Research Article

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Piezoelectric Ceramic-Polymer Composites as Smart Materials: An Overview of Preparation Methods

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ABSTRACT

Discrete types of compounds were involved in preparing smart materials. This study exclusively implicates only piezoelectric compounds owing to their innate piezoelectric effect. The ferroelectric natured materials: ceramic and polymer has been employed to fabricate efficient piezoelectric composites. In this study, efforts were taken to elucidate the methods involved in preparation of heterogeneous composites applied in the field of constructing transducers, sensors, actuators, etc.,. Their methods of preparation and applications vary from one another depending upon their connectivity and each of the methods successfully described with felicitous figures.

Key words: Smart materials; Piezoelectric effect; Heterogeneous; Piezoelectric composites; Ceramics; Polymers.

1. INTRODUCTION

Several traditional and modern ceramic-ceramic, polymer-ceramic, metal-polymer and ceramic-metal combinations were used for material fabrications (Koray Akdogan and Mehdi Allahverdi 2005). The properties of composite combinations with different connectivity kinder their applications in many fields, remarkably 1-3 and 2-2 connectivity were applied in transducer, medical imaging and micro-fabrication applications. In this reviewDice and fill method, Injection molding and Lost-molding technique were some famous methods (Jain *et al.* 2015). The other methods like tape casting, micro-fabrication by co-extrusion, solid freeform fabrication, hot press method, solvent casting, spin coating and cold isostatic pressing were also available andelucidatedbelow.

2. EXPERIMENTAL METHODS

2.1 Dice and Fill Method

The various preparations such as 1-3 and 2-2 connectivity, this method suits in best way. In this method first a ceramic plate is diced by orthogonal slicing for two times as illustrated in fig1(a). The slicing process gives some small rods separated by little distance of width 50µm and thickness about 35µm (Klicker et al. 1981; Klicker *et al.* 1982; Mirza *et al.* 2016). After slicing, epoxy or any other polymer was filled in the place where the rods were embedded. The third process was grinding the ceramic layer present below the rods. Finally the ceramic plate was ready with electroding and polling.

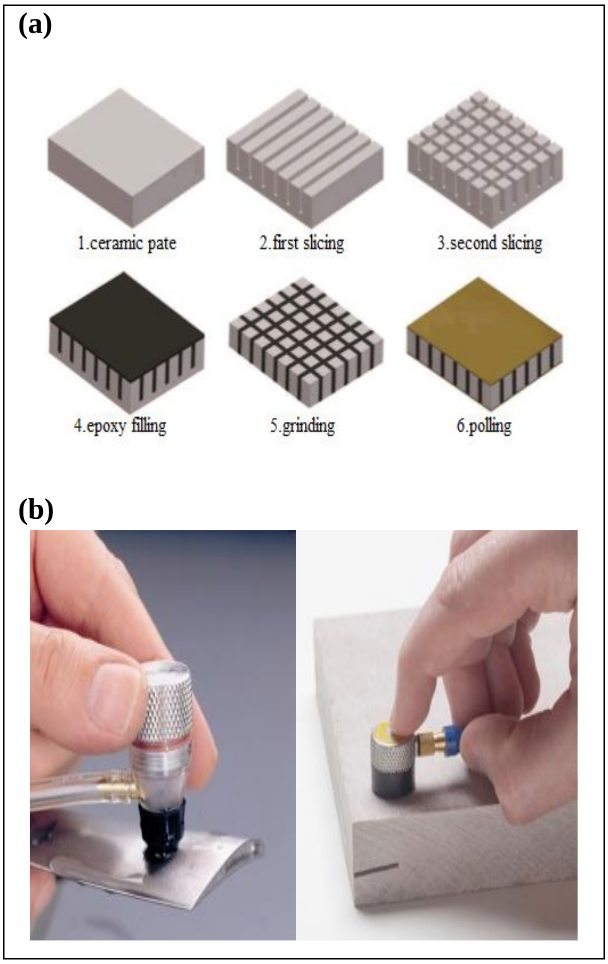


Fig.1: (a) Represents schematic diagram of dice and fill method, (b) Ultrasonic and medical transducer applications.

