Fabrication of Curved Ceramic / Polymer Composite Transducers for Ultrasonic Imaging Applications by Fused Deposition of Ceramics

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Abstract

Fused Deposition of Ceramics (FDC), developed at Rutgers University, is a Solid Freeform Fabrication (SFF) technique where a three-dimensional green ceramic object is built layer by layer, starting from a Computer Aided Design (CAD) file of the object. This technique was used to build novel piezoelectric ultrasonic transducers for medical imaging applications. Curved ceramic skeletons for 2-2 (parallel ceramic / epoxy plates) composite transducers were built by FDC. The design's curvature can be tailored in the CAD file. Therefore, the final composite requires very little machining. In the FDC-built green parts, the ceramic plates were 500 μm thick and the spacing between the plates was 1270 μm. The FDC green samples were subjected to a slow binder burnout cycle at 550°C for 4 hours, using a heating rate of 8°C per hour, then sintered at 1285°C for 1 hour. Physical characterization of the samples revealed that 95% of the theoretical density was achieved. The ceramic plates shrunk 20% in height as well as in width. The shrinkage was of only 16% in the direction parallel to the plates. Optical microscopy and SEM were performed on green and sintered samples. The results of these characterizations are reported in this paper as well as the electromechanical properties of the final composites and of FDC bulk samples.

Introduction

Fused Deposition of Ceramics [1] is a Solid Freeform Fabrication technique in which a green ceramic object can be built layer by layer, due to the deposition of roads by a liquefier moving in the x and y directions. The feedstock for this technique is a ceramic loaded, 1.78 mm diameter filament, which acts as a piston to push the material out of the nozzle. This technique was used to fabricate the ceramic portion of a piezoelectric ceramic / inactive polymer composite transducer for medical imaging applications. Composites are employed because they combine the property advantages of both polymer and ceramic transducers and thus have properties superior to the ceramic phase alone [2]. The material of choice was lead zirconate titanate (PZT) for the ceramic and Spurr epoxy for the polymer [3]. Ultrasound medical imaging transducers are used both as emitters and as receivers. For optimum efficiency in these two modes, the volume fraction of ceramic in the composite should be maintained between 20 and 30 volume percent [4]. This range is determined by compromising to achieve high piezoelectric charge coefficient (d_{33}), moderate acoustic impedance and high thickness coupling coefficient (k_t) [5]. For medical imaging applications, a curvature in the transducer's surface will allow for focusing the beam at a defined distance away from the transducer. This focusing ability leads to a better lateral resolution, as the beam is smaller in the focal zone and therefore has a higher intensity; this increases the signal to noise ratio [6]. The design flexibility afforded by the FDC process allows the designer to freely choose the radius of curvature and all other transducer specifications without penalty of cost or manufacturing time associated with increased complexity of the design.

Composite Design

Composites can have different "connectivities" as described by Newnham et al. [7], meaning that each phase can be self interconnected in either one, two or three dimensions. For this project, a 2-2 connectivity was chosen, where both the ceramic and the polymer are continuous in two directions (x and z), thus forming alternating parallel plates (Figure 1).

The minimum green ceramic plate width (w) is dictated by the nozzle diameter used and the firing shrinkage. The volume fraction of ceramic then gives the spacing (s) between these walls. In order to avoid lateral resonant waves between the plates (Lamb waves) [8], the thickness of the transducer should be larger than the pitch, which is defined as the sum of the width of one ceramic and one polymer plate. The resonant frequency of the transducer is inversely proportional to the transducer's thickness [6].

In order to improve the resolution, it was chosen to use a curved transducer . The radius of curvature should be chosen depending on the application, i.e. the distance of the biological organ to image from the transducer. Two radii of curvature were chosen: 10 and 20 cm. Flat transducers (infinite radius of curvature) were also built for comparison.

