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FABRICATION OF URANIUM CARBIDE/BERYLLIUM
CARBIDE/GRAPHITE EXPERIMENTAL-FUEL-ELEMENT
SPECIMENS

W. A. Muenzer

July 1978

MASTER

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OAK RIDGE Y-12 PLANT
OAK RIDGE, TENNESSEE

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FABRICATION OF URANIUM CARBIDE/BERYLLIUM CARBIDE/GRAPHITE EXPERIMENTAL-FUEL-ELEMENT SPECIMENS

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ABSTRACT

A method has been developed for fabricating uranium carbide/beryllium carbide/graphite fuel-element specimens for reactor-core-meltdown studies. The method involves milling and blending the raw materials and densifying the resulting blend by conventional graphite-die hot-pressing techniques. It can be used to fabricate specimens with good physical integrity and material dispersion, with densities of greater than 90% of the theoretical density, and with a uranium carbide particle size of less than 10 μm .

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SUMMARY

A fabrication process has been developed for fuel-element specimens containing enriched uranium carbide (UC_2), beryllium carbide (Be_2C), and graphite (C). The process involves milling uranium hydride powder, beryllium carbide powder, graphite flour, and a carbonizable binder in a solvent. After milling, the mixture is evaporated to dryness and densified by conventional graphite die hot pressing at 12.5 MPa and 1850° C. The uranium hydride is carbided, in situ, during the densification process.

A series of 12 developmental densifications were made, using depleted uranium to determine the appropriate steps necessary to achieve a satisfactory density ($> 85\%$ TD), UC_2 particle size ($< 10\ \mu\text{m}$), dispersion, and physical integrity. Parameters that were varied included the compounds of uranium, the type of graphite flour, volume percents of beryllium carbide and carbonizable binder, and total milling time. Uranium loading densities of 0.450 to 0.500 g/cm³ and beryllium carbide contents of approximately 30, 50, and 70 vol % were obtained. A maximum porosity of 15% and a maximum uranium carbide particle size of 10 μm were also achieved. Four densification pressings were made, using a four-holed die body-and-punch assembly. Sixteen enriched uranium fuel-element specimens were produced from the four pressings.

INTRODUCTION

Fuel-element specimens containing enriched uranium carbide, beryllium carbide, and graphite were prepared at the Oak Ridge Y-12 Plant^(a) for Sandia - Albuquerque.^(b) The specimens were to be used in core-meltdown studies, utilizing the fast-burst reactor located at Sandia. The specimens were to have a uranium density of 0.450 to 0.500 g/cm³ and contain 30, 50, and 70 vol % beryllium carbide. The remaining volume was to consist of graphite flour, a binder-derived carbon, and porosity. A maximum porosity of 15 vol % and a maximum uranium carbide particle size of 10 μ m were requested.

The fabrication process was to involve blending uranium oxide or hydride powder, beryllium carbide powder, graphite flour, and a carbonizable binder dissolved in a solvent; milling the slurry to deagglomerate and disperse the raw materials; evaporating the solvent to dryness, and densifying the dry blend by conventional graphite die hot pressing (and carbiding of the UO₂ or UH₃) at 12.5 MPa and 1850^o C. Twelve experimental densifications were to be made to determine satisfactory raw materials and to optimize the process parameters. Following this, 16 fuel-element specimens were to be made, containing enriched uranium. Eight of the specimens will contain 30 vol % Be₂C and eight will contain 70 vol % Be₂C. Sandia requested that no 50 vol % Be₂C specimens be made. The specimens were shipped to Sandia for core-meltdown studies.

(a) Operated by the Union Carbide Corporation's Nuclear Division for the Department of Energy.

(b) This work was initiated and partially funded by R. H. Marion of the Ceramics Development Division, 5846, Sandia.

PREPARATION OF EXPERIMENTAL-FUEL-ELEMENT SPECIMENS

CHARACTERIZATION OF THE RAW MATERIALS

High-surface-area (small-particle-size) beryllium carbide and uranium carbide particles were required to achieve maximum density and dispersion in the densified specimens. Available $-44\text{ }\mu\text{m}$ Be_2C was used in the fabrication experiments and is characterized in Table 1.