2.2 Lost Mold Method

Fabrication of composites with 1-3 connectivity was prepared by lost molding. A plastic mold for fillingceramic slurry, LIGA (Lithography Galvano-forming and plastic molding) preparation was employed (Becker *et al.* 1986). In this method, a wax mold has the negative of the desired structure which was made by two methods, namely punching holes in wax sheet or by bundling up of micro-tubular organic fibers. After drying, the mold which was prepared shall burn out and then sintered in a controlled atmosphere to get structure with >98% density; this process was called “binder burnout”. Hexagonal rods ~50µm in diameter, 400µm height and spacing of 50µm apart have been made brilliantly with the help of the instrument shown in figure 2 (a).

2.3 Injection Molding

Injection molding couldbe used to manufacture composite easily. Industries use Injection molding because it gives rapid throughput, minimum amount of material waste, flexibility and low cost per part(Bowen and French 1992; Bowen *et al.* 1993). Different varieties of rod with different sizes, shapes and spacing could be made by injection molding. By the standard injection molding apparatus figured at 3 (a), preformed 2-2 sintered sheet composites with 25µm and 1-3 PZT rods with 30-40µm diameters were produced Lubitz *et al.* 1992). Mold could not be formed each time for different shapes due to its limitation over time and cost, hence lost molding technique was involved to prepare mold.Basically, this method was cost effective approach.

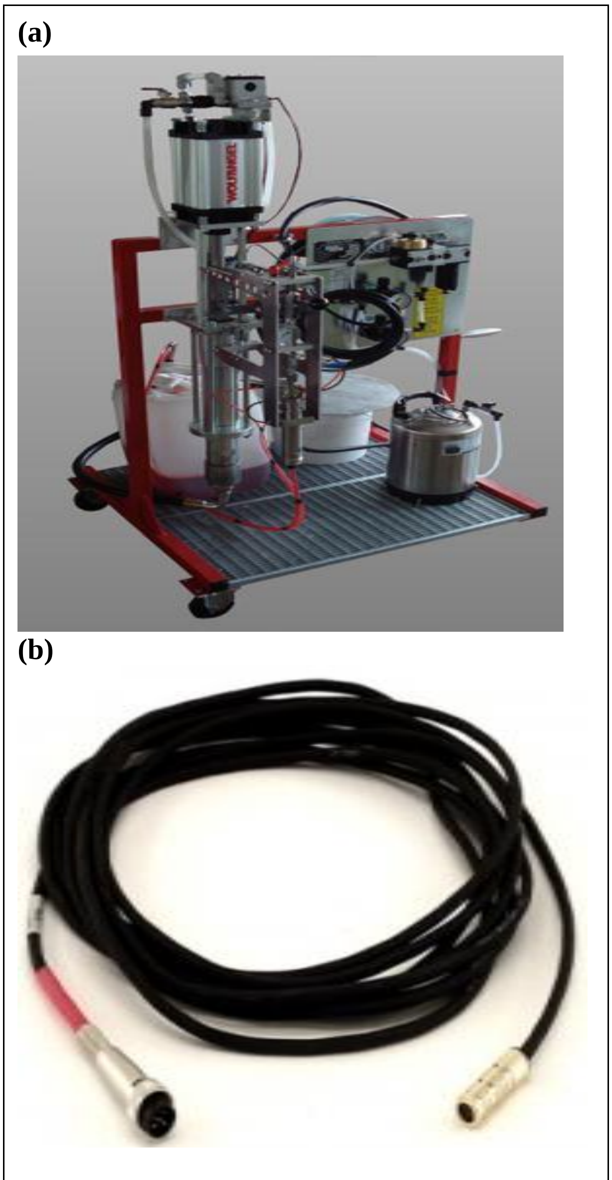


Fig. 2: (a) Lost mold apparatus, (b) Hydrophone.