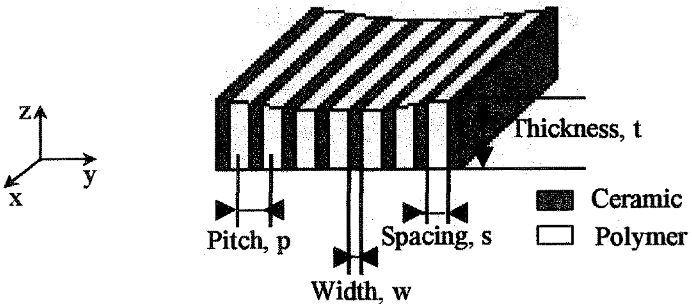


Figure 1: Schematic Representation of a Curved 2-2 Ceramic Composite Built by FDC Technique.

Fused Deposition of Ceramics

The first step in making any structure by the FDC technique is to fabricate the raw material feedstock filaments. The generic processing sequence for PZT particle loaded filament fabrication was as follows: coating the PZT powder (non spray-dried, PZT-5H, TRS Ceramics, Inc., College Park, PA) with 1 wt. % stearic acid as a dispersant to lower the systems overall viscosity, compounding of coated powder (to 60 volume fraction of PZT ceramic) with binder, granulating the compounded material into mm sized pellets, and finally extrusion of filaments with close control over their diameter. Based on previous results, stearic acid was chosen as the dispersant. The binder used is a four component binder [9]. Several processing parameters were optimized to improve the quality of the filament. Capillary rheometer viscosity measurements indicate a sufficiently low viscosity (at the actual FDC temperature of 166°C) for proper FDC behavior. These filaments were successfully used in the FDC process.

The next step was to create a CAD file of the desired transducer. In this project, the ceramic structure had to be curved on the top. However, since SFF techniques are layerwise techniques, the curvature had to be approximated by discrete steps. Figure 2 shows an example of the stepped surface.

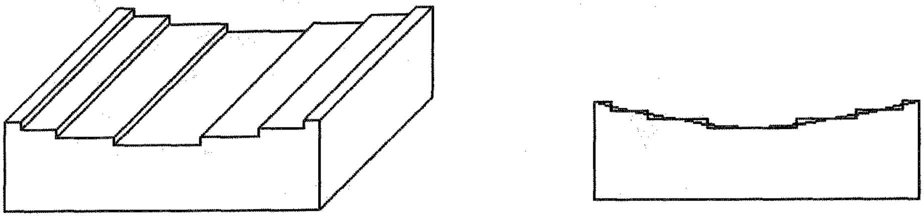
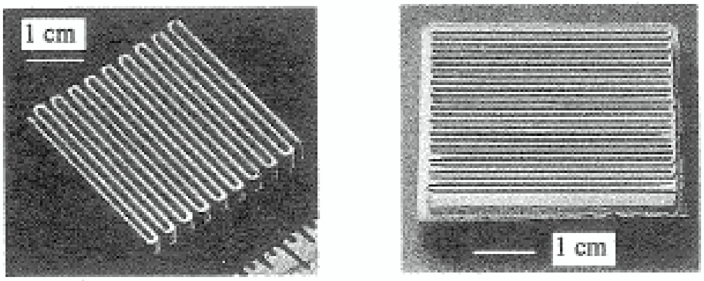

(a) (b)

Figure 2: (a) Computer Aided Design of the 2-2 Composite Structure; (b) Side View of the CAD File Showing the Curvature Approximation by Steps.

Quickslice software was then utilized to slice the CAD file and to create the tool path. In order to create the parallel PZT plates for the 2-2 composite structure, a gap was left between the deposited ceramic roads. For the first parts, the nozzle diameter, road width and spacing between the roads were selected to be 508 µm, 508 µm and 1270 µm respectively. The slice thickness was 254 µm. Later, thanks to the development of a new binder formulation, it was possible to fabricate parts using a 254 µm diameter nozzle; in these parts, the road width, spacing and slice thickness were respectively set at 330 µm, 762 µm and 127 µm. In each case, the FDC build parameters such as liquefier temperature, envelope temperature, flow rate, etc., were optimized to improve part quality. The liquefier and envelope temperature were respectively set at 165 and 50°C. The flow rate was optimized in order to avoid overflow as well as underflow of material at the point where the nozzle starts depositing.

Figure 3 shows as-built ceramic structures for 2-2 composites fabricated by FDC with 508 µm (a) and 254 µm (b) diameter nozzles. It should be noted that the height of each of the parallel PZT plates is different, which results in the needed curvature for the focusing function of this transducer design.