Table 1
CHARACTERIZATION OF BERYLLIUM CARBIDE, URANIUM OXIDE,
AND URANIUM HYDRIDE RAW MATERIALS

Element	Beryllium Carbide	Uranium Oxide	Uranium Hydride
Beryllium (wt %)	58.85	-	-
Uranium (wt %)	-	87.45	98.51
Carbon (wt %)	40.12	< 0.01	0.01
Oxygen (wt %)	1.00	12.16	0.25
Hydrogen (wt %)	-	-	1.21
Nitrogen (ppm)	190	-	1200
Total Elemental Impurities (ppm)	2675	163	135
Surface Area (m^2/g)	-	6.0	1.0
Screen Size, Agglomerates (μm)	44	44	125
Minor Phases	C(graphite), BeO, Be	None Detected	None Detected

Low-surface-area UC_2 ($\sim 0.01\text{ m}^2/\text{g}$) was available, but would have required extensive grinding to reduce the particle size due to its hardness. In the fabrication experiments, the UC_2 was formed from UO_2 and UH_3 , in situ, during the hot-pressing operation, the graphite flour filler being the source of carbon. The UO_2 ^(c) had a surface area of approximately $6\text{ m}^2/\text{g}$ and was screened to $-44\text{ }\mu\text{m}$ to remove the larger agglomerations; the UH_3 had a surface area of approximately $1\text{ m}^2/\text{g}$. Both the UO_2 and UH_3 are also characterized in Table 1. Graphite flours used include natural-flake graphite (NFG),^(d) graphitized Santa Maria coke,^(e) and GL-1008 coke.^(f) The powders were screened to $-420\text{ }\mu\text{m}$ and heated for approximately 16 hours at 150°C to remove moisture. Acenaphthylene (ACN), with a coke yield of approximately 39%, was used as a binder, and toluene was used as its solvent.

PARAMETER STUDY AND EVALUATION

Two experiments were conducted without Be_2C to evaluate the in situ carbiding of UO_2 during hot pressing and the effectiveness of the ACN binder. Uranium dioxide, ACN, and NFG flour were blended in one liter of toluene in a Hobart blender.^(g) The blending was

(c) A product of Tennessee Nuclear Specialities, Incorporated, Jonesboro, Tennessee.

(d) A product of Asbury Graphite Mills, Incorporated, Asbury, New Jersey.

(e) A product of Union Oil Company, San Francisco, California.

(f) A product of Great Lakes Carbon Company, Niagara Falls, New York.

(g) A product of the Hobart Manufacturing Company, Troy, Ohio.

continued beyond the slurry stage until all the solvent had evaporated, leaving the binder dispersed in the UO_2 and NFG.

In the first experiment, dry-blended powder, containing UO_2 , ACN (60 pph), and NFG (a UO_2 -to-graphite ratio of 0.95), was loaded into each hole (33 mm D) of a three-hole die. The powder was densified at 12.5 MPa and 1850°C (a one-hour hold at 1850°C) under an argon atmosphere, using the heating cycle outlined in Figure 1. Pressure was applied at the start of the heating cycle. During the densification, some of the material extruded into the clearance between the punches and die while the ACN was in a liquid state (ACN melts at approximately 92°C and thermosets at approximately 430°C). The specimens had good physical integrity, indicating satisfactory binding. X-ray diffraction analyses of the specimens showed UC_2 and UO_2 as the major constituents, indicating incomplete carbiding of the UO_2 . In the second experiment, dry-blended powder containing UO_2 , ACN (30 pph), and NFG (a UO_2 -to-graphite weight ratio of 0.75) was densified at 12.5 MPa and 1850°C (a one-hour hold at 1850°C) in a single-hole (76-mm-D) die, using the same heating cycle. The pressure was not applied until the temperature reached 600°C , and there was no evidence of extrusion. The specimen had good physical integrity at this binder content, and UO_2 was again a major constituent.

Figure 1. HEATING CYCLE FOR FUEL-ELEMENT-SPECIMEN DENSIFICATION.

