Robocasting of Photonic Band Gap Structures

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Introduction

Photonic band gap (PBG) materials[1] exhibit characteristic frequency bands in which the density of states for electromagnetic wave propagation approaches zero. This behavior has great potential for efficient bending of light, increased efficiency of lasers by inhibiting unwanted modes, and a variety of other applications where control of photons is important. The length scale, symmetry, and dielectric constant contrast of the crystal structure define the domain and directionality of this band gap. Solution of the Maxwell equations for periodic dielectric structures reveals that the PBG scales with the feature size of the crystal. Thus, while the ultimate application of PBG’s may target a 1.5 \( \mu \text{m} \) wavelength, fabrication routes that can create periodicity on the millimeter length scale can be used to rapidly screen new structures and materials with direct correlation to higher frequency applications. Our motivation was to demonstrate that a solid freeform fabrication technique, known as robocasting[2, 3], could be used to rapidly produce PBG structures on a mm length scale.

Robocasting, was invented at Sandia National Labs to utilize colloidal ceramic slurries in the rapid, moldless fabrication of components. The process starts by formulating a well-controlled, weakly flocculated colloidal suspension at high solids loading. The suspension is then extruded through a deposition nozzle onto a mobile x-y translation stage. A computer controls the x-y stage, allowing the extrudate to be deposited in 2-dimensional patterns derived from slices of a 3-dimensional CAD model. After each layer is drawn, the deposition nozzle is incremented in height and the next layer is deposited until the 3-D part is finished.

Robocasting of alumina slurry was used to construct self-supporting, spanning lattice structures that display a PBG. The symmetry of the rods forming the lattice resembled a face centered tetragonal stacking pattern. In addition to model lattices, intentional defects were introduced by periodically omitting rods from the lattice with the aim of producing allowed transmission modes in the PBG. Example transmission spectra are given for three lattices to highlight the frequency dependent attenuation in the 80 to 100 GHz regime.

Experimental Procedure

Materials System

The ceramic chosen for this study was AKP-30 aluminum oxide (Sumitomo Chemical Co., Osaka, Japan). AKP-30 is a high purity \( \alpha \) alumina (>99.99%) with a particle size of \(~0.3 \mu \text{m} \) and BET surface area of \( 7 \text{ m}^2/g \). \( \alpha \) alumina has a refractive index of \( n = 1.765 \). A poly(electrolyte), the ammonium salt of poly(acrylic acid) (Darvan 821A, R.T. Vanderbilt Company Inc., Norwalk, CT), was used as a dispersing agent. Darvan 821A provides electrosteric stabilization of the colloidal alumina particles by adsorbing on their surface at a
concentration of ~0.2 to 0.4 mg/m$^2$. In the robocasting suspensions, the concentration was 2 mg of active polymer per gram of alumina. Darvan 821A behaves similarly to poly(acrylic acid), as described by Cesarano et. al., [4, 5] in acidic and alkaline conditions. Potentiometric titration of Darvan 821A solutions of known concentration revealed that approximately 75% of the monomer groups were ionized at a pH of >8.0. The colloidal stability of the suspensions was tailored through pH and ionic strength adjustments[6]. Hydroxypropyl methylcellulose (Methocel F4M, DOW Chemical Co., Midland, MI) was added as a binder/thickener.

Fabrication of Lattices
The first step in the robocasting process is to formulate a suspension with sufficient yield stress to form the spans of the lattice structure. An aqueous suspension of 55% alumina by volume was achieved by initially dispersing the ceramic powder with the aid of the Darvan 821A and ultrasonication. After dispersion, the methylcellulose was mixed into the suspension from a stock solution. Next, the ionic strength and pH were adjusted to promote weak flocculation. The details of suspension rheology and shape evolution during robocasting are the subject of a future publication[7], however, the shear yield stress of these suspensions has been measured in the range of 50 to 150 Pa.