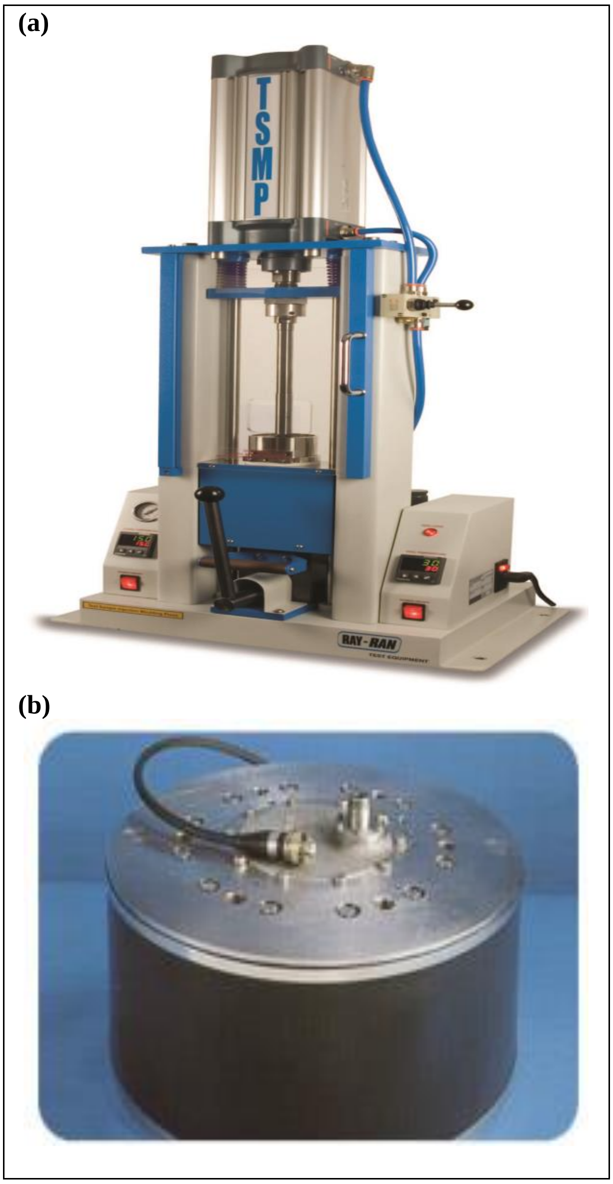


Fig. 3: (a) Injection molding apparatus, (b) Underwater transducer.

2.4 Tape Casting Method

This method was involved to fabricate 2-2 connectivity composites into very thin tapes i.e., down to several micronsas in figure 4 (b) and the tape casting apparatus displayed in figure 4 (a) (Huebner *et al.* 1847; Hackenberger *et al.* 2000). It allows the manufacturer to produce a transducer with high frequency for high resolution medical imaging and recently high frequency ultrasound transducer greater than 20MHz were fabricated using cast staked tapes of PZT and polymers. Composites with 2-0-2 connectivity and1-3 connectivity were employed in tape casting.

