Formulation of Sol–Gel Derived Bismuth Silicate Dielectric Ink for Flexible Electronics Applications

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ABSTRACT: An agile method is demonstrated for the development of a low dielectric constant ink made from nanostructured bismuth silicate (nBSO), synthesized by a sol–gel method. The viscous ink, suitable for screen-printing, was formulated by controlling the organic additives. The viscosity as well as the dynamic viscoelastic properties of the formulated ink was tuned to yield ideal flow characteristics. The microstructure and surface roughness of the printed film were characterized, and the film shows minimum average and root-mean-square surface roughness values ($R_A = 80.3$ nm and $R_g = 102$ nm). Microwave dielectric properties of the printed dielectric film were $\varepsilon_r = 4.36$ and $\tan \delta = 1.6 \times 10^{-2}$ at 10 GHz. The apparent leakage current density of the film is of the order of $10^{-5}$ A/cm$^2$. The improved dielectric and thermal properties, combined with low leakage current, make nBSO dielectric ink a suitable candidate for use in printed electronics.

INTRODUCTION

Printed electronics serve as one of the most versatile emerging technologies, featuring low cost and high volume, for many products that we depend on for everyday life. One advantage of printed technology compared to conventional photolithography-driven electronics is the relative simplicity of fabrication.1

Within the past decade, the field of printed electronics has made rapid strides in both low- and high-performance domains like flexible displays, smart labels, decorative paints,2 antennas,3 radiofrequency identification devices,4 fuel cells,5 sensors,6 solar cells,7 thin-film transistors (TFTs),8,9 and multilayer ceramic capacitors.10 Different techniques are commonly adopted for imprinting conducting, semiconducting, and dielectric inks onto various substrates, such as screen-printing, ink-jet-printing, and gravure printing.11 The screen-printing technique, formerly known as serigraphy, is a stenciling process that was widely used in textiles and printing cards in early days. Later, the process proliferated slowly into niche areas like electronics. Currently it is the most widely used technique for fabricating ceramic thick films in printed electronics.11–13 In screen-printing, a viscous ink is squeezed through a finely woven mesh having the desired geometry onto a substrate by applying force with a rubber squeegee.

A typical ink consists of a solid filler (>60%), vehicle (typically, organic solvents), dispersant, and binder in ideal proportions to enable uniform printing and better adhesion. With screen-printing inks, drying or curing is usually achieved with the help of ultraviolet (UV) or infrared (IR) rays or local heating. In UV-curable inks, the vehicle is composed of a mixture of fluid oligomers, monomers, and initiators that, when exposed to UV light, release free radicals that cause the polymerization of the vehicle, which then hardens to a dry ink film. In the case of IR-assisted curing, the energy emitted by IR radiation is directly used to heat the wet ink film. A vast majority commercial functional inks avoid either of these radiation-assisted curing processes, instead employing local heating with the help of suitably designed flash-driers and ovens. However, oven-drying of the ink above 100 °C precludes the use of a number of flexible substrates, including paper.11 Therefore, the drying efficiency of the screen-printing ink is of key importance today in order to simplify fully automated printing techniques. A lot of research is going on this domain, aimed at developing screen-printing inks that evaporate at room temperature for use in high-performance applications.11,13

This room-temperature evaporation drying relies primarily on the vehicle, requiring a specific combination of organic solvents that evaporate soon after printing. However, the binder must be left behind in the printed film, so as to achieve maximum rub-off resistance. Developing such a fast-curing ink has always been a challenge, since the drying process should be initiated soon after ink is delivered to the substrate but not before the ink passes through the screen grids, which could result in clogging.

The development of conducting and semiconducting inks has seen an appreciable improvement in the past several years.14 A few industries like Creative Materials Inc. (USA) and...
Henkel AG & Co. (Germany) market low-κ dielectric insulating inks. At the same time, there are only a few reports available in the literature on dielectric inks. Wu et al. reported the development of a nanocomposite dielectric ink for TFT application in 2014. Recently, Jobin et al. reported the development of silica- and zirconia-based inks for printed electronics. High-κ dielectric ink has a critical role in a large area of electronics since it is a vital component in TFTs. Some other major areas where high-κ inks have potential applications include dye-sensitized solar cells, field effect transistors, piezoe- and ferroelectric devices, and solid oxide fuel cells. Low-κ ink, on the other hand, is used for insulation in flex circuits and thick films and for improving the moisture, arc track, and resistance to weathering of electrical electronic devices. The development of a dielectric ink that does not require the cumbersome arrangements needed for thermal and UV/IR curing deserves wider attention. Additionally, room-temperature curing enables one to employ a wide variety of substrates.

