



## Influence of deformation passes on interface of SiC<sub>p</sub>/Al composites consolidated by equal channel angular pressing and torsion

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**Abstract:** Powder mixture of pure Al and oxidized SiC was consolidated into 10% (mass fraction) SiC<sub>p</sub>/Al composites at 250 °C by equal channel angular pressing and torsion (ECAP-T). The valence states of Si for SiC particulates and Al for the as-consolidated composites were detected by X-ray photoelectron spectroscopy (XPS). The interfacial bondings of the composites were characterized by scanning electron microscopy (SEM). The elements at the interface were linearly scanned by energy dispersive spectroscopy (EDS) and the EDS mappings of Si and Al were also obtained. The values of the nanohardness at different positions within 2 μm from the boundary of SiC particulate were measured. The results show that after ECAP-T, interfacial reaction which inhibits injurious interfacial phase occurs between Al and the oxide layer of SiC, and the element interdiffusion which can enhance interfacial bonding exists between Al and SiC. As ECAP-T passes increase, the reaction degree is intensified and the element interdiffusion layer is thickened, leading to the more smooth transition of the hardness from SiC to Al.

**Key words:** metal matrix composites; consolidation; deformation; interfacial reaction; element interdiffusion

### 1 Introduction

Particle reinforced metal matrix composite (PRMMC) has generated wide spread interest due to a series of excellent properties such as light mass, high stiffness, high strength and fine abrasion performance which make it a kind of attractive materials for aerospace, machinery and electronics industry [1–3]. This composite fabricated by traditional methods, like powder metallurgy (PM) and squeeze casting [4,5], always requires secondary processing in order to homogenize the particles distribution among the matrix and minimize the amount of pores [6–9]. As a result, severe plastic deformation (SPD) was proposed for directly fabricating qualified composite. Equal channel angular pressing (ECAP), one of the most commonly used methods in SPD, was applied to achieving full density of 1% (volume fraction) carbon nanotube-Cu matrix composites with superior mechanical properties by improved particle bonding and least grain growth [10]. Forward extrusion and equal channel angular pressing (FE-ECAP) was engaged as a technique for consolidating Al matrix

composites with 5% and 10% SiC particulates, which were proved to be better compacted than the composites with the same constituents consolidated by hot forward extrusion [11]. A fully dense Al–Al<sub>2</sub>O<sub>3</sub> composite consisting of mostly nanocrystalline Al and γ-Al<sub>2</sub>O<sub>3</sub>, a small fraction of ultrafine Al grains and amorphous alumina was well consolidated after four passes back pressure equal channel angular pressing (BP-ECAP) and it was found that the formation of the nanostructure was attributed to the interaction between severe shear deformation and in situ oxidation during ECAP [12].

Equal channel pressing and torsion (ECAP-T) is a novel SPD method which improves ECAP processing by manufacturing a spiral channel in the horizontal stage of ECAP, as shown in Fig. 1. This process was firstly applied for consolidating pure Al powder and proved to be a feasible approach for fabricating full dense bulk material with fine grains [13]. Then, the mixtures of SiC particles and Al powders were consolidated into PRMMCs by ECAP-T and ECAP, respectively [14]. It is found that SiC particles distribution in Al matrix was more homogeneous after ECAP-T process, compared with simple ECAP process. The aim of this study is to

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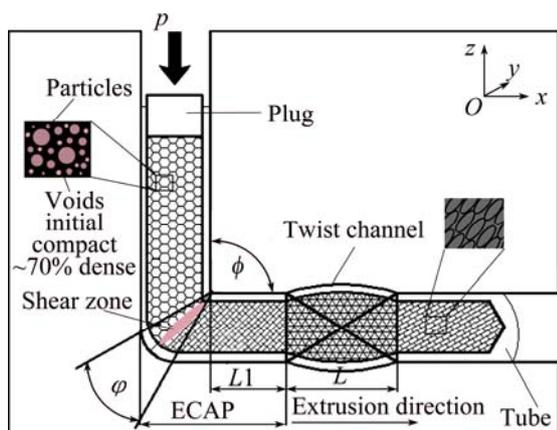


Fig. 1 Schematic illustration of ECAP-T process

investigate the influence of ECAP-T passes at the interface of SiC/Al composites consolidated by this process.