A series of ten experiments were then designed to determine: (1) a heating cycle that would permit complete carbiding of the starting uranium compound; (2) satisfactory uranium and graphite flour starting materials; (3) the feasibility of preparing specimens containing 30, 50, and 70 vol % Be_2C ; (4) a minimum satisfactory binder content, and (5) the milling time necessary to effect maximum dispersion of the UC_2 , Be_2C , and graphite. Milling of the powder/toluene slurry was considered necessary, due to the tendency of the starting materials (UO_2 , UH_3 , Be_2C , and graphite) to agglomerate. The parameters of each experiment are listed in Table 2. The desired final uranium density of 0.500 g/cm^3 was used to calculate the amount of UO_2 or UH_3 feed added.

In an argon-atmosphere glovebox, the uranium compound (UO_2 or UH_3), Be_2C , graphite flour, and ACN were combined with one liter of toluene in a polyethylene container. Ceramic balls ($\sim 25\text{ mm D}$) were added, and the sealed container was placed on a rolling

Table 2
EXPERIMENTAL PARAMETERS FOR DEVELOPMENTAL DENSIFICATIONS

Experiment Number	Target Percent Beryllium Carbide (vol %)	Target Uranium Density (g/cm ³)	Starting Uranium Compound	Type Graphite Flour Feed	Milling Time (hr)	Acenaphthylene Binder Content (ppm)
9	30	0.500	UO ₂	GL-1008 ⁽¹⁾	16	0
11	30	0.500	UO ₂	GL-1008	16	10
14	30	0.500	UH ₃	GL-1008	16	10
4	50	0.500	UO ₂	Natural Flake Graphite ⁽²⁾	8	10
5	50	0.500	UH ₃	Santa Maria ⁽³⁾	8	10
12	50	0.500	UH ₃	GL-1008	16	0
13	70	0.500	UH ₃	GL-1008	8	0
6	70	0.500	UO ₂	GL-1008	8	0
7	70	0.500	UO ₂	GL-1008	16	10
8	70	0.500	UO ₂	GL-1008	8	10

(1) A product of the Great Lakes Carbon Company, Niagara Falls, New York.

(2) A product of the Asbury Graphite Mills, Incorporated, Asbury, New Jersey.

(3) A product of the Union Oil Company, San Francisco, California.

mill for eight (short milling time) or 16 (long milling time) hours. After milling, the slurry was taken back into the glovebox and the toluene evaporated while blending in the Hobart blender. The powder mixture was then loaded in equal amounts into each hole (33 mm D) of a three-hole die and densified at 12.5 MPa and 1850° C, using the same heating cycle. The hold time at 1850° C was extended to three hours to ensure complete conversion of the UO₂ or UH₃ to UC₂. The furnace was allowed to cool to room temperature before removing the die and punch assembly and unloading the specimens. The densified specimens were handled in an argon atmosphere to prevent the Be₂C and UC₂ from reacting with the moisture in the air. Chemical analyses of the densified specimens are presented in Table 3.

Table 3
FUEL-ELEMENT SPECIMENS—DEPLETED URANIUM

Experiment Number	Geometric Density (g/cm ³)	Theoretical Density (g/cm ³)	Percent of TD	Uranium Density (g/cm ³)	Percent Beryllium Carbide (vol %)	Total Carbon (wt %)	Fixed Carbon (wt %)	Percent Oxygen (wt %)	Minor Phases ⁽¹⁾
9	2.12	2.82	75	0.394	22.33	62.14	21.32	1.77	BeO, UC
11	2.30	2.72	85	0.361	23.12	51.66	6.06	1.91	UN, U ₂ N ₃
14	2.71	2.75	98	0.476	29.37	57.21	10.77	0.22	BeO, Be(OH) ₂
4	2.47	2.88	86	0.502	47.28	47.29	20.71	0.70	BeO, UC
5	2.70	2.94	92	0.566	49.71	45.73	22.21	0.34	BeO, UC
12	2.28	NA ⁽²⁾	NA	NA	(50) ⁽³⁾	NA	NA	NA	Be(OH) ₂ , UC
13	2.83	2.87	99	0.500	73.86	37.69	25.44	0.91	BeO
6	2.34	NA	NA	NA	(70) ⁽³⁾	NA	NA	NA	Be(OH) ₂ , UC
7	2.61	NA	NA	0.490	68.47	NA	NA	NA	BeO, UC
8	2.57	NA	NA	0.455	(70) ⁽³⁾	NA	NA	NA	NA

(1) By X-ray diffraction analyses.