Bismuth silicate is a material that finds applications in many sectors due to its excellent dielectric, thermoluminescent, and photocatalytic properties. The microwave dielectric properties of bismuth silicate and its suitability for low-temperature co-fired ceramics (LTCC) applications were explored recently, and the results indicate that this material shows promising microwave dielectric properties for LTCC applications. Bismuth silicate is also successfully used in ferroelectric materials to tune the Ferroelectric modes. The variation of viscosity with the shear rate from 0 to 100 s⁻¹ was measured in rotational mode, whereas the storage modulus (G') and shear modulus (G'') were measured in oscillation mode. The as-prepared colloidal ink was screen-printed on a clean Mylar polyester film using an XPR72 semiautomatic screen-printer (EKRA, Germany). A homemade screen prepared from a 350-count mesh was used for the present study, cleaned thoroughly in order to remove any dust or ink residue trapped in the mesh. The desired shape for printing was fabricated on the mesh by a conventional photoresistive technique. The adhesion strength of the printed ink on the substrate was carried out by tape test analysis based on modified ASTM standard D3359-09. The surface homogeneity and quality of the printed film were characterized using optical microscopy (MRDX, Leica Microsystems, Wetzlar, Germany). The surface roughness of the printed pattern was analyzed using atomic force microscopy (AFM) (Multimode, Bruker, Germany). The analysis was carried out on different areas of 10 μm × 10 μm and 4 μm × 4 μm, respectively, operating in tapping mode. Microstructural analysis of the printed film was done by scanning electron microscopy (SEM) using a JSM 5600LV instrument (JEOL, Tokyo, Japan). The dielectric properties in the low-frequency region (300 Hz to 3 MHz) were studied using an LCR meter (Hioki model 3532-50, Nagano, Japan). The dielectric properties of the printed film were measured using a split post dielectric resonator (SPDR) (QWED, Warsaw, Poland) technique at different frequencies (5, 10, and 15 GHz) with the aid of an ENA series ES071C vector network analyzer (Agilent Technologies, Santa Clara, CA, USA). The I–V characteristics of the printed film were measured using a source meter (Keithley 2410, 1100 V). For this measurement, the prepared ink was printed on a thin copper film, and gold metal was sputtered on the top of it as the front electrode.

## EXPERIMENTAL SECTION

**Preparation of nBSO through Sol–Gel Synthesis.** Bismuth(III) nitrate pentahydrate [Bi(NO₃)₃·5H₂O, 99%, Merck, Mumbai, India] and tetraethyl orthosilicate [TEOS, 98%, Sigma-Aldrich, St. Louis, MO, USA] were used as the precursors for nBSO preparation. Initially, Bi(NO₃)₃·5H₂O was dissolved in acetic acid under continuous stirring. After that, a stoichiometric amount of TEOS was added drop-by-drop to the solution, and stirring was continued for 1 h. The obtained gel was dried in a hot air oven at 70 °C overnight. The dried gel was crushed, ground to a powder, and then calcined at 850 °C for 4 h to complete the phase formation. It was further ground to a fine powder using a high-energy planetary ball mill (Pulverisette 5 classic line, Fritsch GmbH, Idar-Oberstein, Germany) in water with tungsten carbide as the grinding medium. Thermogravimetry/differential thermal analysis (TG-DTA) of the dried gel was carried out from ambient temperature to 900 °C in air atmosphere using a thermogravi- metric analyzer (TGA/DTA instrument, Shimadzu, Japan). The phase purity and crystal structure of the calcined nBSO ceramic material were studied using X-ray diffraction (XRD) analysis with Cu Kα radiation (X’Pert PRO diffractometer, PANalytical, Almelo, The Netherlands). Fourier transform infrared (FTIR) analysis of calcined nBSO powder was carried out using a spectrum II FTIR spectrometer (PerkinElmer, Waltham, MA, USA) in the range 400–4000 cm⁻¹. The morphology and particle size of the nBSO powder were more systematically analyzed using high-resolution transmission electron microscopy (HRTEM) (FEI Tecnai G2 30S-TWIN, FEI Co., Hillsboro, OR, USA). For this purpose, nBSO powder was well dispersed in acetone using ultrasonication and then drop-cast onto a carbon-coated copper grid and dried. The BET surface area of the calcined powder was measured by nitrogen adsorption using a Gemini 2375 surface area analyzer (Micromeritics, Norcross, VA, USA).