## 2 Experimental

High purity (99.81%) Al powder having an average particulate size of 32.22  $\mu\text{m}$  was chosen for raw matrix material. The main impurities in Al were 0.11% Fe and 0.07% Si. The reinforcement used in the study was SiC having an average particulate size of 13.59  $\mu\text{m}$ . SiC powders were pre-heated in a furnace at 250  $^{\circ}\text{C}$  for 2 h to remove the moisture and then oxidized at 750  $^{\circ}\text{C}$  for 2 h to form  $\text{SiO}_2$  layer on the particle surface. The total mass of reinforcement particulates for one composite sample was measured as 0.172 g ( $m_0$ ) before oxidation and 0.188 g ( $m$ ) after oxidation. According to the relative molecular mass of SiC ( $A_1$ ) and  $\text{SiO}_2$  ( $A_2$ ), the oxidation proportion ( $P$ ) of SiC particulates is calculated as 9.3% by the formula as below:

$$P = \frac{A_1(m - m_0)}{(A_2 - A_1)m_0} \times 100\% \quad (1)$$

The matrix Al powder was homogeneously blended with the oxidized SiC powder in the mass ratio of 9:1. The morphology of the mixtures of Al powder and SiC particles is shown in Fig. 2. As can be seen, the blended powders had a random morphology. Al and SiC particles were like near-ball and polygon, respectively.

The powder mixtures were wrapped in copper tubes with a relative density of 0.7 and then inserted into ECAP-T die. The ECAP-T process was carried out at 250  $^{\circ}\text{C}$  under inert gas protection with a ram speed of 1 mm/s in single pass, 2 passes and 4 passes, following route B<sub>C</sub> (the sample is rotated by 90 $^{\circ}$  in the same direction between consecutive passes). During the pressing, the mixed lubricant of graphite and  $\text{MoS}_2$  was engaged in order to reduce the friction. The die

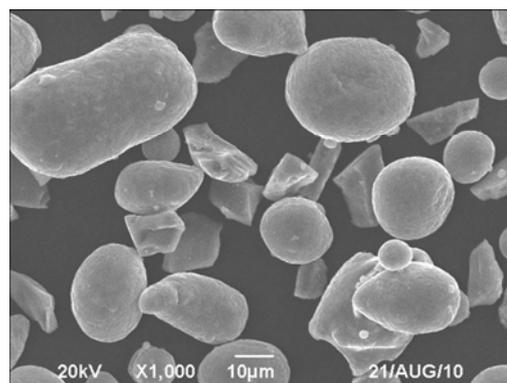


Fig. 2 SEM image of powder mixture

parameters were as follows: the channel angle  $\phi = 90^{\circ}$ , the arc of outer corner curvature  $\psi = 37^{\circ}$ , the twist angle = 90 $^{\circ}$ , the cross-sectional dimensions of the channel 10 mm $\times$ 10 mm, the length of twist channel  $L = 30$  mm and the length of transition channel  $L_1 = 15$  mm.

The as-consolidated composites were examined by XPS (Thermo Fisher Scientific ESCALAB 250Xi XPS System) in order to detect the valence of elements. Then, the microstructures of the composites were observed by SEM (JEOL JSM-6490LV) and the element distribution between the matrix and reinforcement was investigated by SEM-EDS. The nanohardness values at the interface of the composites were measured by nanohardness tester (HVS-1000).

## 3 Results and discussion

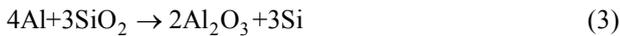
### 3.1 Interfacial reaction at SiC/Al interface

The temperature at which SiC<sub>p</sub>/Al composites were fabricated via ordinary PM method is always around the melting point of Al. During the PM process it was difficult to inhibit the chemical reaction (2) at the interface between Al and SiC.