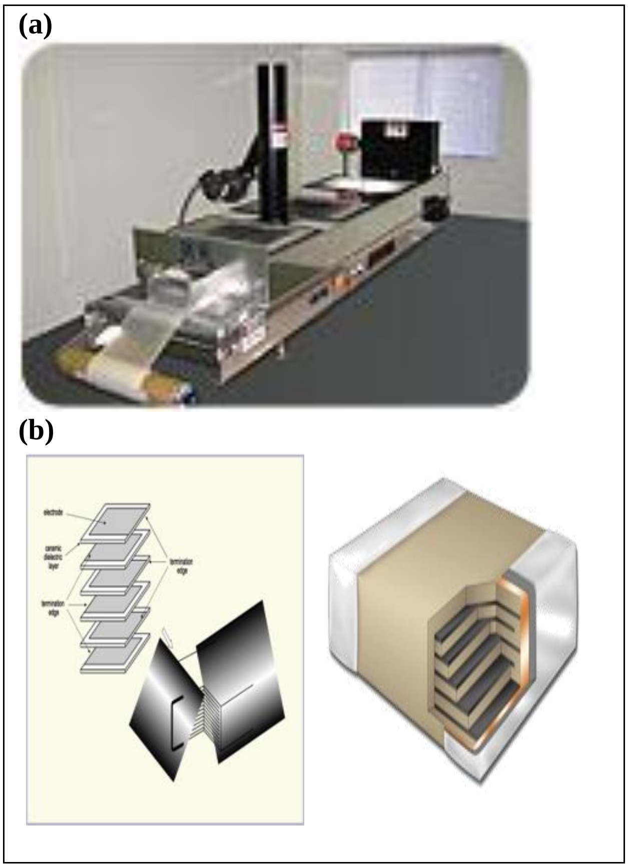


Fig. 4: (a) Tape casting apparatus, (b) Multilayered capacitor.

2.5 Microfabrication by Co-Extrusion

This fabrication process could be done by the method as shown in below figure 5 (a), illustrates twoproduction and extrusion process and figure 5 (b) showing an actuator with and without their inner core. The extrusion feedrod assembly was used to transform macro-scaledfeedrod into micro-scaled parts. Van Hoy et al. and Halloran et al. (Halloran 1999; Hoy *et al.* 2005)were the first to develop microfabrication by Co-Extrusion (MFCX) method that includes forcing of thermoplastic ceramic extrusion compound through an array of the die with a given reduction ratio.There were several stages of extrusion that involve in the production of arrays of objects in the size range of 10µm.

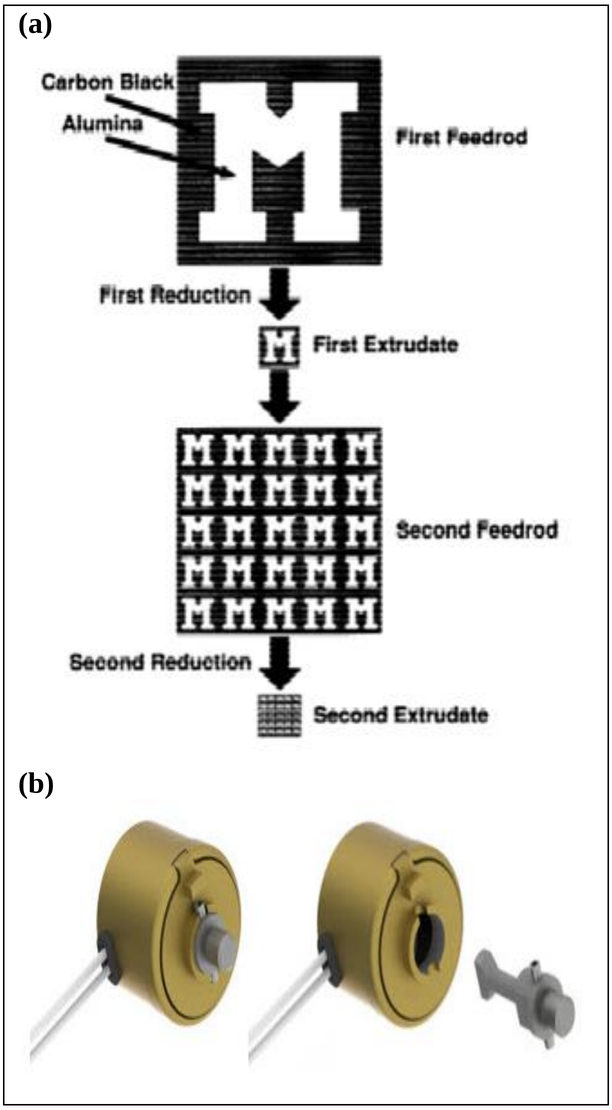


Fig. 5: (a) Schematic representation of microfabrication by Co-Extrusion method, (b) Actuator.

