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A Comparison Study On The Densification Behavior And Mechanical Properties Of Gelcast Vs Conventionally Formed B_4C Sintered Conventionally And By Microwaves

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ABSTRACT

This investigation compares microstructures and mechanical properties of both Gelcast B_4C and "conventionally" die-pressed B_4C, using both microwave and conventional sintering methods. The microstructures and final mechanical properties of B_4C specimens are discussed.

INTRODUCTION

The utilization of microwave energy for reaching high temperatures necessary to densify B_4C powder is compared with conventional means of sintering by evaluating the mechanical properties after densification. Microwave energy has been shown to be an effective means for achieving high sintered densities, even though temperatures of ~2250°C are required. In this study, green preforms of B_4C specimens were sintered by both conventional and microwave heating. This study also utilized an advanced forming method called "Gelcasting" developed at ORNL[1]. Gelcasting is a fluid forming process whereby high solids suspensions of powders containing dissolved monomers are cast into a mold, then polymerized or "gelled" in situ.

EXPERIMENTAL

Appropriate amounts of powders were weighed to yield a 50:50 blend of B_4C (#3000 and #1500 powders)† + 5wt% (Thermax) carbon powder as a sintering aid[2]. This B_4C mixture was first "washed" in methanol, which removes B_2O_3 as trimethyl borate [3]which significantly improves the powder dispersion. The methanol treatment of boron carbide powder consisted of the placement of powders in a plastic (HDPE) beaker, then filling remaining volume with methanol to fully cover all powder. This was followed by drying in an oven at 60-70° C to evaporate the methanol. This was repeated 3 times for efficacy. Samples of mixed powder were uniaxially cold pressed in stainless steel dies to 15KSI using a light coating of stearic acid as a die lubricant, followed by isostatic pressing at 50KSI.

Gelcasting slurries were prepared using a 15% aqueous solution of methacrylamide†† : methylbisacrylamide†† (MAM:MBAM at 2.5:1 respectively). The pH was adjusted to >11 using small additives of tetramethyl ammonium hydroxide (TMAH) ††. Darvan #7††† and PVP K-15†††† each at 0.5% by weight of the B_4C powder were added as dispersants.

† B_4C from E.S.K. Kempten, Germany (#3000 and #1500 powders)
†† Sigma Chemical Co., St. Louis, MO
††† R.T. Vanderbilt Company, Inc., Norwalk, CT.
†††† GAF Chemical Corp., Wayne, NJ: Polyvinylpyrrolidone K-15

The B_4C powder was then added to the monomer premix (to yield a solids loading of 45% by volume) while stirring on a laboratory mixer until fluid. The production of samples was completed as follows: The plastic (HDPE) mold was prepared by spraying with Polyester Parafilm. The B_4C slurry was degassed on ro-vap laboratory evaporator for ~ 30 minutes at approximately 0°C. (to prevent air entrapment in the finished samples) prior to addition of the catalyst tetramethylethylenediamine† (TEMED) first at the rate 0.1 microliter / gram (of slurry) followed by the ammonium persulfate† (APS). Note, the pH of the A.P.S. had to be adjusted as follows by: addition of 500 microlitres TMAH/ 25cc of an aqueous 10% APS solution. (which increased pH to ~8.5). This mixture was then stirred for 1-3 minutes on the Ro-Vap while continuing to degas for ~ 2 minutes. The APS was then added at the rate 4 microliters / gram of slurry, with an additional 2 minutes for stirring, before removal and casting. Note, the order of addition was found to be essential to achieve optimal dispersion.

The casting of samples was accomplished simply by pouring the slurry into molds to yield specimens (approximately .5" thick by 2.5"dia), which were then covered and placed in an oven at 70°C. Samples were allowed ~ 60 mins. for gelation. Specimens were removed from the molds and dried at room temperature. They were debindered in argon (to prevent oxidation and the formation of B_2O_3) to 600° C and were sintered by either conventional or microwave heating. Half of the green preforms were sintered conventionally at (2250°C) and half by microwave heating (estimated at 2250°C).

After sintering, the densities of samples were measured by the Archimedes method. The sintered samples were then diamond machined and polished. Hardness and toughness measurements were calculated from micro-indents made on the polished samples. Polished samples were then etched using an alkaline electrolytic process and photographed on a Leica Reichert MEF4-A microscope at 1000X using differential interference contrast to enhance the microstructural features.