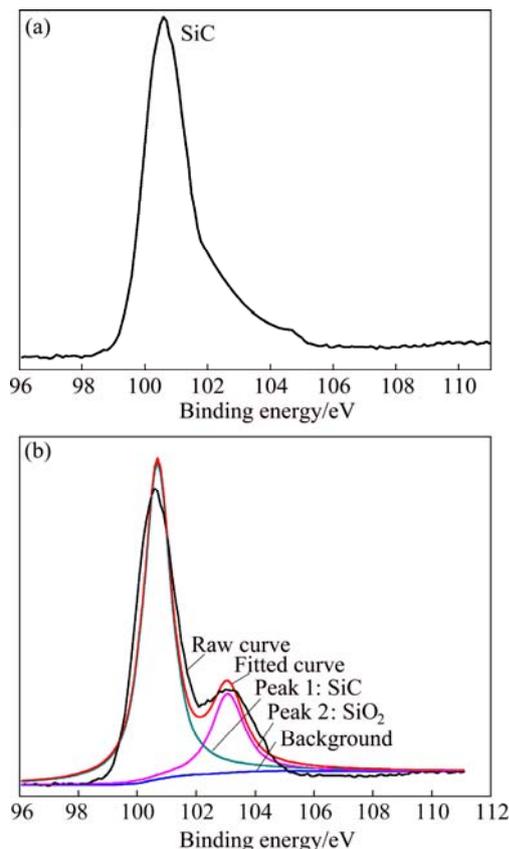


The interfacial phase  $\text{Al}_4\text{C}_3$  forming in this reaction is a kind of brittle compound which can weaken the bonding strength of SiC/Al interface. In this study, the ECAP-T processing temperature is much lower than the melting point of Al. However, the severe hydrostatic pressure during the processing may support the driving power of the harmful interaction which is equal to the thermal power during PM processing. Consequently, there are still possibilities for the generation of  $\text{Al}_4\text{C}_3$  if SiC particles have no pre-treatment. The  $\text{SiO}_2$  layer on SiC particle surface forming in the oxidation treatment can play a great role in avoiding the phenomenon above. The existence of  $\text{SiO}_2$  layer leads to the occurrence of another protective reaction (3) which can successfully

replace the harmful interaction discussed above.



Accordingly, the oxidation treatment for the reinforcement is necessary. Figure 3(a) shows the XPS survey spectrum of non-oxidized SiC particulates in the Si 2p region. It is observed that the surface of non-oxidized particulates exhibits only one significant level of Si 2p core level. The intensity peak is located at 100.6 eV, which corresponds to SiC [15]. Instead, two core levels of Si 2p are observed on the surface of oxidized particulates as shown in Fig. 3(b). According to the deconvolution of the spectra, we fixed the binding energy (BE) at 100.6 eV and 103.2 eV for SiC and SiO<sub>2</sub> [16] core levels, respectively. The volume ratio of SiC to SiO<sub>2</sub> can be estimated by calculating the area ratio of the two intensity peaks as about 0.11:1. This means that the oxidization ratio of SiC particulates is about 9.7% which comparatively accords with the value of 9.3% measured by weighting method. Therefore, the accuracy of quantitative analysis by XPS is proved to be convincing.