(a) (b)

Figure 3: Images of Curved Green Ceramic Structures Built by the Direct FDC Technique; (a) Nozzle Diameter = 508 µm, (b) Nozzle Diameter = 254 µm.

Post FDC Processing

The green PZT structure made by the direct FDC process was subjected to binder burnout (BBO) and sintering heat treatments after FDC. Since the part had fine features, the BBO cycle was modified to maintain part integrity: it was done by heat treating the part in static air atmosphere to 550°C for 4 hours with a heating rate of 10°C per hour. The set up was as follows: the sample was put in an alumina crucible on top of a coarse PZT bed; this bed eased the removal of the binder from the bottom of the sample. Sintering was performed at 1285°C for one hour in an excess PbO atmosphere (to prevent Pb loss during sintering) in a closed and sealed alumina crucible; the heating rate was 3.5°C per minute. The flow of oxygen inside the furnace was not controlled. This was followed by Corona poling of the ceramic using a voltage of 26 kV for 15 minutes. After aging for 24 hours, the ceramic structure was impregnated in Spurr epoxy polymer. The composites were then electroded, repoled and the electromechanical properties were evaluated.

Part Characterization

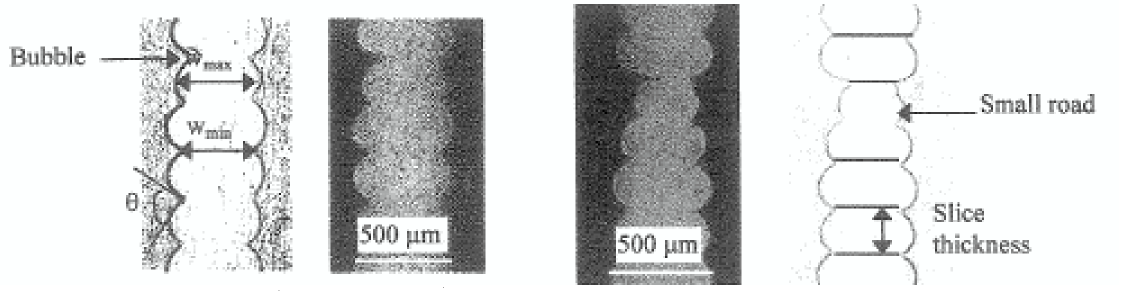


Figure 4: Optical Micrographs (and computer enhanced images) of Cross-Sections of Several Sintered FDC Plates made by FDC SFM Process.

Optical and scanning electron microscopy was performed on green and sintered PZT samples in order to characterize each part's micro- and macro-structure. Defects identified in the first iteration of the process (non-uniform road widths and porosity/bubbles) (Figure 4) were rectified through process improvements as can be seen from the micrograph shown in Figure 5.

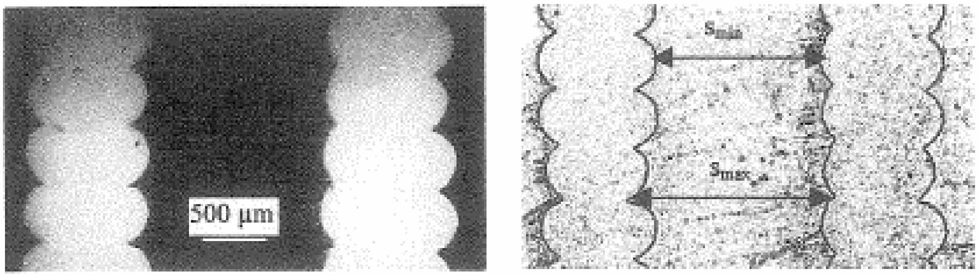


Figure 5: Optical micrographs (and computer enhanced image) of cross-sections of green PZT plates made by the FDC SFM process, showing that the defects such as small roads and bubbles were eliminated by improved processing.

