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PVB, PVAc and PS pressure sensors with interdigitated electrodes

K. Arshak [a,*], D. Morris [b], A. Arshak [b], O. Korostynska [a], E. Moore [a]

[a] *Department of Electronics and Computer Engineering, University of Limerick, Limerick, Ireland*
[b] *Department of Physics, University of Limerick, Ireland*

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Abstract

Miniature sensors, which are rugged and reliable in nature, are essential to meet the growing demand for environmental and biomedical monitoring systems. Developing such sensors using screen-printing and drop coating techniques allows the combination of two cost effective and flexible technologies. Using this approach, a wide variety of materials could be combined to produce sensors with the desired physical properties. Silver electrodes were screen-printed onto alumina substrates, while polymer/carbon-black composites based on polyvinyl butyral, polyvinyl acetate and polystyrene were deposited by drop coating. The addition of surfactant resulted in a more even dispersion of carbon-black throughout the composites, which was observed by TEM. The frequency dependence of the composites also confirmed this result. Pressure was applied using a Lloyd Instruments LR50k in the range 0–2500 kPa and the sensitivity was taken to be the slope of the graph. It was found that the materials displayed a high sensitivity to pressure with good repeatability. Detailed studies of the composites temperature dependency showed that each material has a negative temperature coefficient and were particularly sensitive when the temperature was brought below room temperature. This was improved by coating the sample surface.

Keywords: Interdigitated capacitors; Polymer/carbon-black composites; Pressure sensors; Drop coating; Temperature effects

1. Introduction

Capacitors with interdigitated electrodes have been used for some time in applications such as voltage tunable microwave devices, slow wave devices and integrated optical modulators and deflectors [1,2]. Their popularity is mainly due to advantages including ease of fabrication, flexibility in design, cost effectiveness, no moving parts, less complex packaging constraints, one-sided access to the sensing layer and control of the signal strength [3,4].

More recently, their popularity in strain and pressure sensing applications has also increased [4–6]. For these applications it is possible to fabricate reliable and miniaturized devices, which could potentially be used in biomedical or environmental monitoring.

Perhaps the most popular method of fabrication for strain and pressure sensors is the use of silicon-based or microelectrical mechanical system (MEMS) technology. While these devices have high sensitivity and accuracy and there is a large potential for future development, their inability to withstand unfavourable or harsh environments and the high manufacturing costs are a disadvantage [7]. Furthermore, there are limitations on the type of materials, which can be used in an IC foundry [8].

Thick film and drop coating are alternative technologies, which are capable of producing cost effective, rugged and reliable sensors. Both processes are extremely flexible and a wide variety of materials and designs can be used to achieve the desired sensor properties. Thick film technology depends on a process known as screen-printing and has been used for the commercial production of devices since the 1960's [9]. A variety of devices including strain, pressure, gas, temperature, radiation and magnetic sensors have been fabricated in this way [5,6,10–12].

In drop coating, a microlitre pipette is used to deposit a specially formulated paste onto prepared substrates. The technique is often used in the preparation of samples for electrochemical studies and the deposition of nanosphere solutions onto the desired substrates in nanoscale lithography [13,14]. It is a cost effective process and providing the polymer can be put into

* Corresponding author. Tel.: +353 61 202267; fax: +353 61 338176.
E-mail address: khalil.arshak@ul.ie (K. Arshak).

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solution, there are few restrictions on the type of polymers that can be used. It is thought that the combination of drop coating and screen-printing could produce an alternative to silicon devices, which are suited to environmental and biological measurement applications.

For interdigitated capacitors, changes in the output are caused by deformation of the dielectric layer. Therefore, polymers are preferred for pressure sensing, as ceramic materials are non-compliant [6]. Their sensitivity can be increased through the addition of conductive fillers, for example carbon-black, as this affects the materials dielectric constant due to increased dipole interactions [15]. This is further enhanced when the material is placed under pressure.

The electrical properties of polymer/carbon-black composites are largely dependent on the volume fraction and distribution of the filler particles, by the size, shape and orientation of the phases (conductive and insulative), the interaction between them and the preparation method used [16]. There is a particularly strong dependence on the volume fraction of filler in the composite. Polymers are insulative in nature due to the relatively low amount of available charge carriers. When the composite contains a low concentration of fillers, the conductivity is close to that of the polymer. Further increases in filler concentration lead to a sharp increase in conductivity, which may span several orders of magnitude. This point is commonly known as the percolation threshold and describes the formation of continuous conducing chains of filler throughout the polymer matrix [17]. Further increases will lead to composites with conductivity close to that of the filler material.

