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Short communication

Improvement of interface between Al and short carbon fibers by α-Al_2O_3 coatings deposited by sol–gel technology

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Abstract

This paper describes an investigation on an improvement of the interface between Al and short carbon fibers (SCFs) with α-Al_2O_3 coating by sol–gel technology. The composites of Al/uncoated SCFs and Al/α-Al_2O_3-**coated SCFs were fabricated successfully by vacuum press infiltration. The formation of α-Al_2O_3 coating during calcination was analysed by Fourier transform infrared (FTIR) and X-ray diffraction (XRD). Scanning electron microscopy (SEM), energy-dispersive analysis of X-ray (EDAX) and transmission electron microscope (TEM) were used to observe the coated SCFs and the interface of composites. The results showed that the average thickness of the α-Al_2O_3 coating was about 200–250 nm and the formation of Al_4C_3 at the interface between Al matrix and SCFs was controlled by the α-Al_2O_3 coatings.

Keywords: B. Interface; Aluminum; Short carbon fibers; α-Al_2O_3 coating; Sol–gel

1. Introduction

Carbon fibers (CFs) reinforced metal matrix composites are widely used and studied due to the high specific strength, specific modulus, high thermal and electric conductivity, low expansion coefficient and good self-lubricant [1–3]. Al/CFs composites are promising materials for aerospace and commercial application [4–6]. However, the poor wettability and harmful interfacial reaction between CF and aluminum make the preparation very difficult to obtain Al matrix composite with good interface bonding. Coating technology on fiber surface was the most efficient method to overcome these problems. Electroless plating and electroplating were the simple and common technics to obtain metallic coatings such as nickel and copper coatings, which can improve the wettability evidently. Nevertheless the coatings dissolved in the matrix alloy during the synthesis to give precipitation of brittle intermetallics e.g. $NiAl_3$, $CuAl_2$ in the interface, which result in the poor interface bonding and the decrease of mechanical properties largely [7–9]. Some researchers studied the processes of vapour deposition such as PVD, CVD to obtain the coatings such as TiC, B_4C, pyrolytic carbon, etc., and the effects of these coatings in aluminum matrix are evident [10–12], however, these processes are too expensive and complex to apply in the industry.

Compared with other methods, sol–gel coating process is simple and inexpensive method, and it can be coated on various substrates. Generally, in a sol–gel coating process, isopropoxide was used and hydrolysis proceeded, then the metallic oxide coatings are formed [13–15]. It reported that the coatings of Al_2O_3 or SiC on CFs obtained from isopropoxide solution can protect CFs from high temperature oxidation and chemical interaction with Al matrix [16,17]. In this paper, we developed a simple sol–gel process by using aluminum nitrate and ammonia to prepare the α-Al_2O_3 coatings on the surface of SCFs. The sol–gel process, the phase change of Al_2O_3 and the characterization of interface between SCFs and Al matrix were discussed.

2. Experimental

2.1. Preparation of precursor

SCFs used in this paper, were T300 made by Japan Toray Co. Ltd. The polyacrylonitrile (PAN)-based CFs were desized after being dispersed, and were cut into 2–3 mm length. The

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PAN-based fibers have a density of 1.76 g/cm^3 and a mean diameter of 7 μm.

Aluminum nitrate (A.R.) and ammonia solution (A.R.) were used as the starting materials. The transparent alumina sol was prepared as follows: aluminum nitrate (1 mol/L) was first dissolved in distilled water, and then it dripped into ammonia solution (1 mol/L) at a speed of 100 drips per minute with a rapidly stir at 95 °C. After the precipitation finished, nitric acid (0.9 ml) was added to obtain the clear alumina sol. Four percent polyvinyl alcohol (PVA) solution was dripped slowly into the alumina sol. The ratio of the PVA solution and the alumina sol solution was 1:4 in volume. The mixture was then heated for 20 h at 95 °C under a vigorous stir in order to obtain the homogeneous viscous sol.

2.2. Coating procedure and preparation of Al/SCFs composites

After an ultrasonic dispersion of SCFs in the alumina sol the SCFs were extracted and dried at 80–100 °C, and then calcined at about 1050 °C to obtain α-Al_2O_3 coatings under hydrogen atmosphere. 6061Al alloy was used as the matrix in this study, the Al/SCFs and Al/Al_2O_3 coated SCFs composites were fabricated by vacuum pressure infiltration technique. The temperature of perform was 500 °C, and the melt was 730 °C. The pressure was 7 MPa.

