

# Gel-casting of ceramic components for wireless communications

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## Abstract

Gel-casting is a novel ceramic forming process for fabricating products of complex-shaped ceramics. In this contribution, the gel-casting process is applied to manufacture green body of ceramic components for microwave resonators, oscillators and antennas. Calcined powders, dispersant and organic monomers are mixed together by ball-milling to acquire high solid loading slurry with low viscosity. After addition of initiator and catalyst, the slurry is cast into a non-pours mold and then polymerized. The slurry is thus solidified in situ and the green bodies with the desired shapes are obtained. The dried body is rather strong and can be machined so as to get more precise or complex-shaped components. Then the green bodies are sintered at high temperature. The microwave properties like dielectric constant ( $\epsilon_r$ ), quality factor ( $Q$ ), and temperature coefficient of resonant frequency ( $\tau_f$ ) and microstructures of the ceramics prepared by gel-casting are compared with those of die-pressed ones.

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*Keyword:* Gel-casting

## 1. Introduction

Gel-casting has been developed as a forming process for fabricating green bodies of ceramics since the 1990s. The process is based on the in situ polymerization of organic monomers as the setting mechanism for forming the green body.<sup>1,2</sup>

The advantages of gel-casting can be summarized as follows: (1) capability of producing complex parts of ceramics; (2) possibility to use low-cost mould material, such as glass, metals and wax; (3) high strength of green body; (4) flexible machinability of green bodies that allows to process much finer detail parts; (5) excellent homogeneity of ceramic properties; (6) low organic content for easy binder removal compared with tape-casting process.

Microwave ceramics are important materials for microwave components, which are used in wireless communication technology.<sup>3</sup> Because of the complex shapes of microwave components, it is hard to fabricate them through traditional forming process, such as die-pressing, tape-casting, injection moulding. In this contribution, the gel-casting pro-

cess is applied to manufacture green body of ceramics, BaO–Nd<sub>2</sub>O<sub>3</sub>–TiO<sub>2</sub> (BNT), BaO–La<sub>2</sub>O<sub>3</sub>–TiO<sub>2</sub> (BLT) and MgTiO<sub>3</sub>–CaTiO<sub>3</sub> (MCT), for microwave components such as resonators, oscillators and antennas. Performances and properties of slurries, green bodies and sintered ceramics are discussed in detail.

## 2. Experiment

The essential reagents of gel-casting are commercially available, including monomers: acrylamide C<sub>2</sub>H<sub>3</sub>CONH<sub>2</sub> (AM) and *N,N'*-methylenebis-acrylamide (CH<sub>3</sub>CONH<sub>2</sub>)<sub>2</sub>CH<sub>2</sub> (MBAM), initiator: ammonium bisulphate (NH<sub>4</sub>)<sub>2</sub>S<sub>2</sub>O<sub>8</sub> (APS), catalyst: *N,N,N',N'*-tetramethylenediamine (CH<sub>3</sub>)<sub>2</sub>N–(CH<sub>2</sub>)<sub>2</sub>–N(CH<sub>3</sub>)<sub>2</sub> (TEMED), dispersant: ammonium polymethacrylate (PMAA–NH<sub>4</sub>) (25% aqueous solution with an average molecular weight of 15000). Calcined powders, in which raw materials BaCO<sub>3</sub>, Nd<sub>2</sub>O<sub>3</sub>, TiO<sub>2</sub> and Bi<sub>2</sub>O<sub>3</sub> are ball-milled, dried and then calcined at 1100 for 2 h, are grounded, milled and dried for later use.<sup>4</sup>

The flowchart of gel-casting is shown in Fig. 1. Firstly, AM and MBAM are dissolved in distilled water as a premix

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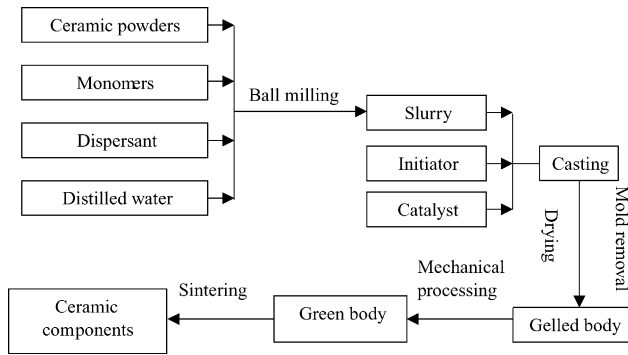


Fig. 1. Flowchart of gel-casting process.

solution. Ceramic powders and dispersant are then added. The slurry is ball-milled for 2–3 h to promote dispersibility and homogeneity. PMAA-NH<sub>4</sub> is selected as dispersant in order to maintain good slurry fluidity while obtaining high solid loading.<sup>5</sup> The suspension is degassed for 10 min until no further release of bubbles was observed. Then the initiator and catalyst are added. Afterwards, the slurry is cast into a nonporous mold in a vacuum chamber. After gelation, the green body is demolded and dried at room temperature. The green body is so strong that it can be machined to get more complex-shaped parts. Finally, the green bodies are sintered at high temperature.

