

# A New Method for Preparation of Cellular Ceramics by Gelcasting Approach

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**Abstract**— This paper presents the results of a novel-processing route for fabricating highly cellular ceramics. The method is based on the generation of foam in two continuously stages: one, is the foaming of ceramic suspension by use foaming agent prior to the polymerization and other, is the evaporation of light organic solvent such as acetone, methanol and ethanol during polymerization because of exothermic reaction of gelation. The resulting ceramic foams consisted of a highly interconnected network of cells with densities as low as  $0.2 \text{ g/cm}^3$ . The distribution of cell size was dependent both on the density of the specimen produced and on the time for polymerization onset. The size ranged from approximately 50 to 1000  $\mu\text{m}$ . Enlargement of cell size to achieve materials of high permeability was possible through expansion of the foam via pressure reduction before polymerization.

**Keywords**— Cellular ceramics; Gelcasting; Idle time; Cell size distribution; Surfactant.

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## I. INTRODUCTION

Cellular ceramics have attracted increasing interest because of their application e.g. as separation for molten metal, hot gases, a variety of liquid filtration processes, as catalyst support, thermal insulators and sensors and in drug delivery system [1].

The interest in cellular ceramics has been associated mainly with their specific properties such as high surface area, high permeability, low mass, low specific heat and low thermal conductivity. In addition to the composition and mechanical properties of the cellular parts, the cell size and morphology are also important factors that influence the suitability for potential applications. Predominantly closed cell ceramics are needed for thermal insulation whilst open interconnected cell ceramics are required for uses involving fluid transport such as filters and catalysts [2].

Different methods have been employed for the production of cellular ceramics, namely, replication of polymers, use of hallow spheres, foaming of sol-gel based systems, GASAR processing and incorporation of fugitive phase. Smith adapted the original gelcasting system for the manufacture of cellular ceramics from foamed suspensions [3].

The development of ceramics by gelcasting has been allied with traditional concepts of the production of ceramic materials with polymer chemistry [4]. In this method, slurry of ceramic powder in a solution of organic monomers is

placed in a mould. The monomer mixture is polymerized *in situ* to form gelled parts.

The association of gelcasting with foaming followed by solidification by *in situ* polymerization forms an internal crosslinked network that transforms the foam in a strong-gelled part. The gel retains the foamed structure in such a way that both microstructures and macrostructures are preserved [5, 6].

This work describes studies on a novel-processing route for porous ceramics with a cellular structure. The technique comprises the foaming of ceramics in two continuously stages: one, is the foaming of ceramic suspension by use foaming agent prior to the polymerization and other, is the evaporation of organic solvent such as acetone, methanol and ethanol during polymerisation because of exothermic reaction of gelation.

## II. EXPERIMENTAL

### A. Materials

The specifications of the materials used in this work are similar to that of Ref. [7].

### B. Instruments

Instruments, which were used in this study, are as follows:

- Brookfield viscometer (Shanghai balance instrument factory, model NDJ-4, type LVF).

- Scanning electron microscopy (Philips XL30, magnifying to  $10^5$ ).
- Programmable furnace with computer controlling (Exciton, model Azar 1600).
- Universal testing machine (Measurements technology Inc, MTI 30K).
- Simultaneous thermal analysis (STA) (Netzsch, STA409-PC Luxx).
- Ultrasonic shaker (Branson model B200, output frequency: 40 kHz).
- pH meter (Eutech instruments, type WP2).