2.3. Characterization

The crystalline phase of the gels after calcination in nitrogen atmosphere was evaluated using a Bruker-axs X-ray diffraction (XRD). An investigation of the gel after calcination was performed via Fourier transform infrared (FTIR) spectroscopy (Bruker, Equinox). Scanning electron microscopy (SEM), field emission scanning electron microscopy (FESEM, Fei Sirion 200) and energy-dispersive analysis of X-ray (EDAX) were used to investigate the coated SCFs. The composites were observed by transmission electron microscope (TEM) of JEM2010 microscope.

3. Results and discussion

Fig. 1 shows XRD patterns of the as-synthesized precursor calcined at different temperatures for 1 h. It is shown that the precursor to be amorphous as well as the powder calcined at 500 °C. The characteristic peaks of γ-Al_2O_3 appear at 700 °C with a rather weak intensity, and the amorphous disappear at 800 °C, which means the amorphous to γ-Al_2O_3 transition is completed at 800 °C. With the increase of calcination temperature up to 900 °C, the crystallinity of γ-Al_2O_3 improve and α-Al_2O_3 appear with weak peaks, which indicate the transition of γ-Al_2O_3 to α-Al_2O_3. At 1000 and 1100 °C there are only the α-Al_2O_3 peaks, indicating the γ-Al_2O_3 to α-Al_2O_3 phase transition be almost completed when the temperature higher than 1000 °C.

The FTIR at 4000–400 cm^{-1} for the precursor calcined at different temperatures are shown in Fig. 2. The broadband at

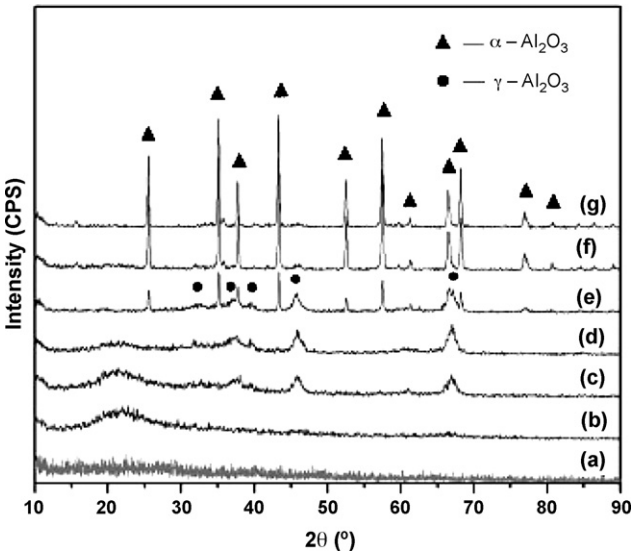

Fig. 1. XRD patterns of the as-synthesized precursor calcined at different temperatures (a) gel, (b) 500 °C, (c) 700 °C, (d) 800 °C, (e) 900 °C, (f) 1000 °C, (g) 1100 °C.

3445 and 1638 cm^{-1} is attributed to the stretching and bonding vibrations of the absorbed and inner water. The curves (a) and (b) are very similar, which means the alumina sol has the same contents of the alumina gel. In the two curves, the characteristic bands at 1053, 810 and 470 cm^{-1} indicate the stable boehmite formed during the preparation of precursor. Peaks localized at 1356 and 1041 cm^{-1} are the vibration of NO^{3-} group, which were disappeared when calcined at 700 °C. As for the curve (c), weak characteristic bands indicate that the water, NH^{4+}, NO^{3-} and other impurities were decomposed and volatilized completely. As the calcined temperature at 1000 °C, significant spectroscopic bands at 640, 588 and 449 cm^{-1} appear which are identified to be the characteristic absorption bands of α-Al_2O_3 [18]. And the weak absorption bands of α-Al_2O_3 can be seen form curve (d), indicating the phase transition of α-Al_2O_3