2.6 Solid Freeform Fabrication (SFF)

Most of the methods described before were used for composite with simple connectivity, but 'Solid freeform fabrication' provides complex internal hierarchy and symmetry in the place of composite fabrication without using molds. In this method from a CAD file, three-dimensional components were formed on a fixtureless platform directly by fabricating polymer, metal or ceramic structures. A surface file was created from a CAD file, whichmust be given as an input to the manufacturing system. From the surface file, cross-sectional slices or a slice file was obtained as such each slice could be constructed uniquely by varying the tool path. Finally slices could be transmitted in a layer-by-layer fashion to the SFF machine to obtain the layered approach. Moreover, this method consumes less time and cost which becomes a good advantage.Solid freeform apparatus along with its biomaterial application over bone regeneration(Yeon Kwon *et al.* 2015)exposed in below figure 6.

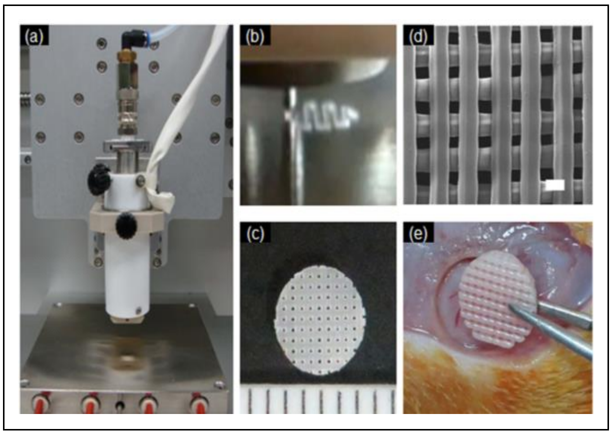


Fig..6: (a) & (b) Solid Freeform Fabrication apparatus, (c) Optical imaging (d) SEM imaging, (e) Implimentation of fabricated scaffold.

Fig. 7: (a) Hot press apparatus, (b) Pellets made by hotpressing.

2.7 Hot Press Method

One of the methods applied for the production of bulk samples and its process was carried out by heating a polymer in a solvent like n-dimethylormamide (DM), dimethylformamide (DMF), imethylacetamide (DAM) or dimethylsulfoxide (DMSO) (Seema *et al*. 2007). The solution was stirred continuously using a magnetic stirrer under particular temperature until it gets required viscosity. Ceramic powder was added as filler to the polymer-solvent mixture, which could be later made into a pellet by the use of stainless steel mold present in the hot press apparatus (Gowdhaman *et al*. 2016). The combination of heat and pressure was applied to make pellets and also its thickness could be varied. Finally a polling apparatus filled with silicon oil was used to pole the pellet. Many connectivity patterns like 1-3, 0-3, 2-2, etc., were easily constructed by hot press apparatus presented in figure 7 (a).