### C. Procedure

The preparation process by gelcasting is similar to that of Ref. [8]. Firstly, by using magnetic or mechanical stirrer (with turbine blade), pre-mix solution that contains monomers, solvent (water) and dispersant (ammonium salt of poly (methacrylic acid)) is prepared. After adding the dispersant to the pre-mix solution, alumina powders with various loading levels were added to the solution and stirred by using mechanical stirrer. Deagglomeration and homogenization were performed by ball milling in a plastic container for 10 h using alumina spherical grinding media with various sizes. In the first stage of foam making, use Triton X100 as foaming agent. It was added to the alumina suspension and vigorously stirring (up to 2000 rpm) was applied in order to generate foam. Setting of the suspension foam was promoted by the *in situ* polymerization of the acrylamide monomer. Before it, the novel second stage of foam preparation was promoted by adding solvent with low-boiling point to the alumina suspension. Since the polymerization is intrinsically exothermic reaction, the heat liberated by reaction could be easily vaporize the solvent and make foam with high surface area and cell size.

A procedure was employed to produce foams with cell size larger than those obtained directly through foaming. The suspensions were initially foamed as first stage and organic solvent was added. Within the time allowed by the induction period the foam was placed in a desiccator and had the pressure reduced using a vacuum pump until the required final volume was achieved. The pressure was held constant until setting of the foam took place.

After polymerization, the sample was cooled down to room temperature and removed from the beakers or moulds. The specimen were initially dried via liquid desiccant drying method [7] for approximately 2 hours and then dried at room temperature in air for approximately 24 hours. Once dried, polymer was burned out under controlled heating program, which was deduced from thermogravimetric analysis (STA) of acrylamide gels.

### III. RESULTS AND DISCUSSION

Figure 1 shows the foam volume versus stirring time for stabilized suspension containing 45 and 50 vol% alumina in two continuously stages of foaming. The foam volume increases initially by gas entraining into the suspension and liquid drawn around each bubble until a thin film is formed. Hence, the maximum foam volume in the first stage is associated with a minimum thickness of film that can sustain stable foam [9].

With respect to foam stability, as expected, bubble enlargement occurred at faster rates for less viscous and more highly foamed suspension, but viscosity had little influence on the final foam volume generated at high solid content slurry [9].

The experimental design and results of gelation process of the acrylamide-based system are shown in Table 1 and Fig. 2. The statistical technique popularly known as analysis of variance (ANOVA) was used to determine the significance of each variable and their interaction effects (using Qualitek-4 software).

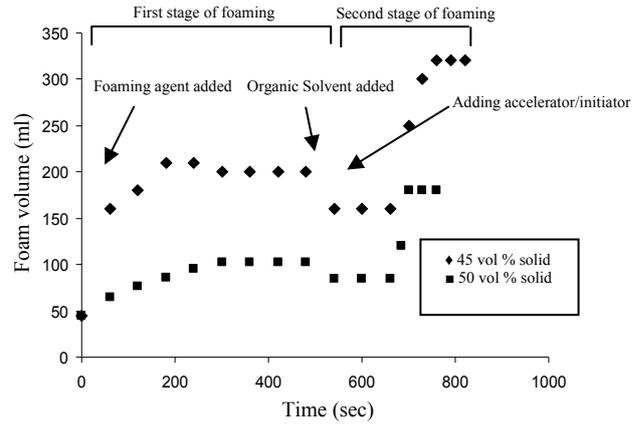


Fig. 1. Foam volume generated in two stages, using ethanol as organic solvent.

The signal-to-noise ratio (S/N) was used in the statistical analysis of the results. In fact, it is a way of isolating the sensitivity to noise factors [10]. The results are shown in Table 3 for idle time.

The statistical analysis of the results shows that, within the selected range of variables, concentration of the initiator has the strongest effect on the gelation behavior. The rest of variables (i.e. volume fraction of powder and molar ratio of monofunctional monomer to crosslinker) do not have strong effects on gelation behavior of gelcasting system. Also, the results shown in Table 2 determine that the counter interacting term of these variables is significant, i.e. 66%. This counter interaction term reduces the pure effects of these variables on the gelation behavior of gelcasting system.