**Formulation and Screen-Printing of nBSO Ink.** A dielectric ink essentially comprises ceramic filler, solvent, dispersant, and binder. In the present case, equal volumes of xylenes (Sigma-Aldrich) and anhydrous ethanol (Merck) were used as solvent. In the first step, the dispersant (2 wt% with respect to filler), Triton X-100 (TCI, Tokyo, Japan), was dissolved in the solvent mixture using an ultrasonic bath for 10 min. At this stage, the filler, nBSO powder (60 wt% with respect to solvent), was added to the mixture and stirred continuously for 6 h using a magnetic stirrer. The binder (5 wt% with respect to filler), ethyl cellulose (Sigma-Aldrich), was then added, and stirring was continued for another 24 h to get a homogeneous viscous ink. The viscosity of the ink was measured using a Rheo plus32 rheometer (Anton Paar, Ashland, VA, USA) operating in rotational and oscillation modes. The variation of viscosity with the shear rate from 0 to 100 s⁻¹ was measured in rotational mode, whereas the storage modulus (G’) and shear modulus (G’’) were measured in oscillation mode. The as-prepared colloidal ink was screen-printed on a clean Mylar polyester film using an XPR72 semiautomatic screen-printer (EKRA, Germany). A homemade screen prepared from a 350-count mesh was used for the present study, cleaned thoroughly in order to remove any dust or ink residue trapped in the mesh. The desired shape for printing was fabricated on the mesh by a conventional photoresistive technique. The adhesion strength of the printed ink on the substrate was carried out by tape test analysis based on modified ASTM standard D3359-09. The surface homogeneity and quality of the printed film were characterized using optical microscopy (MRDX, Leica Microsystems, Wetzlar, Germany). The surface roughness of the printed pattern was analyzed using atomic force microscopy (AFM) (Multimode, Bruker, Germany). The analysis was carried out on different areas of 10 μm × 10 μm and 4 μm × 4 μm, respectively, operating in tapping mode. Microstructural analysis of the printed film was done by scanning electron microscopy (SEM) using a JSM 5600LV instrument (JEOL, Tokyo, Japan). The dielectric properties in the low-frequency region (300 Hz to 3 MHz) were studied using an LCR meter (Hioki model 3532-50, Nagano, Japan). The microwave dielectric properties of the printed film were measured using a split post dielectric resonator (SPDR) (QWED, Warsaw, Poland) technique at different frequencies (5, 10, and 15 GHz) with the aid of an ENA series ES071C vector network analyzer (Agilent Technologies, Santa Clara, CA, USA). The I–V characteristics of the printed film were measured using a source meter (Keithley 2410, 1100 V). For this measurement, the prepared ink was printed on a thin copper film, and gold metal was sputtered on the top of it as the front electrode.
RESULTS AND DISCUSSIONS

In order to elucidate an exact idea of the calcination temperature and the different phase transitions, TG-DTA of the prepared wet gel was carried out. Figure 1a shows the TG-DTA curve of the gel up to 900 °C in air atmosphere.