Next, the suspension was loaded into the robocaster and deposited at a speed of 8 mm/s through a 0.84 mm orifice. The robocaster was programmed to build lattices with the symmetry illustrated in Figure 1 where, $d$ is the pitch, $a$ is the rod diameter, and $c$ is the height of the four layer repeat unit. Similar lattices with this symmetry on the micron length scale have shown PBG’s at much shorter wavelengths[8].

![Figure 1](image)

**Figure 1** Schematic illustration of robocasting models where lattice 1 has no missing rods and lattice 2 has a pair of rods removed.

The filling fraction of dielectric rods is thus defined as $a/d$. Lattice 1 contained all the rods while pairs of rods were periodically removed from lattice 2. After deposition, the dried lattices were sintered at 1650°C for 2h.

Measurement of Photonic Band Gap
The transmission spectra of the lattices was probed through a frequency range of 80 to 110 GHz using a HP8510C network analyzer and mm wavelength test set (Agilent Technologies, Palo Alto, CA). The lattices were mounted on a sapphire substrate using small dots of glue at the corners and placed on a stage between the emitter and receiver of the test set. The microwaves were transmitted through the lattices in the stacking direction (see Fig. 1). The attenuation of each lattice as a function of frequency was determined by comparing their transmission spectra to that obtained for a sapphire window alone. The attenuation in decibels (dB) was therefore
calculated as $20 \log \left( \frac{I}{I_0} \right)$ where, $I$ is the transmitted microwave intensity with the sample present and $I_0$ is the reference intensity.

**Results and Discussion**

Two lattices were successfully robocast and sintered without (Figure 2(a)) and with (Figure 2(b)) periodic line defects.

![Lattice 1 and Lattice 2](image)

**Figure 2** Aluminum oxide lattices with 8 layers (2 unit cells) having parameters (a) $d = 2.25$ mm, $a = 0.64$ mm, filling fraction $\sim 0.28$ and (b) $d = 2.3$ mm, $a = 0.68$ mm, filling fraction $\sim 0.29$.

In each lattice, the filling fraction was $\sim 28$ to $29\%$ based on optical microscopy observation of the rod diameters and pitch. Lattice 1 was fabricated without defects and lattice 2 contained two line defects consisting of pair of missing rods.

The attenuation spectra for lattices 1 and 2 are plotted in Figure 3 for microwaves transmitted in the stacking direction.

![Attenuation Spectra](image)

**Figure 3** Attenuation spectra of (a) lattice 1 and (b) lattice 2. The smooth line through the data is simply an aid to guide the eye.
An attenuation of ~ -10 dB was observed around a center frequency of $f_o = 100$ GHz in lattice 1. In Figure 3(a) the center frequency is denoted by a vertical line and the +/- 6% markers give the bandwidth of the PBG (i.e., ~12% of the center frequency). Lattice 2 displayed a similar center frequency around 100 GHz. Again, the +/- 6% markers denote the band edges for comparison to lattice 1. The feature in this spectrum occurring at ~103 GHz is assumed to be associated with transmission modes allowed by the periodic defect states. Conclusive proof of this hypothesis, however, is impossible from these initial measurements and the limited data set.

Summary

We have demonstrated the ability to construct periodic lattices of aluminum oxide rods using robocasting. The lattices have good definition of the rods and spacing in both the x-y plane and the stacking direction. The filling fractions achieved were on the order of 0.28 to 0.29. Initial PBG transmission measurements indicate an attenuation of -10 dB for the defect free lattice and -15 dB for lattice 2 containing periodic line defects. The bandwidth for both lattices was approximately 12% of the center frequency. Lattice 2 appeared to display transmission modes commensurate with the inclusion of the periodic line defects. Further characterization of PBG lattices needs to be performed to verify the effects of engineered defect states on their transmission behavior. Robocasting provides a facile method to produce periodic dielectrics in the mm length scale. Current efforts are focusing on refining the scale of such lattices to reach into the low THz regime.