Die-pressed ceramic samples with the same starting materials are also prepared in order to compare the difference of ceramics by two kinds of forming processes. As regards of the die-pressing route, the ceramic powder was granulated with PVA solution as binder.

Viscosity of the slurry is measured by rotary viscometer (Shanghai Jinke Co., China). Zeta potential of the powders is measured by a Zetaplus (Brookhaven Instrument Co., USA). The slurry to be tested is 0.5 vol.% and the pH value is adjusted by the amount of HCl or NaOH. The bulk densi-

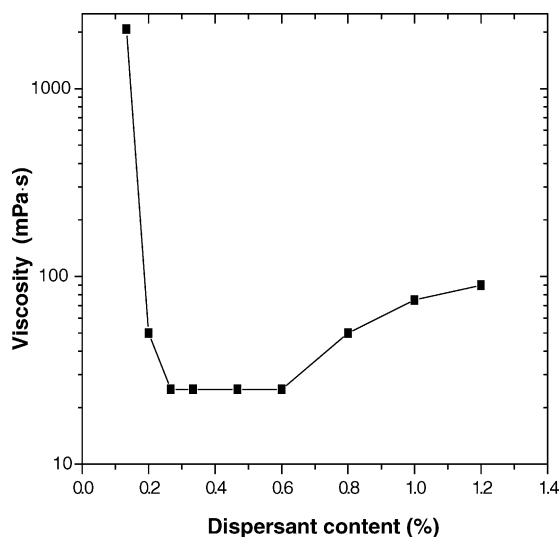


Fig. 2. Relationship between viscosity and dispersant content.

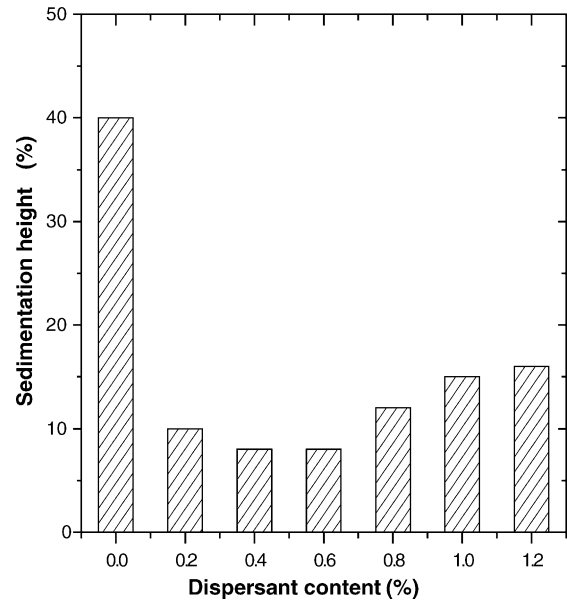


Fig. 3. Diagram of sedimentation and dispersant content.

ties of the samples are measured by the Archimedes method. The microstructures of samples are observed with a ESEM (Philips XL-30). The green bodies were tested in 3-point bending with a universal testing machine (Hongshan Instrument Co., China) at 30 mm span and displacement of 10 mm/min. Dielectric constant ( $\epsilon_r$ ), quality factor ( $Q$ ) and temperature coefficient of resonant frequency ( $\tau_f$ ) were measured by a vector network analyzer (Agilent 8714, USA).

### 3. Results and discussion

The relationship between viscosity and dispersant is shown in Fig. 2. It can be seen that the slurry is hard to flow without dispersant. With the addition of dispersant, the viscosity decreases dramatically and reaches minimum value

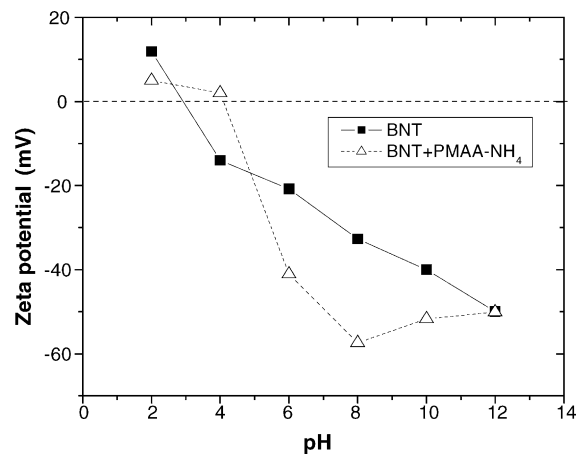


Fig. 4. Relationship between zeta potential and pH value with and without dispersant.