The TGA graph clearly indicates four distinct stages of weight losses. In the first stage, the weight loss starts from 40 °C and shows a sharp decrease up to 110 °C; the total weight loss is around 6%. This may be due to the removal of planar water and ethyl alcohol. In the DTA curve, a corresponding sharp exothermic peak is observed around 54 °C. In the second stage, a slow and steady weight loss of almost 3% occurs up to 240 °C. This is believed to be due to the dehydration of Bi(OH)3 to BiOOH, and a representative endotherm is clearly visible around 236 °C in the DTA curve. A sharp weight loss was observed after 240 °C to around 312 °C, which indicates the decomposition of Si(OH)4. The TG weight loss continued to 530 °C, and thereafter no considerable weight loss was observed. The loss in this region implies the decomposition of the residual bismuth nitrate present in the system. From the entire TGA process, we could observe a total of 24% weight loss from room temperature to 530 °C. The typical exotherms at 286.6, 466.8, 514.9, and 637.1 °C for the decomposition of Si(OH)4, Bi2O3 phase transition, decomposition of Bi(NO3)3, and the formation of Bi5SiO13 metastable phase, respectively, are also evident in the DTA curve. The sharp endotherm at 702 °C corresponds to the crystallization of Bi5SiO13, and a small endotherm around 860 °C indicates the formation of Bi4(SiO4)3.29,30 Based on the TG-DTA results, the prepared nBSO powder was calcined at 850 °C/4 h.

The FTIR spectroscopic technique can be used as a powerful tool for structural studies of colloidal compounds, to identify their constituent chemical species. Figure 1b depicts the room-temperature FTIR spectrum of the calcined nBSO powder. The two sharp bands at ~482 and ~540 cm⁻¹ in the spectra indicate the characteristic bending mode of the Bi–O bond in the BiO6 octahedral unit. The strong band centered at ~887 cm⁻¹ is assigned to the stretching vibration of a Bi–O bond in the BiO6 octahedral unit. A weak diffuse band centered at ~1100 cm⁻¹ is characteristic of a Si–O–Si unsymmetrical stretching mode in a SiO4 tetrahedral unit.31–33 These results indicate that the basic structural unit in the nBSO comprises [BiO6]6—octahedral and [SiO4]4—tetrahedral groups, which is in concurrence with earlier observations.34

The XRD pattern of the calcined nBSO powder is shown in Figure 2a. All the peaks in the XRD pattern were indexed using standard JCPDS file 35-1007. The result indicates that the formation of this phase of nBSO was complete at 850 °C. The crystal structure belongs to the I4̅3d space group, having cubic symmetry. The lattice strain and stress are believed to have a great influence on the physical properties of nanomaterial. XRD profile analysis is a well-executed technique for estimating the micro strain. From the available models, the Williamson–Hall (W-H) method is the most simplified and extensively studied technique35 for analyzing the crystallite size and lattice strain. The instrumental broadening effect was rectified by using silicon as the standard. W-H analysis is an integrated method, in which the crystallite size- and strain-induced line broadening of the XRD profile was calculated from the peak width using the following equation:35

\[
\beta \cos \theta = \frac{K \lambda}{D} + 4 \varepsilon \sin \theta
\]

where D is the crystallite size, K is the shape factor (~1), \( \beta \) is the peak width (fwhm), \( \lambda \) is the wavelength of Cu Kα radiation used, and \( \varepsilon \) is the strain in the crystal. From the plot of \( \beta \cos \theta /
against \(4 \sin \theta/\lambda\), we are able to calculate the strain as well as the size of a crystallite. In principle, the slope of the graph gives the strain, whereas the reciprocal of the intercept should be the crystallite size.\(^{36}\) Figure 2b represents the W-H plot of nBSO calcined powder. A negative strain of around \(-2.64498 \times 10^{-4}\) is obvious from the curve. This implies that the crystal undergoes compressive strain in its lattice that may be mainly due to the lattice shrinkage.\(^{37,38}\) The crystallite size calculated from the intercept is \(~67\) nm. The average crystallite size was also calculated from the Debye–Scherrer formula \((D = 0.9\lambda/\beta \cos \theta)\) by considering the five highest intensity peaks, having \((hkl)\) of \((211), (310), (321), (422),\) and \((431)\), respectively, and obtained an average value of 69 nm.