RESULTS

For the B_4C materials sintered by microwave heating, the die-pressed and the Gelcast materials had comparable densities, with the Gelcast material being slightly higher at 94% of theoretical. The hardness and toughness values were also comparable. Further sintering studies are being conducted and improved methods for temperature measurement in the microwave cavity are being explored. For specimens sintered conventionally, those produced by gelcasting exhibit improvements in the final properties when compared to samples formed by "conventional" methods. For the B4C samples processed by conventional sintering, the densities were ~8% higher for those formed by Gelcasting (95% of theoretical) as compared to die-pressed-isopressed samples, (only 87 % of theoretical density). The higher densities of the sintered gelcast materials resulted in improved mechanical properties.

Densities of the sintered materials are compared in figure 1. Note for all graphs in figures1-4: the specimens A2-A4 were sintered by microwaves, the specimens B1-B2 were sintered conventionally). Hardness and toughness measurements are compared in figure 2. Correlations of density, hardness and toughness are shown in figure 3.

Typical microstructures are presented in figure 4. In the microwave sintering experiments, it was apparent that the expected sintering temperature of 2250°C was exceeded, as evidenced by excessive grain growth and crystallographic twinning of some grains[4] (specimens A2,A3,A4 in figure 4). After sintering, the average grain size of samples fabricated by Gelcasting was ~ 5 μm compared to ~10 μm for the microwave sintered material. For the specimens sintered conventionally, the Gelcast specimen (B1 in figure 4) exhibited finer grain size, a more uniform microstructure, and improved mechanical properties.

† Sigma Chemical Co., St. Louis, MO.

CONCLUSIONS

Although the expected sintering temperature of 2250°C was beleived to be exceeded, the densification of B4C by microwave energy is an effective method for densification. As further studies are conducted and improved methods for temperature measurement in the microwave cavity are resolved, it is expected that improvements in mechanical properties will follow. For the comparison of Gelcast vs die-pressing forming processes, specimen formed by Gelcasting exhibited finer grain size, more uniform microstructures, and improved mechanical properties.

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DISCLAIMER

Fig. 1

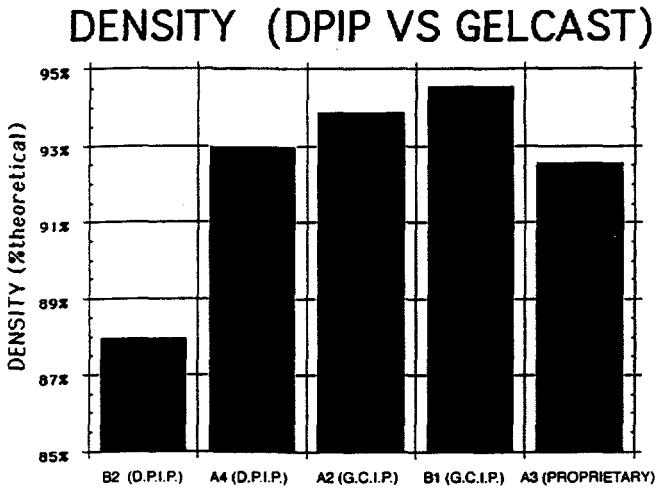
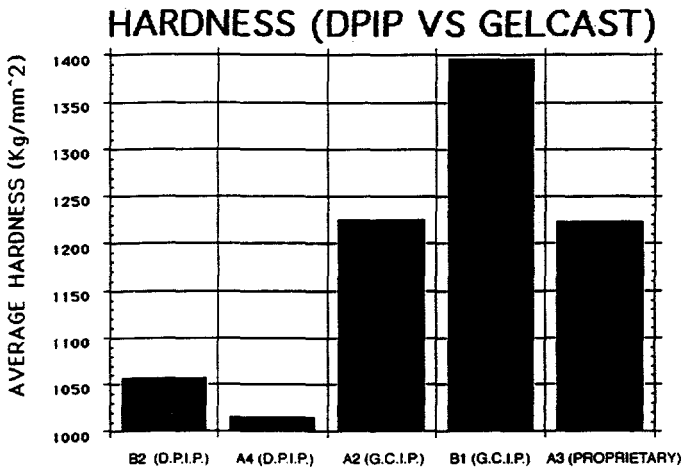


Fig. 1. Density values of die-pressed vs. Gelcast specimens are compared.

