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Acoustics and the Ultrasonic Measurements

Lead-free thick-film ultrasound sensors for industrial process monitoring

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Summary: Sol-gel high-temperature ultrasound transducers (HTUT) have been developed to compensate for the limitations present in many commercial ultrasound sensors, such as limited temperature durability and limited frequency ranges, etc. The most common HTUTs are lead-based and more environmentally friendly options are desired. For industrial process monitoring, ideally HTUTs can be fabricated directly onto machine tools, dies, vessels and moulds to create information-rich but minimally invasive sensors. In this study, two lead-free sol-gel composites are compared with a common lead-based HTUT material for the feasibility of spray-coating as a thick film on a steel substrate. The general procedure and key information for the preparation of the precursors for each composite have been outlined. The firing and annealing procedures necessary for the successful creation of films with good mechanical stability and surface adhesion are also outlined. The characterisation of the film properties demonstrates reasonable adhesion and mechanical stability for each film.

Keywords: HTUT, Sol-gel, Lead-free, Thick-film, Characterisation

1. Introduction

Unlike commercial temperature and pressure sensors, ultrasound transducer measurements can be used to infer a range of material properties in an industrial process non-invasively [1]. Also, HTUTs, when compared to conventional ultrasound probes, are miniature, can tolerate high-temperature, can be applied on curved shapes, and can thus be located in areas of industrial equipment where conventional probes are difficult to fit. Hence, they can be a better choice for the sensorisation of industrial processes and equipment under the framework of Industry 4.0.

Barrow *et al*. [2] invented a method for the fabrication of a crack-free thick-film (thicker than 10µm) to use as HTUTs by dispersion of ceramic powder in a sol-gel solution. In the current study two lead-free CBT/BST (calcium bismuth titanate/barium Strontium titanate) and BiT/ST (bismuth titanate/strontium titanate) thick-film sol-gel composites and commonly used lead-based BiT/PZT (bismuth titanate/lead zirconate titanate) as a benchmark for the development of HTUT were investigated. BiT and CBT are chosen due to their high-temperature durability (almost 700 °C and 800°C respectively), ST and BST are chosen due to their suitable dielectric constant.

The ability to apply each sol-gel solution to a steel substrate by spray coating is investigated. Spray coating was selected as the application method since it is possible to apply to different areas of industrial equipment unlike other coating methods such as spin and dip coating which are not easily portable. All composites were developed on steel substrates with dimensions of 30mm×40mm ×10mm. The quality of the films was characterised by Powder X-Ray Diffraction (XRD) and Scanning Electron Microscopy (SEM).

2. Methodology

The PZT solution was prepared based on the method outlined in [3]. This comprises a water-based solution and metal alkoxide precursors with inverted mixing order to increase the stability. The final solution has a similar consistency to thin paint and has a suitable viscosity for spraying.

The BST and ST solutions were prepared based on the fabrication procedures detailed in [4], [5]. Both solutions were very similar, and the only difference was that the BST formulation contains barium acetate. A successful preparation procedure results in a transparent and clear solution with suitable consistency for spraying.

After the successful preparation of the solutions, the respective ceramic powders were added to make the sol-gel suitable for thick-film applications. The powder was fine enough (~1µm) to make a crack-free film. The commercial BiT powder was purchased from Fisher Scientific and ball milled to reach an accuracy of almost 1µm making it suitable for fabricating a crack-free film. The CBT ceramic powder was manufactured in the lab by following the procedure in [6]. The CBT powder was also ball-milled for around 24hours to produce a fine powder.

For the substrate, surface treatment was necessary before coating to increase the adhesion strength between the film and the sample. For the sample preparation, first, the substrate was polished with sandpaper, then the sample was placed in a methanol ultrasound bath, after which the sample was annealed. Following the annealing process, the sanding step was repeated, and ultrasound cleaning was applied to remove the oxidation layer and clean the surface respectively.

After the sample and sol-gel materials were prepared, the coating method was conducted for each composite using an airbrush. Each layer was fired at 85℃ for 10mins for Bit/PZT, and 150℃ for 5mins for CBT/BST and BiT/ST. The thickness of the final three layers for each composite was just under 70µm.

After the initial heat-treatment, each sample was annealed in a muffle furnace. A similar annealing process was followed for each of the composites based on [7] with slight modifications. Three annealing stages included a slow ramp to 285℃, and a rapid increase to 400℃ to pyrolyse and oxidise the organic components. For the final stage, the annealing process continued at 650℃ with the heating rate of 200℃ per hour and held for 5mins to ensure crystallisation.

3. Experimental Results and Discussion

Fig. 1 presents images of the composites taken after annealing. Some minor cracks were observed at the surface of BiT/PZT due to a non-uniform film caused by difficulty in manual spray coating. These cracks can also be observed in the SEM result of the film in Fig. 2. These cracks may have occurred during the heat treatment because of the large difference in thermal expansion of the steel substrate and the film. These cracks were not observed in the two lead-free composites.

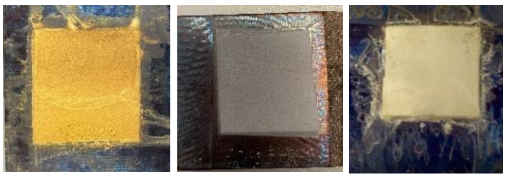

Fig. 1. Sol-gel composites after annealing a) Bit/PZT b) CBT/BST and c) BiT/ST

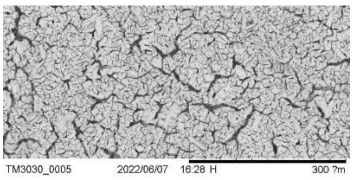

Fig. 2. SEM results of BiT/PZT

The annealed sol-gel samples were characterised through XRD. The XRD results of all three composites are shown in Fig.3. The figure presents that crystallisation occurred for solutions and ceramic powders of each composite and thereby the annealing process and heat treatment have been effective in creating a thick film with suitable material properties for use as a HTUT.

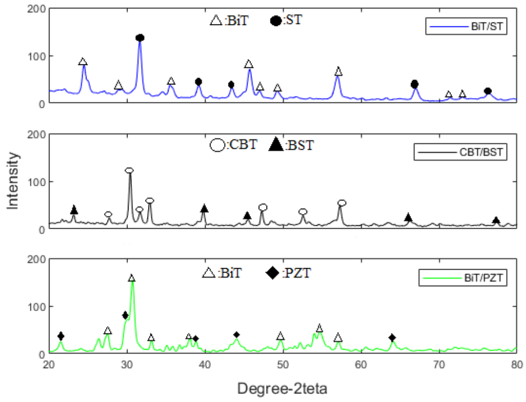

Fig. 3. XRD Results for BiT/ST

4. Conclusions

The initial steps for the preparation of two lead-free HTUTs and a common lead-based comparison HTUT were outlined. Future work will include poling each composite to enable characterisation and evaluation of their piezoelectric performance as HTUT. Once the complete HTUT fabrication process is finalised, the developed sensors will be used in an actual industrial process where the data will be used for real-time monitoring and optimisation of the process.

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