The electrical properties of such composites in the vicinity of the percolation threshold can be described by Eq. (1)

$$\sigma = \sigma_o(\phi - \phi_c)^t \tag{1}$$

where σ is the conductivity of the composite, σ_o the conductivity of the conducting phase, ϕ the volume fraction of filler, ϕ_c the volume fraction of filler at the percolation threshold and t is a universal exponent which typically has a value of 2 [18].

Percolation theory has its limitations, as it only describes the behaviour of the composites in the vicinity of the percolation threshold and can only be valid when the ratio of the conductivity of the two phases is infinite [18]. The general effective media (GEM) equation is a combination of percolation and Bruggeman effective media theory. It can be used to calculate the percolation threshold for any volume fraction of filler [17]. It is shown in Eq. (2)

$$\frac{1-\phi(\sigma_l^{1/t}-\sigma_m^{1/t})}{\sigma_l^{1/t}+[(1-\phi_c)/\phi_c]\sigma_m^{1/t}} + \frac{\phi(\sigma_h^{1/t}-\sigma_m^{1/t})}{\sigma_h^{1/t}+[(1-\phi_c)/\phi_c]\sigma_m^{1/t}} = 0 \tag{2}$$

where σ_m is the conductivity of the composite, σ_l the conductivity of the low conductivity or polymer phase and σ_h is the conductivity of the high or conductive filler phase. A semi-quantitative expression for the behaviour of polymer/carbon-black composites under pressure can also be extracted from the GEM theory. This is shown in Eq. (3)

$$\phi_c(P) = \frac{\phi_c(0)}{1+mP} \tag{3}$$

where P is the pressure, m is a multiplier and $\phi_c(0)$ and $\phi_c(P)$ describe the relationship between the percolation threshold and pressure. The resistance of a polymer/carbon-black composite is generally increased with pressure is applied as the filler particles are pushed together and form conducing paths which span the polymer matrix. The percolation threshold generally occurs for a volume fraction of conductive filler around 0.16 [19]. This varies depending on the degree of dispersion of the filler. Shear mixing and the use of surfactants can improve dispersion, increasing the resistivity of the sample [20]. Therefore, higher volume fractions of carbon-black are required to reach the percolation threshold, resulting in a loss in mechanical properties as the composite becomes stronger and less compliant under pressure.

However, there is some trade off involved. Poor dispersion of carbon-black throughout the polymer matrix can lead to higher frequency dependence as the voltage across the gap separating the agglomerates is expected to be higher than the macroscopic voltage, V, by a factor, M, which represents the ratio of the average size of the conducting aggregate to the average gap width [21].

To investigate the electrical and mechanical behaviour of polymer/carbon-black composites with an even distribution and high filler concentration, sensors were developed on alumina substrates with Ag interdigitated electrodes. Polymer solutions were prepared using polyvinyl butyral (PVB), polyvinyl acetate (PVAc) and polystyrene (PS). Carbon-black and surfactant were shear mixed into the solutions so that a homogenous dispersion of the filler was achieved. Transmission electron microscopy (TEM) was used to verify this for each sample, with PVAc being used as an example. The change in impedance with frequency was also used in order to observe the stability of the response as this can affect the design of suitable interface circuitry. To examine its effect on the mechanical properties of the composite, each device was then tested under pressure ranging from 0 to 2500 kPa. Finally, the effect of temperature on the polymer composites was investigated, as during operation the temperature may change. This may alter the sensor resistance, requiring compensation from the measurement system.

2. Experimental

Three polymers were explored as part of this work, PVB, PVAc and PS. Polyvinyl butyral was chosen, as it is an elastomer, which is commonly used as a binder in the formation of polymer thick film pastes for screen-printing [5]. Polyvinyl acetate is often used as a primary ingredient for the formation of glue, which is odourless, non-flammable and suitable for use at low temperatures [22]. Finally, polystyrene is a thermoplastic, whose properties can easily be transformed from brittle to soft by changing the degree of polymerization [23].

For each material, 160 mg of polymer was dissolved in Tetrahydrofuran (THF), supplied by Labscan Ltd. Following this 40 mg of carbon-black with a particle size of 12 nm, supplied