Effects of metal binder on the microstructure and mechanical properties of Al_2O_3 -based micro-nanocomposite ceramic tool material

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Abstract: The Al_2O_3 –(W,Ti)C composites with Ni and Mo additions varying from 0vol% to 12vol% were prepared via hot pressing sintering under 30 MPa. The microstructure was investigated via X-ray diffraction (XRD) and scanning electron microscopy (SEM) equipped with energy dispersive spectrometry (EDS). Mechanical properties such as flexural strength, fracture toughness, and Vickers hardness were also measured. Results show that the main phases Al_2O_3 and (W,Ti)C were detected by XRD. Compound MoNi also existed in sintered nanocomposites. The fracture modes of the nanocomposites were both intergranular and transgranular fractures. The plastic deformation of metal particles and crack bridging were the main toughening mechanisms. The maximum flexural strength and fracture toughness were obtained for 9vol% and 12vol% additions of Ni and Mo, respectively. The hardness of the composites reduced gradually with increasing content of metals Ni and Mo.

Keywords: ceramic matrix nanocomposite; metal phase; microstructure; mechanical properties

1. Introduction

Ceramic materials have unique mechanical and chemical properties, such as high wear resistance and high hardness, especially at high temperature [1]. However, their low fracture toughness limits their potential applications. Ceramic metal composites, the focus of extended research, include ceramic matrix nanocomposites and ceramic-reinforced metal matrix composites [2–3]. Ceramic-metal nanocomposites that exhibit improved flexural strength and fracture toughness may be developed by combining the “nanocomposite effect” with the ductility of the metals. Co and Ni are commonly used binding phases in cermets. However, compared with WC–Co alloys, WC–Ni alloys demonstrate better corrosion resistance and oxidation resistance [4]. Alumina is the most common matrix, and various metals that have been incorporated into alumina include Ni [5–9] and Mo [10–11]. By controlling the partial pressure of oxygen during the sintering process, the Ni and Al_2O_3 matrix can exist in the form of Ni spinel (NiAl_2O_4) or Ni and Al_2O_3 phases. The fracture strength of Ni-reinforced alumina is lower than that of the spinel-reinforced material [5]. Furthermore, the particle location is a key microstructural pa-

rameter to improve the fracture strengths of Ni-reinforced alumina composite. Occlusion of Ni particles reduces the fracture strength of composites [9]. In high-temperature sintering, metal phases are distributed at grain boundaries or triple junctions in the form of liquid state, which is conducive to strengthen grain boundaries [12].

In recent years, an increasing number of studies have been conducted on multiphase toughening ceramics, such as Al_2O_3 /WC–10Co/ZrO₂/Ni [13], Al_2O_3 /ZrO₂/Ni [14], Al_2O_3 –TiC–Ni [15], Al_2O_3 –TiC_{0.5}N_{0.5}/Co–Ni [16], and TiB–SiC–Ni [17]. In general, the comprehensive mechanical properties of multiphase toughening ceramics are better than that of particle-toughened ceramic. A large number of weak interfaces form at the boundaries between ceramic and metal grains. The weak interface is also an important approach to strengthen ceramics. Crack deflection and particle pullout can absorb the fracture energy to delay the further extension of cracks. A certain solid solution in the grain boundary improves the bonding strength of grain interfaces and the fracture strength of multiphase Al_2O_3 -based ceramic [16]. Kwon reported that an increase in (Ti_{0.93}W_{0.07})C solid solution phase in the system improves fracture toughness (K_{IC} : 9.3–12.7 MPa·m^{1/2}) and maintains high hardness (HV:

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11.9–14.1 GPa) in the TiC–WC–(Ti,W)C–Ni cermet [18]. (W,Ti)C is a solid solution compound of WC with high fracture toughness and TiC with high hardness. The highest toughness values of Al_2O_3 –(W,Ti)C composites are 2.4-fold higher than that of pure alumina ceramic [19–20].

Even though the Al_2O_3 –Ni(Mo) and Al_2O_3 –(W,Ti)C composites have been extensively but separately studied for the past 20 years, research on Al_2O_3 –(W,Ti)C–Ni–Mo ceramic composite is rare. Adding Ni and Mo into Al_2O_3 –(W,Ti)C composite is expected to generate a material with high strength and toughness to improve the impact resistance of cutting tool materials. In this study, the influences of varying Ni and Mo contents on the microstructure and mechanical properties of Al_2O_3 –(W,Ti)C–Ni–Mo composites were discussed.