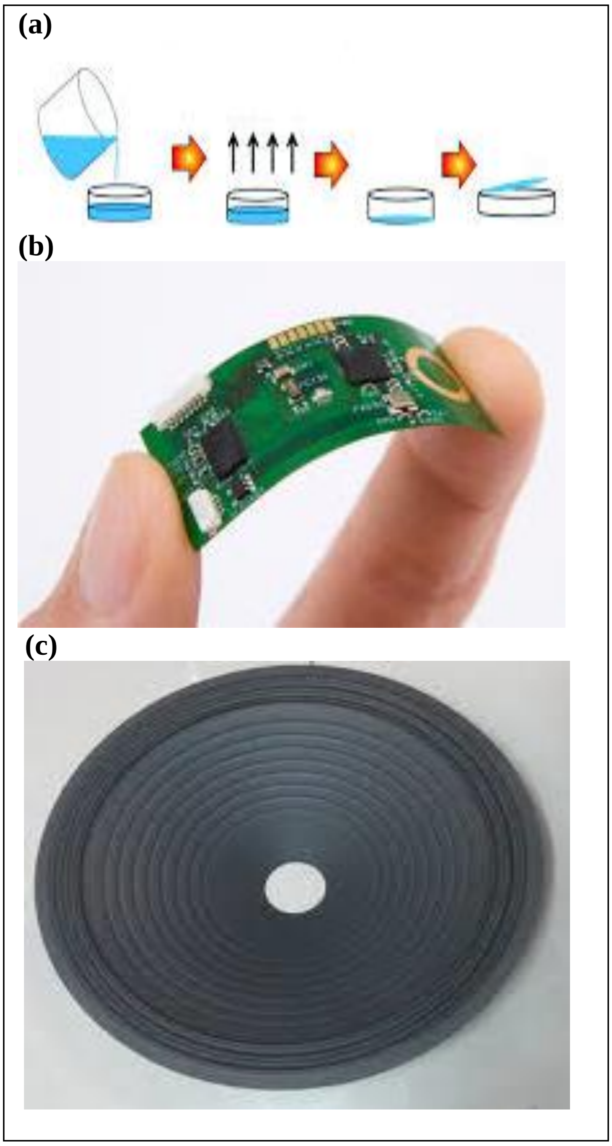
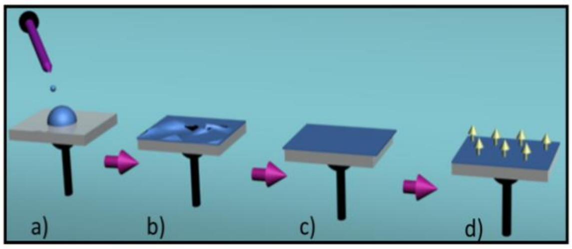


Fig. 8: (a) Schematic representation of solvent casting method, (b) Flexible printed circuit board, (c) Loudspeaker membrane.

2.8 Solvent Casting

This method was one of the oldest in composite preparation.Solvent casting gives thin and uniformly casted tapes. Similar to that of hot press, solvent casting also applies simultaneous heat and magnetic stirring to the filler and polymer mixtures. Thus obtained solution was poured over a glass mold and kept for a particular time at a furnace for solvent evaporation producing crystalline polar phase composites (Siemann 2005; Jain *et al.* 2014). The entire process could be seen in below schematic diagram 8 (a) representing the steps involved in solvent casting process. Composites with 3-1 connectivity were also obtained using PZT rods and PVDF solution over it.



(e)

(f)

Fig. 9: (a), (b), (c) & (d) Schematic description of the spincoating method, e) Spin coater machine, (f) Disk coating.

2.9 Spin Coating

This method involves simultaneous application of heat and magnetic stirringof the ceramic-polymer mixture. Solvent in the mixture gets evaporated during the spin coating process. Thus prepared material could be placed on the surface of the material to be coated and the spinner gets spun. Uniform coating was achieved with the help of centrifugal force (Tyona 2013). Thickness of film could be varied depending upon theviscosity, concentration of both solvent and solution and also speed of the spinner. Figure 9 (a) to (d) represents the procedures involved in spin coating, (e) spin coater instrument and (f) coated disk.