The particle size, as we discussed, has a crucial role in the ink formulation, along with the rheology and microstructure of the printed film. For example, smaller particle size can improve the viscosity as well as the colloidal stability of the ink. However, in screen-printing, it is not smaller particles but uniform particle size distribution that is more advantageous to yield a homogeneous microstructure of the printed film. In order to homogenize the particle size of the nBSO powder and de-agglomerate the particles, the as-prepared powder was ball-milled for 4 h. The actual nanocrystalline nature of the nBSO particles could be tested using TEM analysis. Figure 3a represents the morphology and particle size of the calcined nBSO powder after ball-milling for 4 h. It is clear from the figures that nBSO has an average particle size of \(~70–80\) nm. In Figure 3b the lattice fringes of the crystal are clearly visible, which attests to a highly crystalline nature of the nanoparticles. The \(d\)-spacing value of the two adjacent fringes in this crystal lattice is measured to be around \(2.72\) Å. The result indicate that these fringes are of \((321)\) plane, having a lattice constant of \(10.18\) Å. The fast Fourier transform (FFT) pattern (Figure 3c) specifies the reflections from two crystalline planes. The \(d\)-spacings of these two reflections were measured, and results showed that these reflections originated respectively from the \([310]\) and \([321]\) planes of the crystal.

Ideally, the functional inks require great control over their rheological properties, in order to facilitate smooth printing and efficient functioning of the end devices. This could be achieved only by the judicious selection of different additives and precise control of their physical and chemical properties. The quantity and quality of organic additives such as solvent, dispersant, and binder must be chosen so as to control the rheology of the ink within the printable range.

The rheological analysis of the formulated ink is given Figure 4. From Figure 4a, one can observe that the viscosity of the ink decreases with an increase in shear rate. Such a shear thinning behavior is essential for screen-printing ink. This is because when the shear rate increases, the ink become thinner, causing it to flow freely. One of the crucial steps in the screen-printing process is pushing of the ink through the mesh opening with a squeegee and subsequently registering the pattern over the carrier substrate with good leveling.\(^{39}\) The shear stress curve also shows the typical nature of pseudoplasticity, which helps to prevent “bleeding”, i.e., the tendency of ink to flow beyond the printed boundaries after printing. In fact, it is the organic binders and dispersants that play a crucial role in converting the viscosity of the ceramic filler to follow a pseudoplastic behavior. Pseudoplasticity is a result of aggregation of the suspended particles.
particles by attractive van der Waals forces as well as the repulsive steric effect on the particles. The combination of these forces prevents the particles from joining together, rather than separating the particles with weak physical bonding. When a shear force is applied, the particle-to-particle network is broken, and as a consequence, the viscosity is decreased.\(^4^0\) In addition, the storage modulus (\(G'\)) and the loss modulus (\(G''\)) also influence screen-printing: the former indicates the elastic (solid-like) component, while the latter symbolizes the contribution of the viscous (liquid-like) component in the fluid.\(^3^9\) For effective printing, it is likely that the ink has more viscous-like character than elastic nature. The dynamic viscoelastic properties of the nBSO ink are given in Figure 4b. A better understanding of the nature of \(G'\) and \(G''\) will be useful in understanding the discrepancy between the influences of organic and inorganic segments on the viscosity of the screen-printing ink. From Figure 4b, it is obvious that the value of \(G''\) is higher than that of \(G'\) in the entire range of strain. This reveals that the developed ink has more liquid-like behavior, which is essential to prevent slumping and also to guarantee better homogeneity and a high aspect ratio in printing.\(^3^9\) The variation of stress against strain of the formulated ink is shown in Figure 4c, which is another characteristic of pseudoplastic fluid and gives more insight into the role of the polymer binder in controlling the rheology of the formulated ink.