2. Experimental

2.1. Preparation of materials

The nominal compositions of the Al_2O_3 –(W,Ti)C–Ni–Mo ceramics with different Ni and Mo contents studied in this work are given in Table 1. Commercially available Al_2O_3 (0.5 μm), (W,Ti)C (the weight ratio of WC: TiC = 7:3) with an average particle size of 3 μm (purity 99.7%, Changsha Langfeng Metallic Material Co., Ltd., China), nano- Al_2O_3

(20 nm), Mo (0.6 μm), and Ni (0.6 μm) (purity 99.5%, Shanghai Chaowei Nanotechnology Co. Ltd.) were used as starting powders. The volume ratio of Ni:Mo was 2:1. MgO and NiO were used as sintering additives. The (W,Ti)C powder was ball-milled in ethanol medium with Al_2O_3 ceramic balls for 10 d to decrease the average particle size to about 1 μm . Nano- Al_2O_3 was placed in ethanol with the assistance of 0.5wt% (relative weight ratio to the nanoparticles being dispersed) dispersant (Shanghai Chaowei Nanotechnology Co. Ltd.). The dispersion process was assisted by mechanical stirring and ultrasonic vibration (with HS10260D ultrasonic instrument, China) for 30 min to attain the best dispersion effect. According to Table 1, a certain amount of microscale Al_2O_3 , (W,Ti)C, Ni and Mo was added into a uniform suspension of nano- Al_2O_3 and ultrasonically stirred for another 20 min. The mixed slurries were ball-milled using a planetary mill (XQM-2, China) at a speed of 70 r/min for 48 h. The weight ratio of balls-to-powder was 5:1. The milled slurry was dried in a vacuum drying oven (Model DZ-2AII, China) at 116°C. Subsequently, the dry mixed powder was placed in a graphite mould and then sintered using a vacuum hot-pressing sintering furnace (ZRC85-25T, China) at 1650°C and 30 MPa for a soaking time of 20 min.

Table 1. Composition of different nanocomposites

vol%

Composites	Al_2O_3 (500 nm)	Al_2O_3 (20 nm)	(W,Ti)C (1000 nm)	Mo (600 nm)	Ni (600 nm)
AWT	44	11	45	0	0
AWTN4	38	11	45	2	4
AWTN6	35	11	45	3	6
AWTN8	32	11	45	4	8

2.2. Specimen processing

The columned sinters with a size of $\phi 42 \text{ mm} \times 3.4 \text{ mm}$ were wire-cut and ground into rectangular bars with dimensions of $3 \text{ mm} \times 4 \text{ mm} \times 30 \text{ mm}$ by a MQ6025A-type universal tool grinder. Finally, the bars were ground with boron carbide powers and polished with diamond spray to a surface roughness R_a of about 1 μm . The flexural strength of nanocomposites was measured by an electromechanical universal testing machine (Model WDW-50E, China) with a 20 mm span at a loading velocity of 0.5 mm/min. Vickers hardness was also tested on the polished surfaces by a diamond pyramid indenter (Model MHVD-30AP, China) at an applied pressure of 196 N for an identical dwell time of 15 s. The fracture toughness was calculated using the length of the radial cracks of the Vickers indentations according to the formula proposed by Evans and Charles [21]. To minimize error, five specimens were tested in each experimental con-

dition. The phase compositions of the ceramic samples were analyzed by X-ray diffraction (XRD, RAX-10A-X, Hitachi). The fractured surfaces and crack propagation paths on the polished surfaces were observed by scanning electron microscopy (SEM, JSM-6510LV) equipped with energy dispersive spectroscopy (EDS).

3. Results and discussion

3.1. Effect of Ni and Mo contents on phase composition

XRD patterns from sintered micro-nanocomposite ceramics are presented in Fig. 1. The existing phases (Al_2O_3 and (W,Ti)C) were as expected for each process. Only Al_2O_3 and (W,Ti)C were detected for the AWT specimens without Ni and Mo. In Figs. 1(b)–1(d), the XRD patterns of AWTN4, AWTN6, and AWTN8 contained strong peaks that corresponded to the characteristic peaks of compound MoNi. In the sintering stage, the addition of Mo could improve the

wettability between metal Ni and ceramic phases, thereby enhancing the interface bonding strength [22]. According to previous research [18], although the dissolution rate of (W,Ti)C is lower than that of other carbides because of its thermodynamic stability, a small amount of (W,Ti)C can dissolve in Ni at high temperature, especially in the presence of heavy elements such as Mo. However, the spinel NiAl_2O_4

appearing in the Al_2O_3 –Ni system [23] was undetected.