Fig. 2



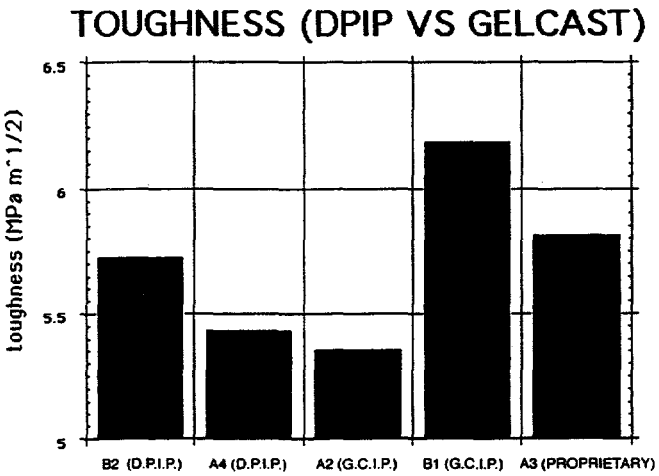


Fig. 2. Hardness and toughness measurements of die-pressed vs. Gelcast specimens are compared.

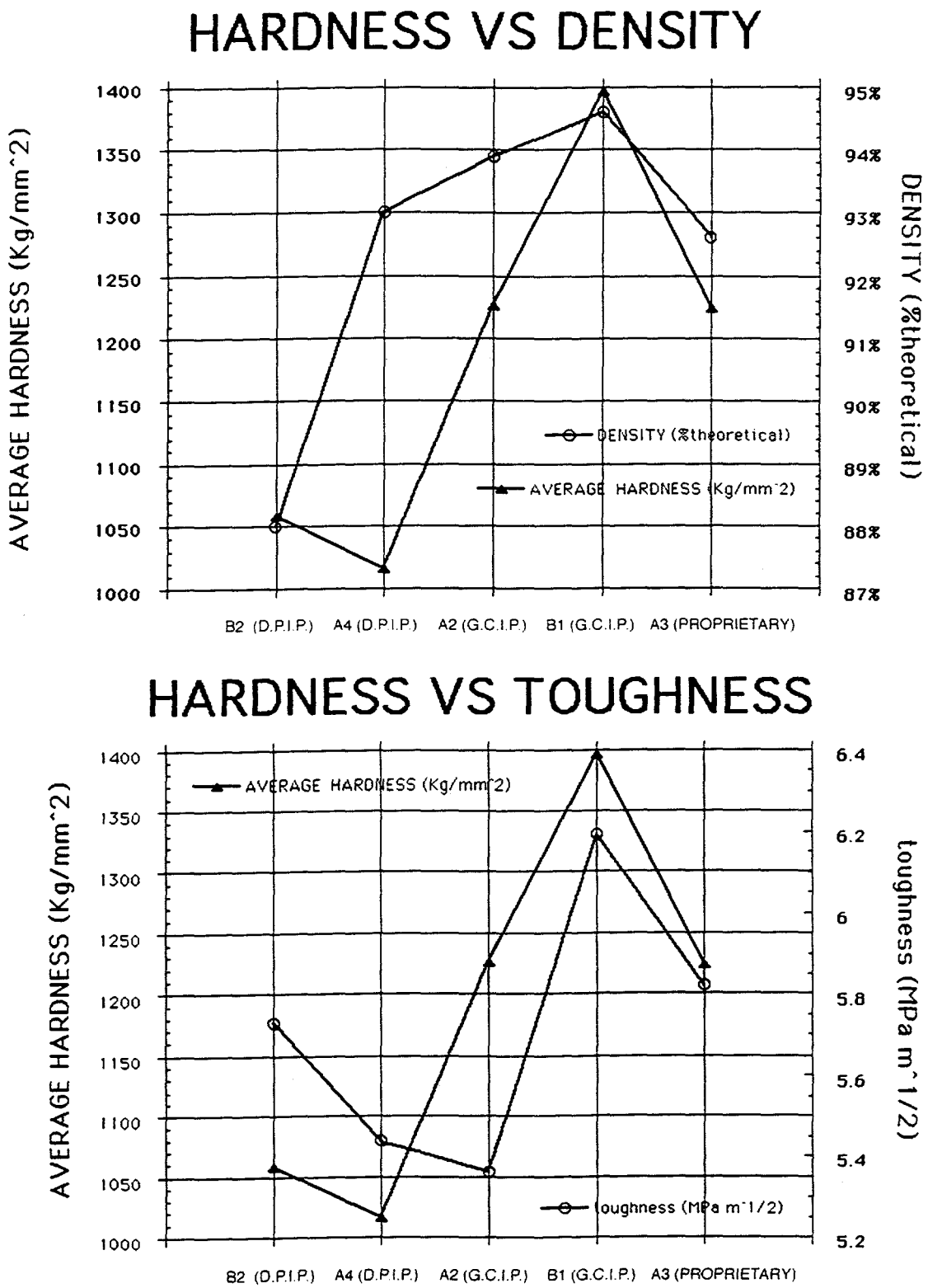


Fig. 3. Correlations of density, hardness and toughness values of die-pressed vs. Gelcast specimens are compared.