Figure 5a shows the NIIST-CSIR emblem screen-printed on flexible Mylar film, and Figure 5b is the optical micrograph of the printed film. Obviously, in the screen-printing process, the ink is deposited on the substrate as tiny rectangles, and once the screen is removed, the ink flows out to form a smooth surface. Leveling is strongly controlled by a number of factors, including flow time, surface tension, viscosity, and thixotropic nature of the ink, mesh size, quality of the squeegee and screen, etc.\(^1^3\) In the present study, double-stroke printing was adopted in order to mitigate the island formation. It is evident from Figure 5b that the printing was more or less uniform, without much porosity. However, some mesh patterns were also visible in the printed film which may be due to the rheological variations at high curing rate, leading to leveling imperfections. In fact, the fast curing nature of the developed ink will tend to increase its viscosity very rapidly, soon after the printing. This, in turn, will restrict the ink materials to get a more homogenized leveling before curing. The scanning electron micrograph given in Figure 5c is also recorded from the same printed film, which obviously looks smoother only because the scanning area is smaller. A uniform distribution of the nBSO particles with minimum porosity was observed in the microstructure. A minor amount of porosity is unavoidable, since it arises from the volatilization of the solvents used in the ink formulation. The cross-sectional microstructure of the ink printed on Mylar film is shown in Figure 5d. From the figure, we can clearly distinguish the printed pattern from the Mylar substrate. The thickness of the printed pattern is measured from the micrograph, and it is almost 6.3 \(\mu\)m, more or less uniform throughout, signifying homogeneity of the printed pattern.

Surface smoothness is of prime concern in printed patterns, which depends to a greater extent on the particle size distribution of the filler, the porosity created by the volatilization of the organic vehicle, and also the leveling behavior of the ink during printing. A couple of typical AFM micrographs of the printed pattern, after double-stroke printing, is depicted in Figure 6. The average surface roughness (\(R_s\)) and the root-mean-square roughness (\(R_{rms}\)) of the printed film are around 80.3 and 102 nm, respectively. These values were lower compared to earlier reports on room temperature curable dielectric inks,\(^1^1,1^3\) indicating that the developed ink can be used to generate good-quality printed patterns. The 3D images of the printed film show the bumps-and-valleys nature of the printing. Kurtosis topography, which is the measure of unevenness of the printed film, gave a value of around 2.99. If the kurtosis value is less than 3, which is usually called “platykurtotic”, it can be taken as a testament that the surface has relatively few high mountains and low valleys.\(^1^3\) This bumps-and-valleys nature is mainly due to the presence of unavoidable porosity created by the fast evaporation of the solvent. However, this has no major significance in the quality of the printed film, since the roughness values are comparatively lower. The skewness (measure of asymmetry of the surface) value of the printed film is around \(-0.237\); the negative value indicates that the surface has a greater number of longer tails compared to the reference plane.\(^1^3\)

The printed ink is found to be cured within seconds after printing. Even though drying process of evaporation-drying-type inks can be accelerated by blowing hot air over the surface to be dried, no such cumbersome arrangements need to be made in the present case. The rub-off resistance of the printed film on the substrate was characterized by adhesion test using Scotch tape (3M, Maplewood, MN, USA). In order to measure
shows the photograph of the film and Scotch tape after testing. It is clear from the photographs that no portion of the printed ink is removed during this adhesion test, which indicates a strong adhesion of the formulated ink on the carrier substrate. To get a better idea, optical and SEM analyses of the film were carried out after the test. Figure 7b shows an optical image of the film taken exactly along the cut line. It is very evident from the image that no part of the ink is removed during the test, which is further confirmed by the SEM image shown in Figure 7c.

The dielectric properties of the printed film at microwave frequency were measured, and the results are tabulated in Table 1. The printed film has a dielectric constant ($\varepsilon_r$) of 4.46 and tan $\delta = 0.0131$ at 5 GHz. It is interesting to note that, with an increase in frequency, the dielectric constant shows a slight decrease, whereas the tan $\delta$ value shows a reverse trend, the difference being within the experimental error limit. The tan $\delta$ value of a material is highly influenced by external factors like humidity, porosity, impurities, etc. at this frequency range. For practical purposes, it is desirable that the dielectric layer have a stable and low tan $\delta$ value ($<0.05$). In the present case, the dielectric loss of the printed film for all the measured frequencies comes within this range, reiterating the suitability of the developed ink for printed flexible circuit applications. Additionally, it should be noted that the $\varepsilon_r$ of the nBSO-printed film was comparatively lower while tan $\delta$ was higher than those of bulk ceramic ($\varepsilon_r = 13.3$ and tan $\delta = 0.0007$ at 15 GHz). This divergence is believed to be due to the presence of the organic binder, combined with the inherent porosity of the prepared film. The low $\varepsilon_r$ value of the developed ink also indicates its aptness for application in interlayer dielectrics and as a protective coating for microelectronic devices. The fast curing nature is an added advantage for the developed ink compared to the currently available UV-curable inks, besides other advantages like durability, compatibility with all kinds of substrates, resistivity to abrasion, and high flexibility. The stability of dielectric properties with varying temperature as well as mechanical deformation is one of the major concerns in printed electronics. Toward this objective, we have performed reliability tests of the printed film with temperature and mechanical bending. The results are shown in Figure 8.