3.2. Effect of Ni and Mo contents on microstructure

To further investigate the effects of a metal binder on the mechanical properties and microstructures of four ceramics with different Ni and Mo contents, the fracture surfaces after bending strength test were analyzed by SEM (Fig. 2). Both

Fig. 1. XRD patterns of the nanocomposite with different Ni and Mo contents: (a) 0vol%; (b) 6vol%; (c) 9vol%; (d) 12vol%.

Fig. 2. Fracture surfaces of AWT (a), AWTN4 (b), AWTN6 (c), and AWTN8 (d).

transgranular and intergranular fractures were observed, but more transgranular fractures were found in sample AWT. Large broken grains were characterized by cleavage fracture with many steps in samples AWTN4 and AWTN6, as shown in Figs. 2(b) and 2(c), and more fracture energy was consumed. In Fig. 2(d), a few dimples and fracture edges after necking were observed in sample AWTN8. EDS results of points A–E in Figs. 2(b) and 2(c) are shown in Table 2. The main phase of points A, B, and D was (W,Ti)C, whereas Al_2O_3 was the main phase of points C and E with a small amount of metals Ni and Mo. Mo and Ni diffused around (W,Ti)C and Al_2O_3 grains. (W,Ti)C, Ni and Mo could form a solid solution in the sintering process. Furthermore, the cohesion between metal and ceramic phases was enhanced. These results were consistent with the findings of previous reports [16,22]. Thus, high-strength metals played a significant role in improving the flexural strength of materials. Meanwhile, Al_2O_3 grains almost displayed their original size because of the “pinning effect” caused by the presence of metals Ni and Mo. Coarse tough (W,Ti)C and fine Al_2O_3 grains, as well as strong interfaces, were the source of high strength in Al_2O_3 –(W,Ti)C–Ni–Mo nanocomposites.

SEM micrographs of crack paths of AWTN4 and AWTN8 are shown in Figs. 3(a) and 3(b), respectively. When metal particles were incorporated into the ceramic matrix with nanoparticles, more metal inclusions in the Al_2O_3 matrix enhanced the toughness of the material at point B in Fig. 3(a). Both transgranular (framed by rectangle)

and intergranular fractures (framed by ellipse) were observed in Fig. 3(b). Crack bridging caused by the small black grain with nickel phase was observed in specimens AWTN4 and AWTN8 (Figs. 3(a) and 3(b)). Furthermore, the zigzag growth path of crack with a branching crack locally was also found. Fig. 3(c) shows the EDS spectrum of micro-nanocomposite ceramic for point A in Fig. 3(a). The main crystal phase of white grains was (W,Ti)C. Fig. 3(d) shows that the main crystal phase of small black grains (point B in Fig. 3(a)) was Al_2O_3 . Given the relatively low Ni and Mo contents for points A and B, it could be inferred that Mo and Ni were distributed at grain boundaries or triple junctions. Moreover, the growth of the particles was inhibited. Compared with covalent bond in the ceramic phase,

Table 2. EDS results of points A–E in Figs. 2(b) and 2(c) at%

Element	A	B	C	D	E
C K	9.26	8.07	0.00	7.26	0.66
O K	27.97	22.07	43.07	26.36	59.84
Al K	3.97	2.37	32.47	2.52	24.50
Ti K	29.81	44.02	15.93	44.85	3.84
Ni K	3.99	1.88	3.93	1.36	9.07
Mo L	10.04	4.56	1.46	0.00	0.44
W M	14.96	17.03	3.14	17.65	1.02
Mg K	0.00	0.00	0.00	0.00	0.63

Fig. 3. SEM micrographs of crack extension paths of AWTN4 (a) and AWTN8 (b), respectively; EDS spectra of the composite ceramic for points A (c) and B (d) in Fig. 3(a), respectively.

ceramic/metal interfaces demonstrated weak links, and dislocations could be introduced. Therefore, plastic deformation of metal particles could absorb more energy and prevent the further expansion of cracks. Plastic deformation of metal particles and crack bridging were the main toughening mechanisms.