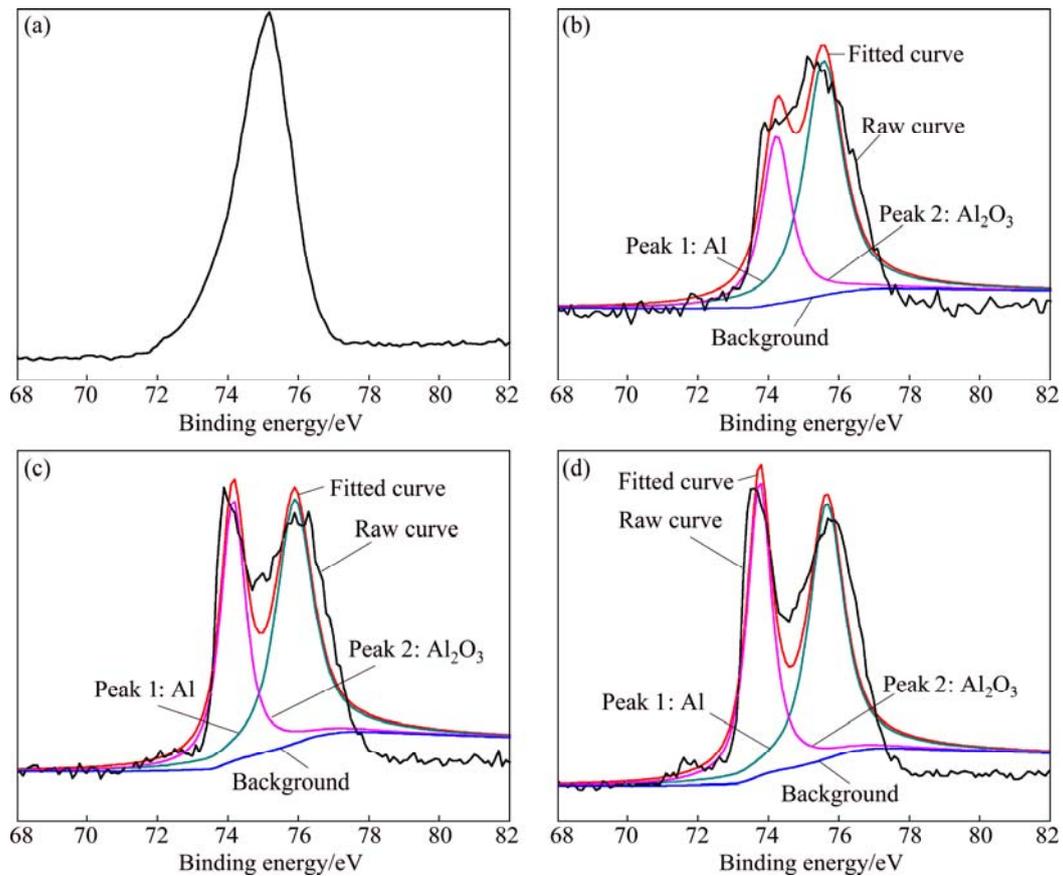
**Fig. 3** XPS survey spectra in Si 2p region for non-oxidized (a) and oxidized (b) SiC particulates

Figure 4 shows the effect of ECAP-T processing passes on the XPS spectra of Al 2p core level for the SiC<sub>p</sub>/Al composite sample. Before ECAP-T processing (Fig. 4(a)), there is only one peak of great intensity localized at 75.6 eV in the Al 2p region, which is