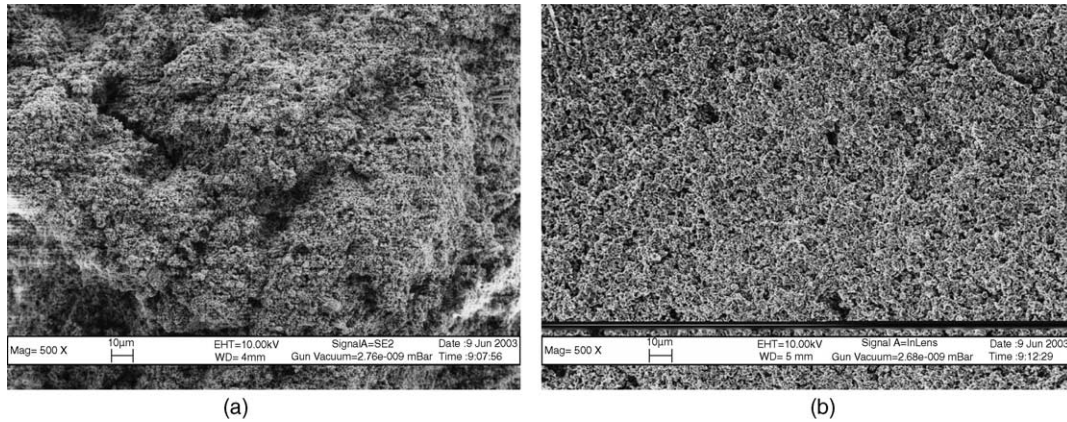


Fig. 5. SEM images of green bodies by two forming processes: (a) green body by die-pressing and (b) green body by gel-casting.

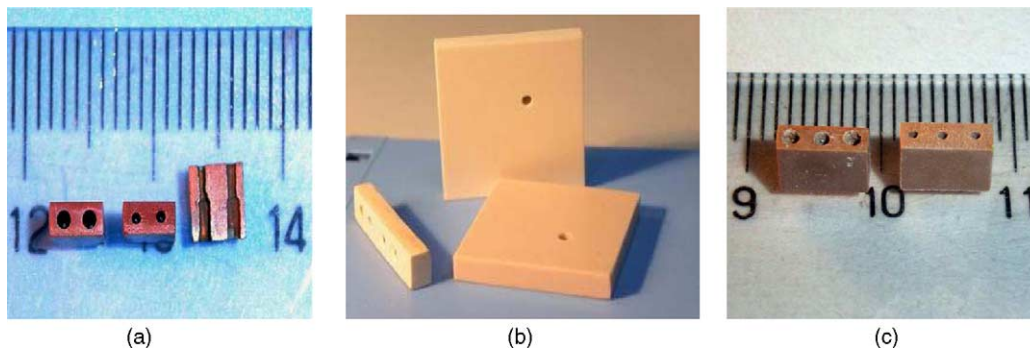


Fig. 6. Microwave components prepared by gel-casting: (a) BNT ceramics; (b) BLT ceramics and (c) MCT ceramics.

of 25 mPa s for the amount of dispersant from 0.3 to 0.6%. After that, the viscosity increases slightly if more dispersant is added. This may be due to the excessive addition of dispersant that leads to flocculation of the slurry.

The slurry is quite stable, which is tested by sedimentation experiments with 50 vol.% slurry placed for 24 h (Fig. 3). At a certain solids loading, a slurry with good dispersion results in lower sedimentation. The sedimentation height decreases with the increasing of dispersant content and reaches minimum value at the dispersant amount of 0.4–0.6 wt.%. Then sedimentation height increases slightly. This result agrees with the result of viscosity experiment. The optimized content of dispersant is therefore 0.4–0.6 wt.%.

The zeta potentials at various pH values are shown in Fig. 4. A higher zeta potential is expected to provide strong repulsion force between particles. At pH 8, the zeta potential reaches minimum value of  $-60$  mV.

Once dried, the gel-casting green bodies are sufficiently strong (7.9 MPa) for machining operations, such as surface polishing, hole drilling and band sawing without any observable damage. The density of green body prepared by gel-casting ( $2.89 \text{ g/cm}^3$ ) is slightly larger than the one by die-pressing ( $2.78 \text{ g/cm}^3$ ). The microstructure of the gel-casting green body is more homogeneous than the die-pressing ones, as shown in Fig. 5. This may be attributed to the good dispersion and in situ gelation of the suspension. These green

Table 1

Dielectric properties of ceramics prepared by two kinds of forming processes

Dielectric properties	$\epsilon_r$	$Q$ (GHz)	$\tau_f$ (ppm/ $^\circ\text{C}$ )
Forming process			
Die-pressing	92.6	1247	38
Gel-casting	95.3	1272	32

bodies are ideal for the consequent sintering of solid-state reaction.

Several kinds of simple and complicated parts for ceramic components have been formed by gel-casting in the experiments (Fig. 6). Table 1 shows dielectric properties of ceramics prepared by two kinds of forming processes. The dielectric properties of samples prepared by gel-casting are slightly better than those by die-pressing. This may be due to the gel-casting bodies that are more homogeneous than the die-pressing ones.

#### 4. Concluding remarks

Complex ceramic bodies for microwave components were successfully prepared via gel-casting route. The slurry reaches best dispersion state with the addition of 0.4–0.6 wt.% PMAA-NH<sub>4</sub>. The green body prepared by gel-casting with flexible machinability is more homogeneous

than that by die-pressing. The dielectric properties of samples prepared by gel-casting are slightly better than those by die-pressing.

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