3.3. Effect of Ni and Mo contents on mechanical properties

The effect of Ni and Mo contents on fracture toughness is shown in Fig. 4. The fracture toughness increased greatly with the increase in Ni and Mo contents. The maximum fracture toughness ($10.60 \text{ MPa}\cdot\text{m}^{1/2}$) was observed for 12vol% addition of Ni and Mo. The factors influencing fracture toughness included the interface bonding strength, the state of stress at the $\text{Al}_2\text{O}_3/\text{Ni}(\text{Mo})$, $(\text{W,Ti})\text{C}/\text{Ni}(\text{Mo})$, and $\text{Al}_2\text{O}_3/(\text{W,Ti})\text{C}$ interfaces, the size of $\text{Ni}(\text{Mo})$ grains, and the way by which a crack grows through metal grains. These factors ultimately affected the size of a frontal process zone (FPZ) ahead of a crack tip. The FPZ is thought to be composed of nanocracks in ceramic and dislocations in metals [24]. According to Ref. [25], if the metal content increases, dislocations will appear when cracks grow in the micro-nanocomposite, and more energy is needed for cracks to extend. Consequently, the fracture toughness of the micro-nanocomposite with metal phases is enhanced. The fracture toughness for AWTN4 and AWTN6 was similar. Possible reasons are as follows. The fracture surfaces exhibited the same features, as shown in Figs. 2(b) and 2(c). The fracture surfaces of large grains were cleavage planes with many steps, and few fracture edges after necking appeared because of low levels of Ni and Mo contents. The fracture surfaces did not present ductile fracture features. Therefore, metal phases made nearly the same contribution to improving the fracture toughness of AWTN4 and AWTN6.

Fig. 4. Effect of Ni and Mo contents on fracture toughness.

The effect of Ni and Mo contents on flexural strength is shown in Fig. 5. Flexural strength is closely related to composition, microstructure, binder phase, and porosity. The flexural strength of $\text{Al}_2\text{O}_3-(\text{W,Ti})\text{C}-\text{Ni}-\text{Mo}$ ceramics initially increased and then decreased with increasing Ni and Mo contents. The maximum flexural strength was observed for 9vol% addition of Ni and Mo. When the contents of Ni and Mo were less than 9vol%, the size and distribution of metal grains were beneficial to the increase in strength. The strength of the nanocomposites was dominated by the biggest microdefects, which were created on grain boundaries or inside the ceramic grains due to synergetic effects of the residual stresses and processing defects [25]. Therefore, the microdefect size dominating the strength of materials was considered to be the largest cavity in nanocomposites. When the additions of Ni and Mo were from 0vol% to 9vol%, the liquid phase accelerated the re-arrangement of the grains and enhanced the densification by filling the clearance between solid grains. An appropriate amount of metal between the ceramic grains in AWTN6 could reduce the size and number of the microdefects. The size of the microdefects in the matrix was expected to be smaller than the grain size. When the Ni and Mo contents were 12vol%, more pores occurred and grew due to the loss of Ni under high temperature and pressure (Fig. 2(d)). The EDS results in Table 2 also reflect the loss of Ni and Mo at some points with the increase in Ni and Mo contents. The decrease in flexural strength of AWTN8 was attributed to the increase in size of the biggest microdefect.

Fig. 5. Effect of Ni and Mo contents on flexural strength.

The effect of Ni and Mo contents on the hardness of $\text{Al}_2\text{O}_3-(\text{W,Ti})\text{C}-\text{Ni}-\text{Mo}$ nanocomposites is shown in Fig. 6. As expected, the hardness of the composites reduced gradually with increasing the content of soft metal Ni according to the law of mixtures.

Fig. 6. Effect of Ni and Mo contents on hardness.

4. Conclusions

In this study, Al_2O_3 -based nanocomposites were prepared by vacuum sintering with varying contents of Al_2O_3 , (W,Ti)C, Ni, and Mo as raw materials. The influence of Ni and Mo additions on the microstructure and mechanical properties of Al_2O_3 -(W,Ti)C-Ni-Mo nanocomposites was investigated. The following conclusions were drawn from the study.

(1) The main phases Al_2O_3 and (W,Ti)C were detected by XRD. Compound MoNi also existed in sintered nanocomposites. The adhesion between Ni/Mo and (W,Ti)C was improved by forming a solid solution.

(2) The fracture modes of nanocomposites were both intergranular and transgranular fractures. The fracture surfaces of nanocomposites with metal phases were cleavage planes with steps. The “pinning effect” caused by metals Ni and Mo, and the strong bond between metal phases and hard phases, as well as fine Al_2O_3 grains, improved the flexural strength of the nanocomposites. The plastic deformation of metal particles and crack bridging were the main toughening mechanisms.

(3) The maximum flexural strength and fracture toughness were obtained for 9vol% and 12vol% additions of Ni and Mo, respectively. The hardness of the composites reduced gradually with increasing Ni and Mo contents. Overall, the strength and toughness of the nanocomposites with Ni and Mo additions were both higher than those of the nanocomposites without Ni and Mo additions, but hardness slightly decreased.

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