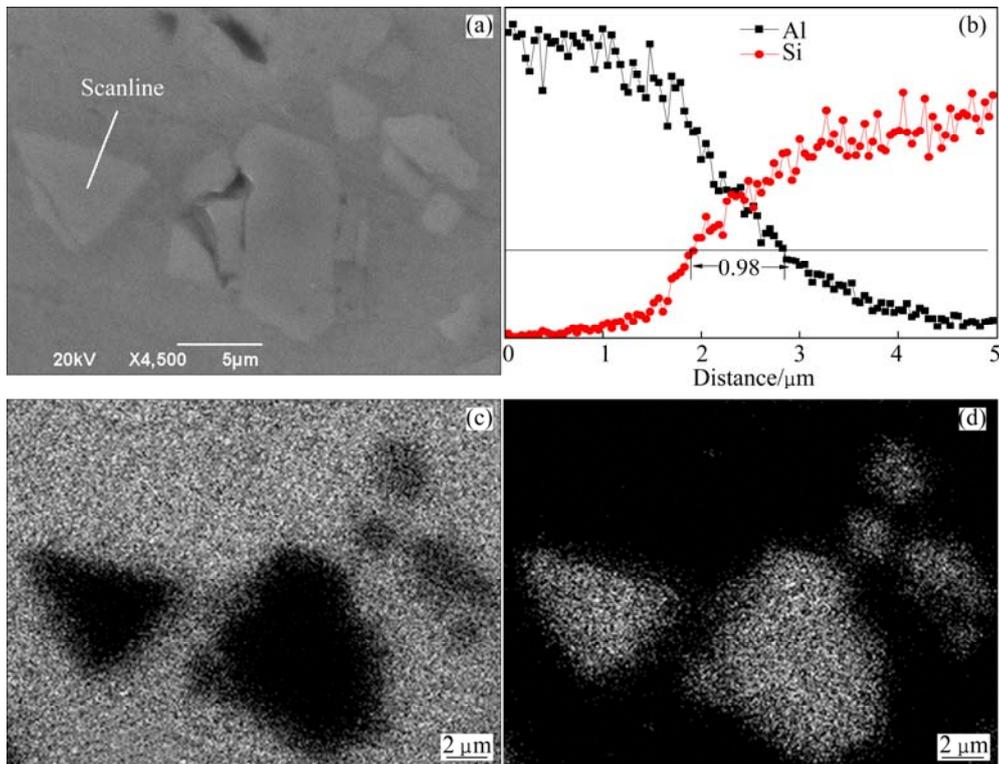
associated to aluminum simple substance [17]. This means that raw Al powder was not contaminated under inert gas protection before the consolidation. However, Figs. 4(b)–(d) show that the composite samples ECAP-T processed exhibit one more intensity peaks localized at 74.2 eV in the Al 2p region, which is associated to Al<sub>2</sub>O<sub>3</sub> [18]. This is a kind of chemical shift about Al element in BE, which verifies the occurrence of the protective reaction with the oxidation layer of SiC particulates. Meanwhile, no intensity peak appears at 73.6 eV, which means that the harmful phase Al<sub>4</sub>C<sub>3</sub> was inhibited to generate [19]. We name the peak localized at 75.6 eV as Peak 1 and the peak localized at 74.2 eV as Peak 2. After single pass ECAP-T processing, the area ratio of Peak 1 to Peak 2 is calculated as 0.55:1 from the fitted curve after deconvolution. When the number of process passes increases to 2 (Fig. 4(c)), the area ratio ascends to 0.75:1, which means that the amount of reaction product Al<sub>2</sub>O<sub>3</sub> rises up. This directly illustrates that the increase of ECAP-T process passes deepens the degree of the protective reaction. As the number of processing passes is raised to 4, the area ratio still increases to 0.82:1. Therefore, it is seen that the degree of protective action is positively correlated with the number of ECAP-T process passes.

### 3.2 Interdiffusion of elements at interface

Figure 5 shows the distribution of the elements (Al and Si) on the polished surface of the composite sample processed by single pass ECAP-T. The EDS scanline is perpendicular to the straight boundary of polygonal SiC particles as shown in Fig. 5(a). The scanline is 5 μm long and divided into two halves by the reinforcement boundary. Figure 5(b) shows the EDS spectra of Al and Si along the scanline. It can be observed that the intensities of Al and Si gradually vary from the matrix to the reinforcement with opposite trends. The intensity transition reveals that there is a diffusion zone around the boundary of SiC particle where the interdiffusion of the elements between the matrix and reinforcement exists. The effective diffusion zone is defined as the area where the intensities of Al and Si element are both above 100 cps. Based on this definition, the thickness of the diffusion zone in Fig. 5(b) is measured as about 0.98 μm. The EDS mapping of Al and Si for the SEM image can be observed distinctly in Figs. 5(c) and (d). According to the mapping, the regional distribution of the elements is visually presented. As can be seen, Si disperses from particle boundary into the matrix and Al also diffuses into SiC particles. Therefore, the interface bonding of SiC<sub>p</sub>/Al composite should not be simply attributed to the chemical reaction. The interdiffusion of the elements between the reinforcement and matrix should also be taken into account.