(2) Not analyzed.

(3) Target vol %.

The three-hour hold at 1850° C was sufficient time to effect complete carbiding of both UO₂ and UH₃ since X-ray diffraction analyses indicated no UO₂ or UH₃ in the finished specimens. Specimens produced, using UH₃ as the starting material, had higher densities than those produced using UO₂. Experiments 7 and 13 were identical, with the exception of the different uranium starting compounds (UO₂ in Experiment 7 and UH₃ in Experiment 13). However, specimens from Experiment 7 had densities of 2.61 g/cm³, whereas those from Experiment 13 had densities of 2.83 g/cm³. Similarly, specimens from Experiment 4 (UO₂ feed) had densities of 2.47 g/cm³ and the one from Experiment 5 (UH₃ feed) had a density of 2.70 g/cm³. The greater densities achieved by using UH₃ instead of UO₂ can probably be attributed to the different carbiding reactions of the two compounds. The carbiding reaction of UO₂ involves a single reaction:

which does not occur until the temperature reaches approximately 1800° C.¹ At this temperature, densification of the powder blend was essentially complete in this operation. Any outgassing of carbon monoxide would probably have introduced porosity, resulting in a decrease in density. On the other hand, the UH₃ dehydrided during hot pressing at approximately 300° C, and the evolved hydrogen gas escaped before any densification had occurred (the pressure was not applied until a temperature of 600° C was reached). The dehydrided uranium then was carbided at approximately 600° C, and no gaseous reaction products were formed:²

Although three different types of graphite flour were used, GL-1008 graphitized coke was used in the majority of the experiments because it had a lower coefficient of thermal expansion than the Santa Maria graphitized coke and a higher degree of isotropicity than the NFG.³

Satisfactory specimens were produced with 30, 50, and 70 vol % Be₂C. The 70 vol % Be₂C specimens had a slightly greater percent of the theoretical density than the lower vol % Be₂C specimens (when all other parameters were constant) due to the superior densification properties of Be₂C compared to graphite flour. Specimens from Experiment 13 (70 vol % Be₂C, UH₃ feed) were 99% of the theoretical density (TD), compared to 98% TD for specimens from Experiment 14 (30 vol % Be₂C, UH₃ feed).

A binder content of 10 pph was determined to be necessary and sufficient to provide good physical integrity of the specimens. All specimens made without the ACN binder (Experiments 6, 9, and 12) had cracks throughout, and several fell apart upon removal from the die. Specimens made with 10 pph binder had no cracks, and a diamond-tipped saw was required to cut analytical samples from the specimens.

Although milling the mixture was considered necessary to achieve satisfactory dispersion of the UC₂, Be₂C, and graphite, there was no discernable difference in the dispersions in

specimens produced with a short milling time (8 hours) or a long milling time (16 hours). The powder/toluene slurry for Experiment 7 was milled for 16 hours; the slurry for Experiment 8 was milled for 8 hours. Photomicrographs of pieces from the two specimens (Figures 2 and 3) showed no noticeable difference in the dispersions. Further evidence of good UC_2 dispersion is shown in the secondary electron images and uranium X-ray responses of the same areas of specimens from Experiment 5 (Figure 4) and Experiment 7 (Figure 5). Fewer UC_2 particles were visible in the SEM and uranium X-ray response of the specimen from Experiment 5 which had a higher carbon content (~ 45 vol %) than the

Figure 2. SPECIMEN FROM EXPERIMENT 7. (70 Vol % Be_2C ; 500X)

Figure 3. SPECIMEN FROM EXPERIMENT 8. (70 Vol % Be_2C ; 500X)

specimen from Experiment 7 (~ 35 vol % C). Since UH_3 and UO_2 utilized the graphite flour as a source of carbon in the carbiding reaction, the majority of UC_2 was located in or at the boundary of the graphite agglomerations and could be pulled from the specimen surface when it was being polished for the photomicrographs. Thus, the higher the carbon content of a specimen, the fewer UC_2 particles would be seen in the SEM and in the X-ray response.