TABLE I  
THE ORTHOGONAL ARRAY OF EXPERIMENTS AND RESULTS

No of exp.	Monofunctional monomer (vol %)	Molar ratio of monofunctional monomer to the crosslinker	Solid loading (vol %)	Molar ratio of initiator to total monomer	Results		
					1 <sup>st</sup> trial	2 <sup>nd</sup> trial	3 <sup>rd</sup> trial
1	12.5	16	35	0.50	320	310	315
2	12.5	18	40	0.75	150	153	145
3	12.5	20	45	1.00	80	78	82
4	12.5	22	50	1.25	40	43	40
5	15.0	16	40	1.00	40	40	42
6	15.0	18	35	1.25	20	19	21
7	15.0	20	50	0.50	230	226	234
8	15.0	22	45	0.75	120	118	123
9	17.5	16	45	1.25	40	38	40
10	17.5	18	50	1.00	65	63	68
11	17.5	20	35	0.75	75	70	72
12	17.5	22	40	0.50	135	130	138
13	20.0	16	50	0.75	85	87	82
14	20.0	18	45	0.50	110	110	112
15	20.0	20	40	1.25	15	14	15
16	20.0	22	35	1.00	40	42	40

As shown in Fig.1, evaporation of organic solvent during polymerisation generates high foam volume. The resulting ceramic foam consisted of a highly interconnected network of cells.

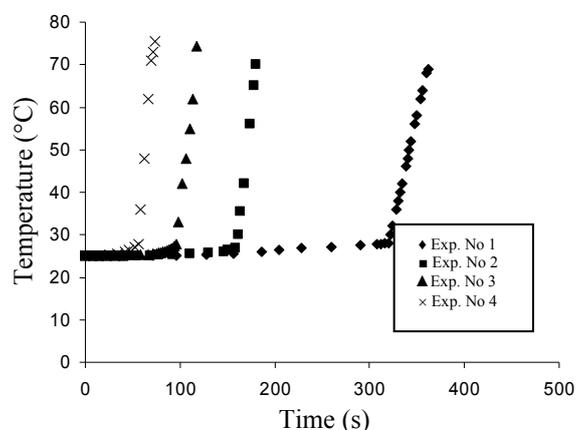


Fig. 2. The gelation behavior of the foamed acrylamide-based gelcasting system in the orthogonal array of experiments.

TABLE II  
PERCENT OF INTERACTION BETWEEN VARIABLES IN GELATION PROCESS.

Variables interaction	SI (%)
Crosslinker × Solid loading	66.14
Crosslinker × Initiator	26.3
Solid loading × initiator	25.7
Monomer × Solid loading	23.72
Monomer × Initiator	1.6
Monomer × Crosslinker	0.6

Crosslinker = Molar ratio of monofunctional monomer to the crosslinker

Initiator = Molar ratio of initiator to total monomer

Monomer = Monofunctional monomer

SI = Interaction severity index (100% for 90 degrees angle between the lines, 0% for parallel lines)

#### IV. CONCLUSIONS

A novel processing route for fabricating highly interconnected cellular ceramics based on gelation of acrylamide has been evaluated using commercial alumina powders with specific characteristics. The process was found to have the following features:

- The alumina slurry has low viscosity at high solids loading (50 vol % or >80 wt %) by the aid of

polyelectrolyte and TEMED as an accelerator in polymerisation reaction.

- Cell sizes was enlarged from 30-600  $\mu\text{m}$  in foams which is generated only by foaming agent, and expand up to 1000  $\mu\text{m}$  if also low boiling point solvent was used during gelation.
- Expansion of the fluid foams prior to gelation, generate a large range of cell size up to 2000  $\mu\text{m}$ .
- The distribution of cell size was dependent both on the density of the specimen produced and on time for polymerisation onset ( $t_{\text{idle}}$ ). In lower idle time, the mean cell size decreased and narrower cell size distribution was achieved.
- The statistical analysis of the results shows that, within the selected range of variables, concentration of the initiator has the strongest effect on the idle time.

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