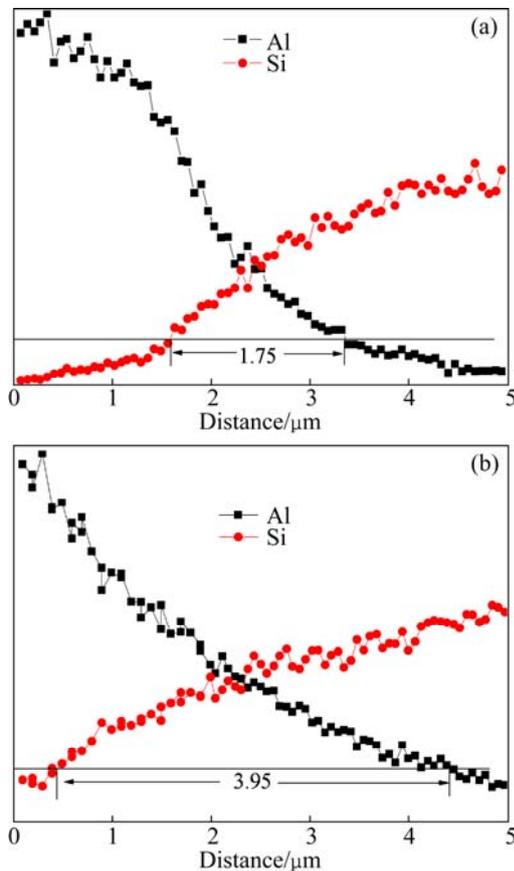


**Fig. 4** XPS spectra of Al 2p core level for powder mixture before ECAP-T (a) and after single pass (b), 2 passes (c) and 4 passes (d) ECAP-T process



**Fig. 5** Distribution of elements on polished surface of composite sample processed by single pass ECAP-T: (a) SEM image of composite; (b) EDS spectra of Al and Si along scanline; (c) EDS mapping of Al; (d) EDS mapping of Si

In order to investigate the influence of the deformation created by ECAP-T on the interdiffusion of the elements, the composite samples fabricated by different passes ECAP-T process were also analyzed by SEM-EDS. From Figs. 6(a) and (b), it can be observed that the thicknesses of the diffusion zones belonging to the sample fabricated by two and four passes are 1.75 and 3.95  $\mu\text{m}$ , respectively. Compared with the sample processed by only one pass deformation, we find that increasing the number of ECAP-T pass has a positive effect on thickening the diffusion layer of the composite. The diffusion in solids is a process relying on heat activation of atoms. Atoms in the crystals are always vibrating around their equilibrium positions. With the increase of temperature, the vibration of atoms becomes more intense, which makes atoms more easily activated for the migration behavior. Atoms must have a sufficiently high energy to migrate from its original equilibrium position to a new equilibrium position. It is known that the deformation energy supported by ECAP-T process can partially convert into thermal energy. At a given surrounding temperature, the thermal energy of the atoms in the crystals tends to increase with the growth of the deformation degree and the atoms migrate further consequently. When Al atoms migrate further in the reinforcement and Si atoms migrate further

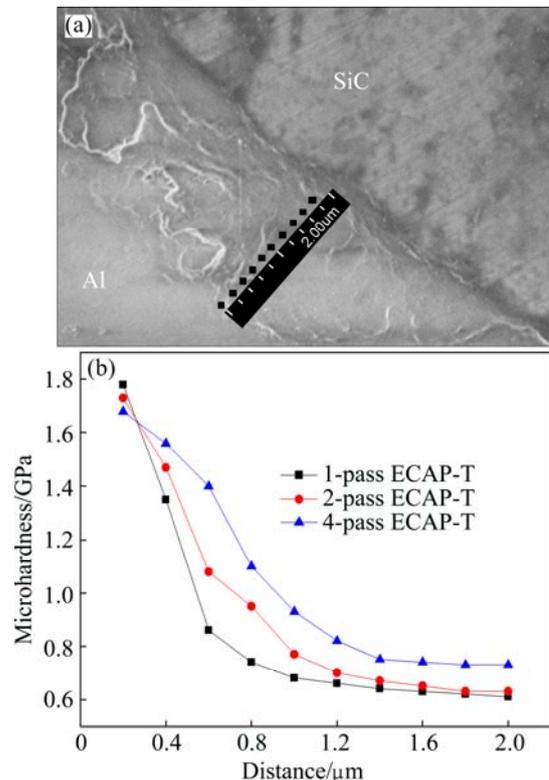


**Fig. 6** EDS spectra of Al and Si for samples fabricated by 2 (a) and 4 (b) passes ECAP-T

in the matrix, the interdiffusion layer of the composite tends to thicken. It is known that the deepening of the diffusion degree is beneficial to the enhancement of the interfacial bonding between the matrix and the reinforcement. Therefore, increasing the pass number of ECAP-T can be a kind of method for promoting the interfacial bonding.