Fig. 3

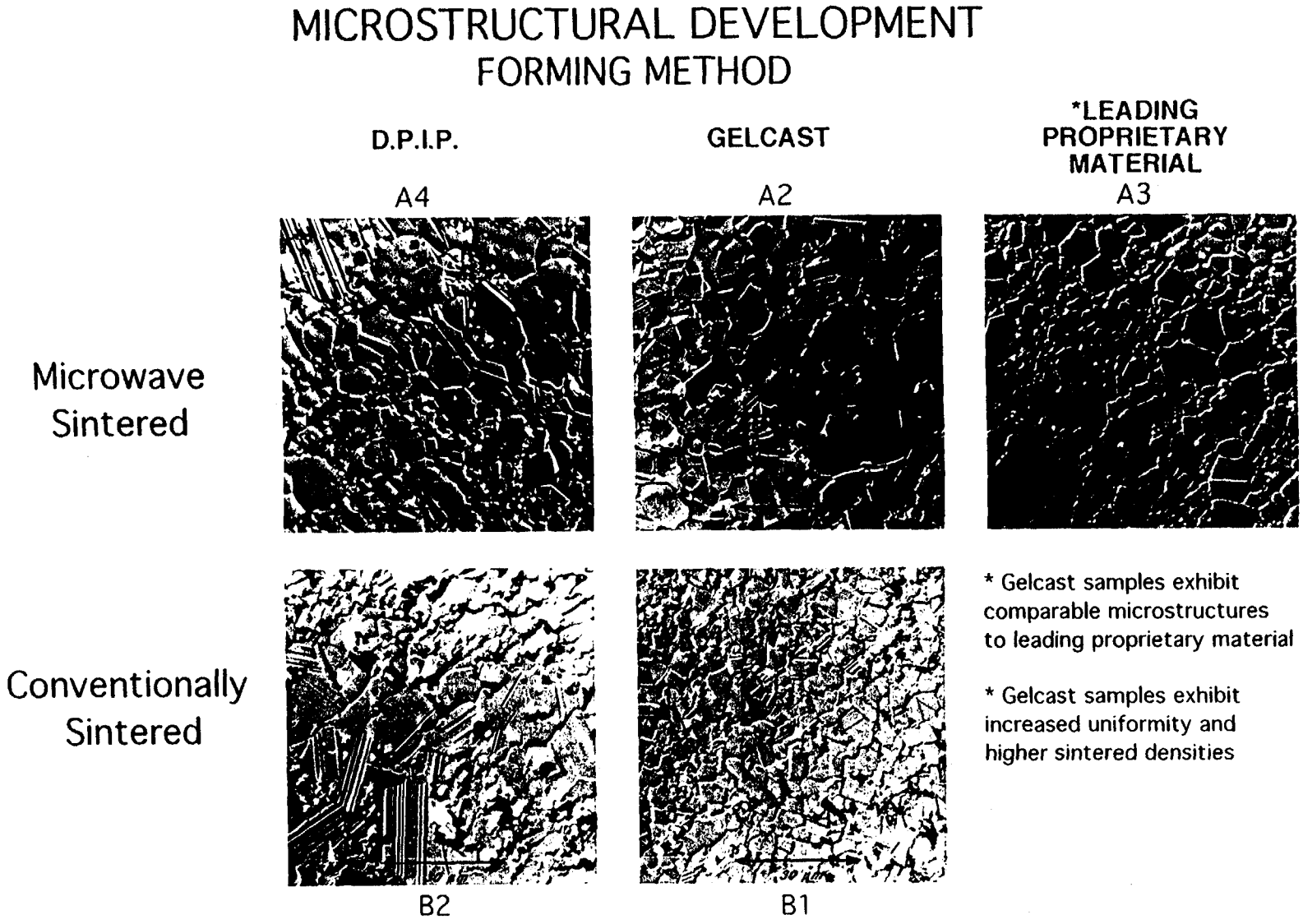


Fig. 4. Microstructures of die-pressed vs. Gelcast specimens are compared.

Fig. 4