Table 1. Microwave Dielectric Properties of Mylar Substrate and Printed nBSO Film

<table>
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<tr>
<th>material</th>
<th>thickness (µm)</th>
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<td>tan $\delta$</td>
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<td>0.0131</td>
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</table>

Figure 8a represents the variation of dielectric constant and tan $\delta$ at 5 GHz with temperature from ambient up to 60 °C. As is obvious from the figure, $\varepsilon_r$ and tan $\delta$ show a negligible variation with temperature, that variation being linear. The results indicate that the dielectric properties of the film are quite stable at the operating temperatures. The variation of the dielectric properties with increasing bending cycle (up to 500 bending cycles) also emphasizes that the dielectric properties are independent of bending deformation.

A representative $I$–$V$ plot of the printed film is shown in Figure 9a. The result indicates that the current density shows an exponential increase with electric field that is typical for a dielectric material. Furthermore, one can notice that the printed pattern has a low leakage current density. In conjunction with the other results discussed above, we can infer that the formulated nBSO ink is a suitable candidate for creating a thin dielectric layer for electronics applications. The variation of dielectric and electrical properties of the developed ink in the low-frequency region is shown in Figure 9b,c. It is obvious from Figure 9b that $\varepsilon_r$ and tan $\delta$ decrease with increasing frequency. This is because at lower frequency, all the polarization mechanisms that are responsible for the dielectric constant of the materials are active, yielding high dielectric constants. With an increase in frequency, the effects of interfacial and dipolar polarization decayed, lowering the dielectric constant. The ac conductivity of the ink is presented...
The material has a very low conductivity, on the order of $10^{-14}$ S/cm. At lower frequency, the conductivity remains almost constant, and after a particular frequency, the conductivity starts to increase exponentially. This can be well explained using Jonscher’s power law:

$$\sigma(\omega) = \sigma_{dc} + A\omega^n$$

where $\omega$ is the frequency, $\sigma_{dc}$ is the frequency-independent conductivity (low-frequency region), $A$ is the temperature-dependent pre-exponential factor, and $n$ is the frequency exponent. As per the equation, at low frequency the conductivity is practically frequency-independent ($dc$ conductivity), and with increasing frequency the second term in the power law equation starts to dominate. On the other hand, at higher frequencies, the ac conductivity is directly proportional to $\omega^n$, which indicates that the conductivity increases exponentially.

## CONCLUSIONS

Low permittivity dielectric inks have potential applications in printed electronics due to their ability to isolate printed conductive patterns in multilayer circuits, besides possessing flexibility and mechanical robustness. With the objective of formulating a fast-curing dielectric ink, we prepared nBSO powder through sol–gel synthesis. The calcination temperature of the prepared powder was optimized using TG-DTA analysis. The phase formation of the nBSO powder was revealed using XRD and TEM analysis. Crystallite size of the nBSO was measured by W-H analysis and found to be around 67 nm. FTIR studies were also carried out to understand the structural units present in the prepared ceramic. The stable viscous ink of nBSO was formulated using a xylene–ethanol mixture as the solvent system, while Triton X-100 and ethyl cellulose were used as dispersant and binder, respectively. The microstructure, surface roughness, and dielectric properties of the developed ink were measured. The printed film has a dielectric constant of $\varepsilon_r = 4.41$ and tan $\delta = 0.0131$ at 5 GHz. The reliability of the printed film with temperature variation and mechanical bending was tested, and the results concluded that the film is stable during temperature and mechanical (bending) changes. The newly developed low-$\kappa$ ink can be used as insulation in flex circuits and thick films, for improving moisture, arc track, and weathering resistance of electrical/electronic devices.

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