### 3.3 Nanohardness at interface

Figure 7 shows the distribution of the nanohardness at different positions within the range of 2  $\mu\text{m}$  from the boundary of SiC particulate. The nanohardness values of the samples after different passes ECAP-T were measured at the points every 200 nm from 0.2 to 2.0  $\mu\text{m}$ . The values tend to decline with the increase of the distance and be stable at a certain distance. The declining rate of the hardness against the distance differs among the samples and decreases with the deformation passes which the sample experiences. The sample fabricated by four-pass ECAP-T shows the slowest declining rate of hardness and the declination ends at the point 1.4  $\mu\text{m}$  from SiC boundary. Instead, the hardness of the sample experiencing single pass deformation declines most rapidly and keeps stable at the point of 0.9  $\mu\text{m}$  from SiC boundary. In addition, it is observed that the maximum of the samples decreases with the growing process pass. However, the minimum of the sample processed by 4 passes is above the minimum of the other two samples. The distribution law of the hardness is closely related



**Fig. 7** Nanohardness at different positions: (a) Different positions at interface; (b) Variation of hardness

with the interface of the composite sample. The interdiffusion layer supports the gradual transition of the hardness from the reinforcement to the matrix. As discussed above, the interdiffusion layer can be thickened by increasing ECAP-T passes. Therefore, the distance of the hardness declination becomes longer due to the thicker diffusion layer. Meanwhile, due to the more severe deformation, more Si atoms are detected in the Al grains far from SiC boundary. In contrast, more Al atoms originally distributed outside SiC boundary diffuse into SiC particulate. As a result, local softening occurs at the position where the hardness reaches the maximum and becomes more intense with the growing ECAP-T passes, as well as local hardening at the position where the hardness reaches the minimum.

## 4 Conclusions

1) Al-based composites (10% SiC particulates) were fabricated via ECAP-T process. The oxidization layer of SiC inhibited the generation of the harmful interfacial phase. The interface bonding of the composites has the characteristics of both interfacial reaction and element interdiffusion.

2) With the growth of ECAP-T passes, the degree of the interfacial reaction and element interdiffusion were intensified and the hardness transition from the reinforcement to the matrix became smoother. Local hardening in the matrix and softening in the reinforcement became more obvious when ECAP-T passes were added.

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## 变形道次对基于等径角挤扭法制备的 SiC<sub>p</sub>/Al 复合材料界面的影响

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**摘 要:** 在 250 °C 下, 利用等径角挤扭变形工艺(ECAP-T)将纯 Al 和经氧化处理的 SiC 混合粉末固结成 10% SiC<sub>p</sub>/Al 复合材料。采用 X 射线光电子能谱仪(XPS)测定 SiC 颗粒氧化处理前后 Si 元素的价态以及固结后复合材料中 Al 元素的价态, 通过扫描电镜(SEM)对复合材料的界面进行观察, 并利用能谱仪(EDS)对界面结合处进行元素线扫描和面扫描, 最后对复合材料界面进行纳米硬度的测量。结果表明, ECAP-T 变形后, Al 和 SiC 的氧化层之间发生了界面反应, 抑制了有害界面相的产生, 并产生了基体和增强体之间的元素互扩散; 随着 ECAP-T 变形道次的增加, 界面反应程度加剧, 元素扩散层加厚, 使复合材料从增强体到基体的硬度过渡更加平缓。

**关键词:** 金属基复合材料; 固结; 变形; 界面反应; 元素互扩散

(Edited by Xiang-qun LI)