(a)

(b)

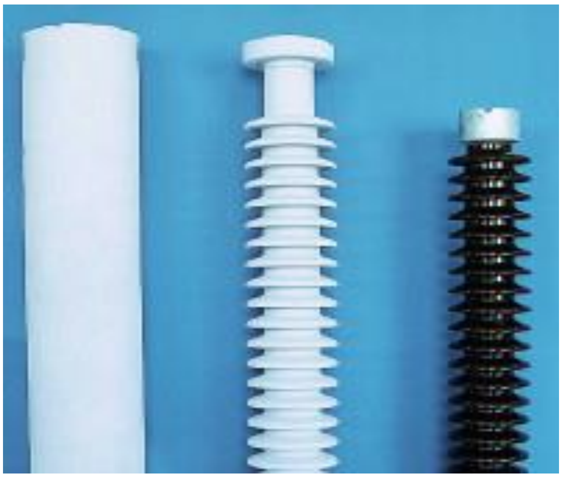


Fig.10: (a) Cold isostatic press apparatus, (b) Isolators.

2.10 Cold Isostatic Pressing

Isostatic pressing was applied to the chosen powdered materials like metal, ceramic, composite or plastic which were placed inside the mold made out of rubber or urethane or by PVC. Using water the powder composite inside the mold was pressurized in the range of 400 to 1000MPa. Thus obtained green compact could be sintered if it was theoretically denser(Henderson *et al.* 2000). There were two methods in preparing, one was dry-bag method and another was wet-bag method. Either one could be used for the preparation of mold depending on their application of complex structures like a turbine wheel. Figure 10 (a) represents the cold isostatic apparatus and (b) stand for isolatorwhich was made by cold pressing method.

3.RESULTS AND DISCUSSION

The composites having 1-3 and 2-2 connectivity were utilized in most of the transducer application. The methods (1), (2), (3), (4) and (7) were involved in fabricating 1-3 and 2-2 connectivity. Dice and fill method deceits ultrasonic transducers and medical transducers for NDT applicationsshown in figure 1 (b) (Taghaddos *et al.* 2015). Lost molding differs in invention by including LIGA process producing transducers, sensors, actuators and hydrophones as pictured in figure 2 (b) (Becker *et al.* 1986).Using this method, rods with different shapes, structures and spacing could be made. The limitation was about the inability to rapidlyprototype samples and time consuming LIGA process. Underwater transducersused in deep seas depicted in figure 3 (b) were processed by injection molding (Lubitz *et al.* 1992). The above discussed methods fabricate single layered material, but tape casting method involves casting of multiple layers of composites like multilayered capacitor portrayed in figure 4 (b) (Tok *et al.* 1999). In the hot press method, 0-3 connectivity was used for manufacturing the contended application (Seema *et al.* 2007) and also its design over pellets couldbe seen in figure 7 (b).

In microfabrication, minute micro-scaled architectures and advanced actuatorspresented in figure 5 (b) were made (Halloran 1999; Hoy *et al.* 2005). Solid Freeform fabrication mainly involved in the bone regeneration process (Yeon Kwon *et al.* 2015). Flexible printed board and loud speaker membrane were exhibited in figure 8 (b) and (c). Comparatively, thesewere few applications over solvent casting method (Jain *et al.* 2014). Spin coating method could not describe any connectivity factor for its composites. Photolithography, dielectrics, magnetic disk coating, compact disk, television tube phosphor and antireflection coating were done by this technique (Chang *et al.* 2005). Hardest composites/plastic, madedevices, namelyisolators, turbine wheels could be obtained using cold isostatic press due to its high pressure range.

4. CONCLUSION

Contrasting methods with different components were discussed for the preparation of piezoelectric ceramic-polymer composites and their employment has been brought in this review. Among the different methods discussed in this study three methods (Solid freeform fabrication, Spin coating, Cold isostatic pressing) have not debated about the connectivity, whereas other techniques have discussed on different connectivity in choosing their composites.The piezoelectric composites prepared in these methods have been widely used by the researchers around the globe for transducers, actuators, sensors, hydrophones and many other applications due to its fascinating properties.

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CONFLICTS OF INTEREST

The authors declare that there is no conflict of interest.

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