Table 1 reports the measured values for road widths, slice thickness, pitch and the surface angles. The road width in the green state is what the FDC system builds and in the sintered state is after shrinkage during BBO and sintering. The surface angle refers to the angle that the material makes at the intersections of two successive layers. The main concern at this point is to decrease both the standard deviations of the values of road thickness, road width and inter-plate thickness (in both the as-built and as-sintered conditions), in order to maximize control over the component and insure that it accurately reproduces what is specified in the original CAD file. It is noted that, as with other ceramic materials made by SFF techniques, the green to sintered shrinkage measurements show anisotropy. For these parts, the shrinkage in the direction perpendicular to the roads (20%) was larger than in the direction parallel to the roads (16%). However, this can be incorporated into the original CAD design and can also be used to an advantage to obtain finer features. Scanning electron microscopy was performed on a fracture surface (Figure 6). These parts showed good layer to layer bonding and no delamination between the roads after sintering.

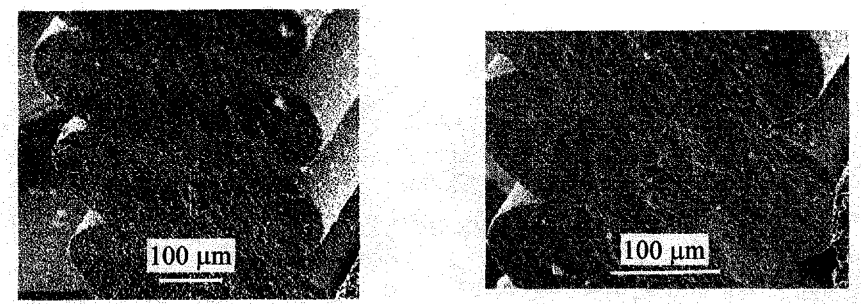


Figure 6: Scanning Electron Micrographs of fracture surfaces of sintered PZT plates showing good bonding between roads and high sintered density, i.e. no pores.

	Green part		Sintered part
	Measured values	**CAD values**	**Measured values**
Slice thickness	264 ± 7 µm	254 µm	212 ± 12 µm
Left angle	81 ± 15°		80 ± 13°
Right angle	71 ± 12°		81 ± 14°
Maximum width	553 ± 24 µm	508 µm	452 ± 39 µm
Minimum width	391 ± 34 µm		320 ± 44 µm
$(w_{max}-w_{min})/2$	82 ±12 µm		68 ± 24 µm
Maximum spacing	916 ± 15 µm		
Minimum spacing	751 ± 18 µm	813 µm	
Pitch= $w_{max}+s_{min}$	1307 ± 21 µm	1321 µm	

Table 1: Statistical results regarding dimensional tolerances of green and sintered PZT plates made by FDC. The sintered parts were made before process improvements, the green parts were made after (See Figures 4 and 5).

Piezoelectric properties were measured on the flat PZT / polymer composites and on bulk samples made by FDC. The results are shown in Table 2. The values obtained are comparable with those obtained by conventional methods.

Sample	V_f PZT (%)	k_t (%)	k_p (%)	K	tan δ	d_{33} (pC/N)	ρ (g/cc)	Z (MRayls)
Flat FDC composite	27	68	32	627	0.023	397 ± 16	2.88	8.34
Dice-and-fill composite	25	63	---	780	0.085	355 ± 14	2.80	9.79
Bulk sample	100	54	71	3340	0.023	664 ± 4	7.7	30.54

Table 2: Piezoelectric Properties of Composites and Bulk Samples Made by FDC.

Summary and conclusions

In summary, curved PZT composites with 2-2 connectivity have been designed and fabricated by the direct FDC technique. The curved 2-2 composite structure was successfully built by FDC using 508 micron and 254 micron diameter nozzles. Optical microscopy was performed on early samples, showing irregularities in the road widths for the green and sintered samples. Subsequent powder processing improvements have led to significantly better control over defects and road widths, etc. No significant irregularities were observed on recent samples as a result of these processing improvements. The slice thickness showed good uniformity. Scanning electron microscopy showed no delamination and good bonding of the roads. The shrinkage behavior of the FDC samples was shown to be anisotropic, as is the case for other ceramics made by the FDC process. The piezoelectric properties of the flat composites as well as those of bulk samples made by FDC were found to be similar to those obtained in samples made by conventional methods.

Acknowlegments

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