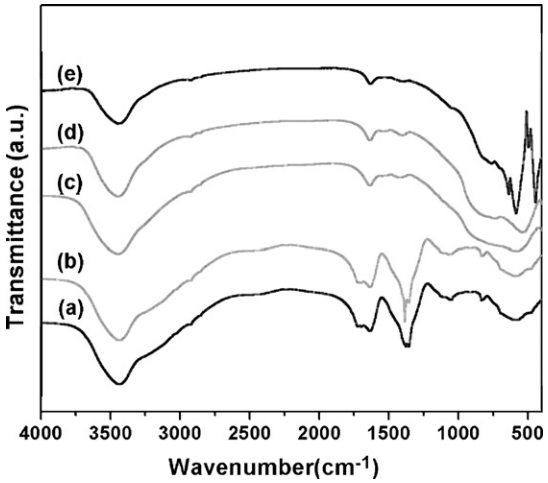

Fig. 2. FTIR spectra of as-synthesized precursor calcined at different temperatures (a) sol, (b) gel, (c) 700 °C, (d) 900 °C, (e)1000 °C.

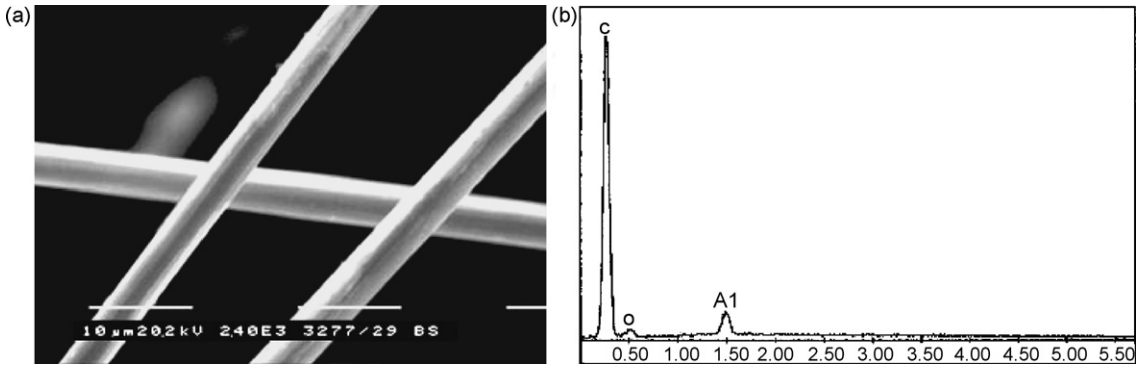

Fig. 3. SEM micrograph of (a) α-Al_2O_3-coated SCFs and (b) the EDAX analysis of the SCFs surface.

started at 900 °C, which is in accordance with the results of XRD analysis.

Fig. 3(a) shows the SEM image of α-Al_2O_3 coated SCFs. It can be seen that the diameters of the fibers are uniform. The Al and O elements peaks, as Fig. 3(b) shown, prove that the α-Al_2O_3 coatings exist in the surface of SCFs. Fig. 4 is the FESEM image of α-Al_2O_3 coating on the SCF surface, which shows the coating bond with the SCF well and the thickness is about 200–250 nm.

TEM observation (see Fig. 5) reveals that there are chemical reactions of the interface between aluminum matrix and uncoated SCF, the presence of needle phase and the SAD analysis indicate that the Al_4C_3 formed in the SCF surface. The brittle and fiber-like phase of Al_4C_3 will result in the weakening interfaces, destroy the fibers and drastically reduces the mechanical properties. Compared with the uncoated composite, the Al/α-Al_2O_3-coated SCFs composite presents neater interface. The SCF is coated α-Al_2O_3 uniformly and the thickness is about 200 nm, as Fig. 6 shown. Fig. 7 is the high magnification of the interface zone, it can be seen clearly that SCF bond with α-Al_2O_3 coating quite well and no harmful phases formed on the fiber surface. The analysis of the SAD pattern indicates the coating is α-Al_2O_3, which is agreed with the XRD results. The α-Al_2O_3 has good chemical stable and heat resistance, so it can play as a barrier layer in the Al/α-Al_2O_3-coated SCFs composite. Though no other compound can be formed in the Al/α-Al_2O_3 interface, Al and α-Al_2O_3 form quite a strong interface, and it is help to obtain higher mechanical properties [19,20]. