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Processing and characterization of B_4C/Cu graded composite as plasma facing component for fusion reactors

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Abstract: A new approach for fabricating B_4C/Cu graded composite by rapid self-resistance sintering under ultra-high pressure was presented, by which a near dense B_4C/Cu graded composite with a compositional spectrum of 0-100% was successfully fabricated. Plasma relevant performances of sintered B_4C/Cu composite were preliminarily characterized, it is found that its chemical sputtering yield is 70% lower than that of SMF800 nuclear graphite under 2.7 keV D^+ irradiation, and almost no damages after 66 shots of in situ plasma discharge in HL-1 Tokamak facility, which indicates B_4C/Cu plasma facing component has a good physical and chemical sputtering resistance performance compared with nuclear graphite.

Key words: boron carbide; plasma facing component (PFC); functionally graded material (FGM); composite

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The first wall in Tokamak is subject to the incidence of not only high heat flux (0.1-1 MW/m^2 during normal operation) but also runaway energetic particles which would cause erosion of plasma facing wall surface and may result in an uncontrolled influx of impurities into plasma. The conditions that exist in a fusion reactor often require very specialized materials; it is desirable to have the material composed of low-Z elements to minimize radiative cooling of the plasma. Graphite has been considered as one of the candidate plasma facing materials because of its high thermal shock resistance, however, there are several problems associated with its use such as enhancement of hydrogen recycling, large erosion due to oxygen and radiation enhanced sublimation where the temperature exceeds about 1 000℃[1] that urges the fusion designers to search for alternative materials. Boron carbide has the advantages of being a low-Z material, a good oxygen getter and having a high melting point [2] and will be one of the most competitive candidates, but its low thermal conductivity and brittleness are the main shortage of this material. The combination of B_4C ceramic with high thermal conductivity and mechanical properties of copper is of interest as first wall plasma facing material. Many processes such as plasma spray, CVD, vacuum arc deposition, in situ glow discharge and so on have been proposed to form B_4C coating/film on carbon-based material or metal substrate [3-13], but the thickness is limited and/or crack even exfoliation on the interface of coating and substrate is always experienced due to great thermal stresses originated from their mismatch coefficients of thermal expansion. Without exception, the remarkably different physical properties of B_4C and copper, especially on melting point and CTE (coefficients of thermal expansion), always construct obstacles for material processing.

In the present work, in view of the reduction of thermal stresses, the concept of FGM is adopted and based on the characteristic of distinct resistivity between B_4C and Cu, the feasibility of using a new powder metallurgical process *via* self-resistance sintering under ultrahigh pressure for preparation of B_4C/Cu graded composite is explored. The plasma-relevant properties on chemical sputtering yield and physical sputtering damage of B_4C/Cu composite after in-situ plasma irradiation in a Tokamak facility are preliminarily reported.

1 Experimental procedures

In view of their distinct resistivity between B_4C and Cu, it can be expected that a gradual resistance distribution and thus a elevated temperature zone will be established along the compositional changing direction (from Cu to B_4C) when strong electric current passing through FGM green sample, the temperature profile in FGM during self-resistance sintering has been roughly

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deduced and it was approximately proportional to the square of thickness and current density applied [14]. By adjusting the inherent resistance distribution and external electric current input, graded sintering of B_4C/Cu FGM, which has characteristic of large melting point difference, may be practicable. However, ultrahigh pressure is required to shorten sintering time and accordingly inhibit elements macro diffusion in FGM specimen.

The experimental setup is schematically illustrated in **figure 1**. The device consisted of a mechanical press and associated electrical and hydraulic systems. A FGM assembly, which contained B_4C/Cu FGM green compact, was first placed in the pressure vessel. The pressure was applied by raising the bottom anvil with force provided by lower hydraulic ram. The sample assembly was encapsulated in pyrophyllite sleeve, which was acted as heat/electric insulator in graded sintering. Graphite and steel platelet were used as sealing and pressure-enforcing components respectively as well as conductors. The alternating current passed through and the FGM compact was mainly heated by joule exothermicity.

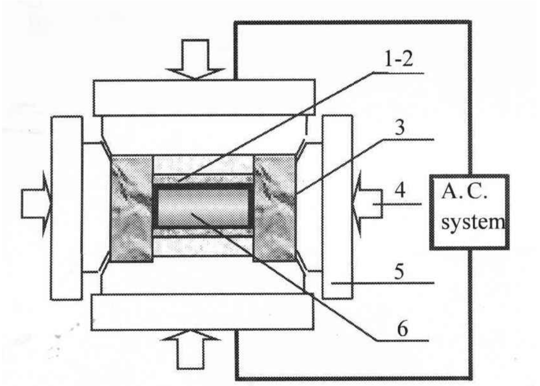

Figure 1 Schematic illustration of experimental setup. 1, 2—steel and graphite platelet; 3—pyrophyllite sleeve; 4—pressurized orientation; 5—anvil of WC hard alloy; 6—B_4C/Cu green compact.

B_4C powder with an average particle size of 0.2 μm, purity (mass fraction) of more than 95% (the balance are B_2O_3, SiC and C) and SiC powder (15% (mass fraction) of total ceramics) as a sintering additive of B_4C, with mean particle size of 0.15 μm and purity of more than 99%; copper powder with particle size less than 74 μm and purity of 99% were used as raw materials. B_4C-SiC powder and Cu powder of intermediate layers of B_4C/Cu FGM were mixed according to designed composition. The ratios of B_4C/Cu in the graded layers were determined by the following formula [15]: $C=\left(\frac{x}{d}\right)^p$, where, C is the volume fraction of ceramics; x the relative distance from the transition layer, d the thickness of the FGM layer and p the compositional distribution exponent. In this work the total number of layers of FGM is 6, the first layer is pure B_4C-SiC ceramics, while the sixth is pure copper. Different p value has different effects of thermal stress mitigation, in this work, four p values, *i.e.*, $p=0.6$, 1.0, 1.4, 1.8, were selected to evaluate the densification effect of graded sintering.

The mixture powders with different compositions above mentioned were stacked layer by layer in a steel mould to form green compact with dimension of $\phi 20\ \text{mm}\times 10\ \text{mm}$ pressed by a jack. The pressed B_4C/Cu FGM compact was integrated with graphite platelet, steel platelet and pyrophyllite sleeve to constitute sample assembly. Graded sintering was performed under pressures ranging from 2 000 to 4 000 MPa, electric power input was about 12 kW (with 7.5 A×1600 V) and sintering time was 40 s. After sample polishing and wax applying, the density of sintered B_4C/Cu FGM was measured by Archimedes method; the microstructure was examined by SEM (Scanning Electron Microscopy) and the plasma-relevant properties of B_4C/Cu FGM were tested by in-situ irradiation with high-energy deuterium beam.

2 Results and discussion

2.1 Densification effect

The densification effect of B_4C/Cu FGM was depicted as shown in **figure 2**, it can be seen that the density of 6-layered overall B_4C/Cu FGM reduces with the increase of compositional distribution factor (p). Higher p values in this work mean higher ceramics content in the FGM sample that may bring two synergic influences on densification of material sintering. Firstly, the sintering mechanism at monolithic ceramic layer belongs to solid-state diffusion for lack of liquid phase, of which process depends mostly on time, particle size,

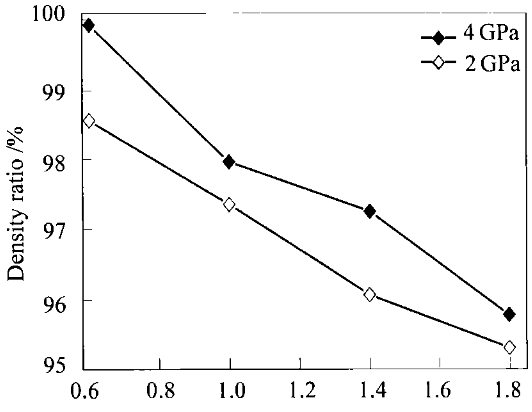

Figure 2 Effects of pressure on the densification of bulk B_4C/Cu composites with different p values.

sintering aid, *etc.*; in this case ceramics cannot be easily densified by rapid sintering. Secondly, the increase of ceramics content directly lead to the increment of resistance in the overall FGM, the high temperature by self-exothermicity and heat conduction on ceramics matrix dispersed by copper may cause partial evaporation of liquid copper. Besides, the dilative gas absorbed by raw fine powders forms pressurized bubbles and thus liquid copper was extruded out of its initial position and therefore po res maintain after rapid cooling.

Pressure has important influence on the densification, which can be also seen from figure 2 that the density of B_4C/Cu FGM elevates with the increase of pressure, but this factor is no longer an eminent one to some extent. It can be inferred that the consolidation mode including grain boundary slip (crystal shattering and plastic transforming) and extrusion creep is not a leading one when pressure exceeds 4 GPa, a pressure far great than that of conventional hot pressing. For further densification, amelioration of internal phases should be emphasized under such circumstances.

To verify above hypotheses, heat treatments of sintered B_4C/Cu FGM of $p = 1.0$ with original parameters of pressure of 2 GPa, 12 kW and 40 s were conducted and the results were shown in **figure 3**. It is obvious that a near dense (99.85% of theoretical density) has been achieved when secondary power ratio of 60% initial input and a 30 s duration of extra treatment were employed. Heat treatment, by analogy, can further improve ceramic sintering and facilitate liquid copper relocating or re-infiltrating into the apertures of ceramic matrix, that is to say the secondary stage of densification proceeds. Nevertheless, it can be reasonably expected that even in this case the large-scale diffusions between ceramics and copper are unlikely to happen due to very short heating duration, despite micro-migration in copper-rich layers is inevitable.

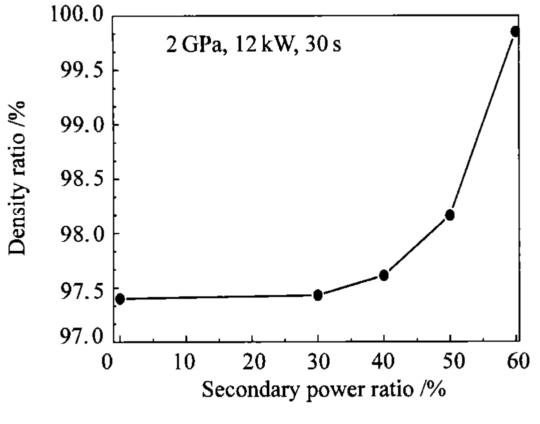

Figure 3 Effects of heat treatment on densification of bulk B_4C/Cu composite with p=1.0.

2.2 Microstructure analysis

Figure 4 displays the overall 6-layered FGM (after heat treatment with 60% power of initial input) backscattering image and a good graded compositional change is apparent, especially at the ceramics-rich side. However, it should be noticed that the layer boundaries are hard to be distinguished when copper content more than 60% (volume fraction), it might be attributed to copper's well-smelting and significant inter-diffusion (micro-flow) between B_4C and Cu. **Figure 5** demonstrates quite good sintering microstructure with fine and homogenous particle distribution in monolithic ceramic layer. From figure 4 and 5, it might be concluded that B_4C/Cu FGM can be well fabricated by itself resistance sintering under ultrahigh pressure; more importantly, the graded compositional transition initially designed can be also satisfactorily retained by delicate process control.

Figure 6 and **7** display the SEM of 20% (volume fraction) Cu layer after heat treatment and its elemental distribution of 5 dots selected which is quantitatively assayed by energy spectrum analysis. It can be observed that the matrix of fine B_4C and SiC particles is impregnated by liquid copper, which can be confirmed that heat treatment after sintering is conducive to the densification of FGM.

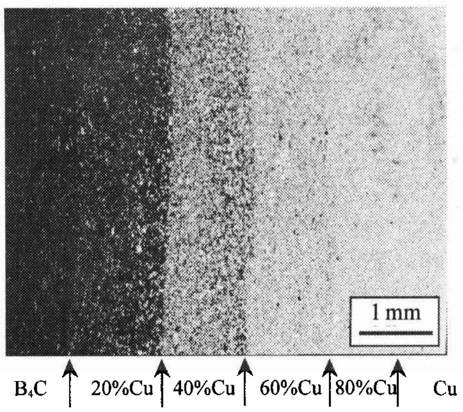

Figure 4 Back scattering image of 6-layered B_4C/Cu graded composite.

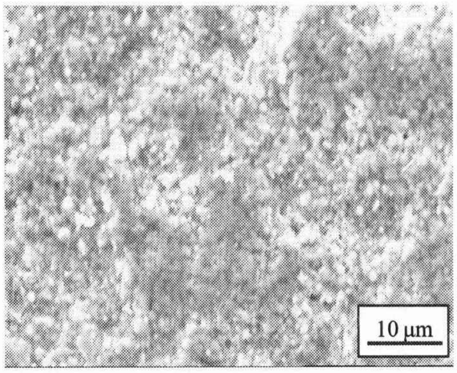

Figure 5 SEM of pure ceramic layer of B_4C/Cu FGM.

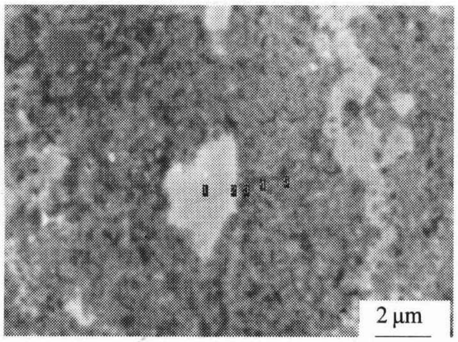

Figure 6 SEM of 20% Cu layer of B_4C/Cu FGM.

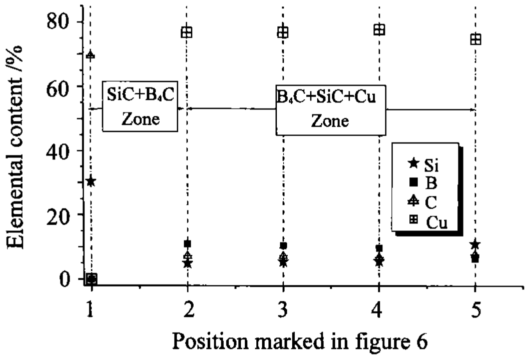

Figure 7 Elemental distribution of figure 6.

2.3 Properties characterization of B_4C/Cu PFC

(1) Chemical sputtering performance.

The chemical sputtering performance of B_4C/Cu plasma facing component was conducted in LAS-2000 apparatus. The experimental procedures were: firstly, vacuum degassing for about 20-30 min at about 1 073 K, then, after the temperature of the sample cooled down to room temperature (RT), a deuterium ion with 2.7 keV energy was implanted and the total fluence was 1.2×10^{18} ions. At the base pressure of 1×10^{-6} Pa, the thermal release spectra were measured by fast temperature rising from RT to 1 223 K. The result is shown in **figure 8**; the CD_4 production of B_4C/Cu FGM is 70% lower than that of SMF 800 nuclear graphite, and its peak value shifts 30 K to lower temperature. The low chemical sputtering yield of B_4C/Cu PFM reflects that the sintering bonding strength of B_4C ceramics is higher than those of graphite and further more the activation energy of CD_4 from B_4C eroded by D^+ should be higher than that of SMF800 nuclear graphite.

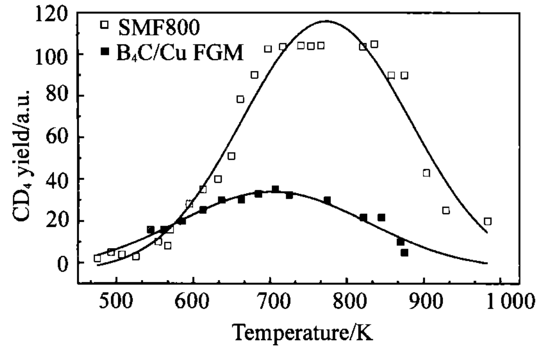

Figure 8 CD_4 production of B_4C/Cu PFC and graphite *vs.* temperature.

(2) Physical sputtering damage-resistant performance.

Physical sputtering damage-resistant property of B_4C/Cu PFC was performed in HL-1 Tokamak facility. The irradiation parameters are as follows: discharge times, 66; intensity of perpendicular magnetic field, 2 T; Toroidal current, 120-200 kA; average lifespan of plasma, 1.2 s; linear mean electron density, 1-1.4×10^{13}cm^{-3}; perpendicular electron density, (2-4)$\times10^{12}$cm^{-3}; perpendicular electron temperature, 100-200 eV.

The plasma physical sputtering damage of the surface of B_4C/Cu PFM can represent its physical sputtering performance, the morphology (of the same area selected) before/after in-situ plasma irradiation is shown in **figure 9**, no discernible cracks on the irradiated surface of B_4C/Cu PFC was found except for slightly etched trace. XRD (X-Ray Diffraction) results show no distinct change of crystalline structure, which means it has good thermal shock resistance and physical sputtering-resistance properties on the above irradiation conditions. While small increase in peak width existed in graphite SMF800 specimen, indicating change of crystalline style or increase of crystal lattice defects, besides, evident characteristics of plasma sputtering damage were also noticeable in graphite SMF800 [12].

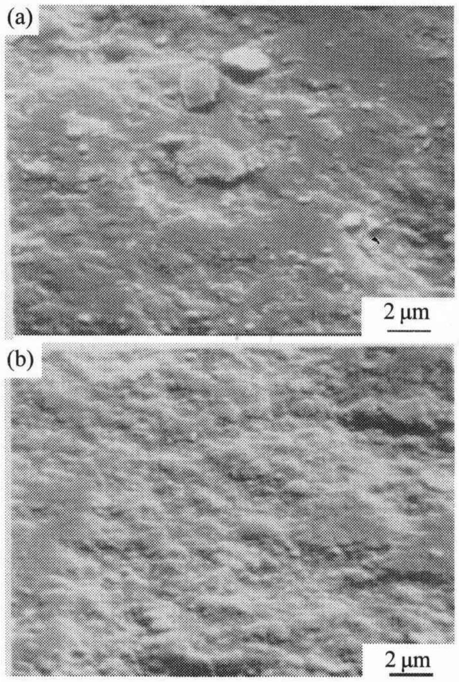

Figure 9 SEM micrographs of B_4C/Cu PFC before (a) and after in-situ plasma irradiation (b).

3 Conclusions

(1) A new approach for fabricating functionally graded material by rapid self-resistance sintering under ultra-high pressure was proposed, and a new B_4C/Cu composite with compositional spectrum of 0 to 100% was successfully prepared.

(2) Pressure and heat treatment have strong influence on the consolidation of B_4C/Cu FGM. A new dense graded B_4C/Cu composite has been obtained on conditions of 2 000-4 000 MPa applied, 12 kW power input for 40 s and subsequently proper heat treatment.

(3) The chemical sputtering yield of B_4C/Cu PFC is 70% lower than that of SMF800 nuclear graphite under 2.7 keV D^+ irradiation, and almost no damages on material surface after 66 shots of in-situ plasma discharge in HL-1 Tokamak facility. B_4C/Cu PFC has better physical and chemical sputtering resistance performance than that of nuclear graphite.

References

[1] V.Barabash, M.Akiba, and I.Mazul, *et al.*, Selection, development and characterisation of plasma facing materials for ITER [J], *J. Nucl. Mater.*, 233-237(1996), p.718.

[2] J.Winter, Comparison of tokamak operation with metallic getters、(Ti, Cr, Be) and boronization [J], *J. Nucl. Mater.*, 176-177(1990), p.14.

[3] K. Nakamura, M. Akiba, and S. Suzuki, *et al.*, High heat flux experiment on B_4C-overlaid C/C composites for plasma facing materials of JT-60U [J], *J. Nucl. Mater.*, 196-198(1992), p.627.

[4] S. Veprek, Large-area boron carbide protective coatings for controlled thermonuclear research prepared by in situ plasma CVD [J], *Plasma Chemistry and Plasma Processing*, 12 (1992), No.3, p.219.

[5] P.G. Valentine, P.W. Trester, and J. Winter, *et al.*, Boron carbide based coatings on graphite for plasma-facing component [J], *J. Nucl. Mater.*, 212(1994), p.1146.

[6] H. Bolt, M. Araki, and J. Linke, *et al.*, Heat flux experiments on first wall mock-ups coated by plasma sprayed B_4C [J], *J. Nucl. Mater.*, 233-237(1996), p.809.

[7] A. Yehia, R. Vassen, and R. Duwe, *et al.*, Ceramic SiC/B_4C/TiC/C composites as plasma facing components for fusion reactors [J], *J. Nucl. Mater.*, 233-237(1996), p.1266.

[8] I. Fujita, Y. Hirohata, and T. Hino, *et al.*, Thermal response for divertor mock-up using surface-modified CFC title [J], *J. Nucl. Mater.*, 241-243(1997), p.1185.

[9] W. Mallener, R. Duwe, and K.H. Rauwald, Plasma-sprayed coatings for plasma-facing components [J], *Proceedings-Symposium on Fusion Engineering*, 2(1997-1998), p.884.

[10] I. V. Opimach, O. I. Buzhinski, I.N. Arkhipov, *et al.*, Hydrogen isotope retention in B4C coating on RGT graphite under high heat fluxes of DIII-D divertor plasma [J], *Proceedings-Symposium on Fusion Engineering*, 2(1997-1998), p.881.

[11] T. Hino, Japanese universities' activities for PFC development and PMI studies [J], *Fusion Engineering and Design*, 39-40(1998), p.439.

[12] C.C. Ge, J.T. Li, and Z.J. Zhou, *et al.*, Development of functionally graded plasma-facing materials [J], *J. Nucl. Mater.*, 283(2000), p.1116.

[13] C.C. Klepper, J. Niemel, and E.J. Yadlowskv, *et al.*, Vacuum arc deposited boron carbide films for fusion plasma facing components [J], *Fusion Technology*, 39(2001), No.2, p.910.

[14] Y.H. Ling, C.C. Ge, and J.T. Li, *et al.*, Fabrication of SiC/Cu functionally gradient material by graded sintering [J], *Ceramic Transactions*, 111(2001), p.333.

[15] T. Hirano, J. Teraki, and T. Yamada, A fundamental study on the application of FGM to high-temperature rotating members, [in] edited by M. Yamanouchi, M. Koizumi, T. Hirai, I. Shiota, *Proc. 1st Int. Symp. On Functionally Gradient Materials (FGM'90)* [C], Functionally Gradient Materials Forum and The Society of Non-Traditional Technology, Tokyo, p.5.