The specimens produced in Experiments 4 through 14 had oxygen contents ranging from 0.22 to 1.91 wt %. The majority of the oxygen was from the BeO which was present in the

(a) Scanning Electron Microscope Photomicrograph. 172387

(b) Uranium X-Ray Response. 172388

Figure 4. SPECIMEN FROM EXPERIMENT 5. (50 Vol % Be_2C ; 500X)

(a) Scanning Electron Microscope Photomicrograph. 172389

(b) Uranium X-Ray Response. 172392

Figure 5. SPECIMEN FROM EXPERIMENT 7 (70 Vol % Ba_2C , 500X)

Be₂C starting material. Aluminum contents of 0.5 to 1.0 wt % were found in the finished specimens. (The aluminum contamination was traced to the aluminum blade of the Hobart blender which apparently was worn away by the particulate Be₂C during the solvent evaporation step.) Specimens produced in Experiment 11 were found to contain two uranium nitrides (UN and U₂N₃), instead of UC₂. This change was due to an operational error in which the densification run was under a nitrogen gas blanket instead of argon, causing the UO₂ feed to form the nitride instead of the carbide.

The best specimens produced in the experimental densifications were those from Experiments 13 (70 vol % Be₂C, UH₃ feed) and 14 (30 vol % Be₂C, UH₃ feed). The

Figure 6. SPECIMEN FROM EXPERIMENT 13. (70 Vol % Be₂C; 500X)

specimens were 99% and 98% TD, respectively, and photomicrographs (presented in Figures 6 and 7) reveal well-dispersed Be_2C , graphite, and UC_2 . Few UC_2 particles are visible in Figure 7 as they were pulled from the surface when the specimen was polished. Uranium carbide dispersion of the specimen from Experiment 13 is further defined by the SEM and X-ray responses (of the same area), shown in Figure 8.

Figure 7. SPECIMEN FROM EXPERIMENT 14. (30 Vol % Be_2C ; 500X)

Results of the experimental densifications were then used to determine the best parameter choices to be used in the fabrication of the enriched uranium fuel-element specimens. The heating cycle given in Figure 1 with a three-hour hold at 1850°C was found to be

(a) Scanning Electron Microscope Photomicrograph.

172391

(b) Uranium X-Ray Response.

172390

Figure 8. SPECIMEN FROM EXPERIMENT 13. (70 Vol % Be_2C ; 500X)

satisfactory in preparing sufficiently dense specimens and in allowing the complete conversion of the uranium starting compound to UC_2 . Uranium hydride was superior to UO_2 as a starting uranium compound in that higher-density specimens were produced with the UH_3 . Milling of the slurry was used to achieve good UC_2 , Be_2C , and graphite dispersions in the densified specimens, but there were no noticeable dispersion differences whether the slurry was milled for 8 or 16 hours. For convenience sake, a 16-hour milling time was selected since this would make it possible to mill the slurry overnight. An ACN binder content of 10 pph was judged to be necessary and sufficient to obtain good physical integrity of the specimens. Satisfactory specimens were made with 30, 50, and 70 vol % Be_2C . For the enriched uranium fuel-element specimens, 30 and 70 vol % Be_2C contents were chosen as this would permit performance evaluation (high and low Be_2C contents) with a minimum of reactor testing.⁴

ENRICHED-FUEL-ELEMENT SPECIMEN FABRICATION

Sixteen fuel-element specimens, containing fully enriched uranium, were produced. Four individual specimens were produced in each of four densification runs. Eight of the 16 specimens (2 runs) were designed to have uranium densities of 0.450 to 0.500 g/cm³ and a Be_2C content of 30 vol %; the other eight specimens were designed to have uranium densities of 0.450 to 0.500 g/cm³ and a Be_2C content of 70 vol %.

