e.g., thermal degradation does not occur.

Out of the six investigated solvents, Belsil DM 1 Plus and Isopar M are the ones which showed the best results for the observed characteristics and are thus recommended for the manufacturing of DETs via the screen-printing method.

The six investigated parameters, namely, chemical stability, screen-printability, breakdown strength, electrical resistance, vapor pressure, and optical conspicuities, are important parameters for the applicability of solvents in the screen-printing manufacturing process for DETs. Nevertheless, additional important DET parameters such as electrode thickness, electrode influence on the mechanical behavior, or long-term stability of DETs need to be investigated in regard to the used solvent. Desired DET characteristics could also be achieved by combining different solvents, rather than using a single one. These investigations would go beyond the scope of this article and will therefore be investigated in future works. This work provides a first guidance and fundamental knowledge of how DET characteristics can be affected by various solvents during the screen-printing process. While the results here are specifically for the screen-printing process, some of these might also be transferred to other manufacturing methods, e.g., pad-printing. In conclusion, this work highlighted the importance of the consideration of all materials required for a manufacturing process and not only the materials present in the final product.

6. Experimental Section

This section provides an overview of the conducted tests and testing devices used. Firtsly the experiments conducted on silicone membranes treated with pure solvent are described, while afterwards the experiments for the characterization of the DETs after screen-printing the electrodes are described.

Experiments on Silicone Membranes Treated with Pure Solvent: Samples were prepared as described in Section 2.2.2. Photos of the samples were taken before applying the solvent, after waiting 1 min at ambient conditions and applying a subsequent heat treatment in the oven (60 min at 150 °C). After this heat treatment, the samples were investigated with a light microscope (digital microscope YW MS3200D, Di-Li Distelkamp-Electronic, Kaiserslautern, Germany). Subsequently, they were investigated by ATR–FTIR spectroscopy (Vertex 70 spectrometer, Bruker Optics, Ettlingen, Germany; see Figure S1a in the Supporting Information), which is a widely used method to characterize polymer films. Based on ATR–FTIR spectroscopy, it is possible to analyze the surface of polymer films and obtain structural information about the composition of the matrix or structural changes.[38,42,43] The solvent-treated polymer films were pressed onto the ATR crystal with a stamp and measured at points of interest, as well as at points that showed no abnormalities. Solvent residues and changes in the chemical structure of the matrix or in the environment of the samples can be detected. The working principle is schematically shown in Figure S1b in the Supporting Information.

After preparing samples with the second geometry 130 × 30 mm, they were tested in an automated breakdown tester (see Figure S2 in the Supporting Information) located in a climate chamber to allow controlled environmental conditions (here 20 °C and 55% relative humidity), validated in prior research.[44] With this setup, the breakdown strength of the dielectric without (screen-printed) conductive electrodes could be determined. Two gold electrodes were brought into contact with the silicone membrane and an electric field was applied to the membrane, while the voltage increased with a slope of 500 V s^{-1} until dielectric breakdown occurred. In this way, 11 measurement spots could be tested per sample.

Experiments Conducted on DETs with Screen-Printed Electrodes: Electrodes were first rated by their screen-printing image. Afterward, the electrical resistance was measured across and perpendicularly to the screen-print direction (see Figure S3a, Supporting Information). Measuring electrical resistance is preferred over measuring electrical resistivity, because of two main reasons. First, in an application scenario, the electrical resistance of the system is a more meaningful quantity than the intrinsic resistivity. Additionally, by measuring along two different paths, direction-dependent differences in the resistance occurring due to the manufacturing process were observable. By measuring the resistivity, e.g., with the Van-der-Pauw method, such influences would be lost. After measuring the resistance of the electrodes, the samples were placed into a specially designed breakdown box, see Figure S3b, Supporting Information. A metal plate on the bottom acts as the counter electrode to the CB electrode. A voltage ramp signal with a constant increase rate of 500 V s^{-1} was applied until breakdown occurred. Compared to the test method for pure silicone membranes, here the whole printed area of the film was simultaneously measurable. After breakdown, the frame was lifted, and a piece of tape was put on the metal plate which corresponded to the breakdown spot in the silicone membrane. The tape prohibited a current flow through the breakdown hole, thus allowing to measure the same sample multiple times. Lastly, the electrode thicknesses were measured with two confocal sensors (confocalDT IFS 2405, Micro-epsilon, Ortenburg, Germany) as described in previous research.[46]

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

dielectric elastomer transducers, material characterization, screen-printing, smart materials, solvents

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