For Densification Runs I and II (30 vol % Be_2C), 45.1 g UH_3 , 65.2 g Be_2C , 115.8 g GL-1008 graphitized coke, and 22.6 g ACN (10 pph) were added to one liter of toluene, and the slurry was milled for 16 hours. The toluene was evaporated while blending, and 60.7 g of the dried powder (11.0 g UH_3 , 15.9 g Be_2C , 28.3 g GL-1008, and 5.5 g ACN) were loaded into each hole (33 mm D) of a four-hole die. The powder was densified at 12.5 MPa and 1850° C with a three hour hold at 1850° C. After unloading the die, six of the 33-mm-diameter by 26-mm-high specimens were cut, using a diamond-tipped saw, into 33-mm-diameter by 13-mm-high and 33-mm-diameter by 6.5-mm-high specimens. Two each 26-mm-high, four each 13-mm-high, and five each 6.5-mm-high 30 vol % Be_2C specimens were provided for fuel-element evaluation at Sandia. For Densification Runs III and IV (70 vol % Be_2C), the same procedure was followed. This slurry contained 45.1 g UH_3 , 152.1 g Be_2C , 39.0 g GL-1008 graphitized coke, and 23.6 g ACN (10 pph). When dried, 63.4 g of the blended powder (11.0 g UH_3 , 37.1 g Be_2C , 9.5 g GL-1008, and 5.8 g ACN) were loaded into each hole of the four-hole die. Two each 26-mm-high, four each 13-mm-high, and five each 6.5-mm-high 70 vol % Be_2C specimens were also provided for fuel-element evaluation at Sandia. Chemical analyses of the specimens are presented in Table 4 and representative photomicrographs are included as Figures 9 and 10.

The 30 vol % Be_2C specimens were approximately 89% TD (Runs I and II) and had uranium densities of 0.442 and 0.444 g/cm³, respectively, which were slightly below the requested range of 0.450 to 0.500 g/cm³. Identically made specimens with depleted UH_3 starting material (Experiment 14, Table 3), instead of enriched UH_3 , were 98% TD. The enriched UC_2 loading is not visible in the photomicrograph (Figure 9) for the same reason noted for the depleted- UC_2 -loaded, 50-vol % Be_2C specimen seen in Figure 4.

Table 4
CHEMICAL ANALYSES OF ENRICHED URANIUM FUEL ELEMENT SPECIMENS

Densification Run	Geometric Density (g/cm ³)	Theoretical Density (g/cm ³)	Percent of TD	Uranium Density (g/cm ³)	Percent Beryllium Carbide (vol %)	Total Carbon (wt %)	Fixed Carbon (wt %)	Percent Oxygen (wt %)	Minor Phases ⁽¹⁾
I	2.45	2.75	89	0.442	28.27	NA ⁽²⁾	NA	NA	NA
II	2.46	2.75	89	0.444	27.90	62.20	15.35	2.27	BeO, UC
III	2.63	2.64	100	0.264	66.53	43.84	26.52	NA	NA
IV	2.54	2.58	98	0.204	68.80	45.25	27.17	2.63	BeO, UC

(1) By X-ray diffraction analyses.

(2) Not analyzed.

Figure 9. SPECIMEN FROM RUN II. (30 Vol % Be₂C; 500)

Figure 10. SPECIMEN FROM RUN III. (70 Vol % Be_2C ; 500X)

The 70-vol % Be_2C specimens were 100% TD (Run III) and 98% TD (Run IV), and had a uranium density of 0.264 g/cm^3 . Identically made specimens with depleted UH_3 starting material (Experiment 13, Table 3) were 99% TD. The low uranium densities were probably due to an operational error in that insufficient UH_3 starting material was added to the slurries. The enriched UC_2 loading in the 70 vol % Be_2C specimens (Run III) is only slightly visible in the photomicrograph (Figure 10) due to the fact that insufficient UH_3 starting material was added to the slurry.

CONCLUSIONS

It can be concluded from the experimental densifications and the enriched-fuel-element fabrications that enriched $\text{UC}_2/\text{Be}_2\text{C}/\text{C}$ fuel elements with a length-to-diameter ratio (L/D) less than or equal to 0.75 can be made with good physical integrity and material dispersion, have densities $\geq 90\%$ TD, and a UC_2 particle size of $\leq 10\ \mu\text{m}$. This goal can be accomplished by: (1) using enriched UH_3 as a starting material (the UC_2 being formed, in situ, during hot pressing); (2) using 10 pph ACN as a binder; (3) using Be_2C in quantities which would give 30 to 70 vol % Be_2C in the finished specimens; (4) using graphitized coke (GL-1008) as a carbon filler and source of carbon for the carbiding reaction of UH_3 to UC_2 ; (5) milling the starting materials slurried in toluene and evaporating the toluene while blending, and (6) using a conventional-graphite-die (1 to 4 holes) hot pressing at 12.5 MPa and 1850°C , with the heating cycle given in Figure 1.

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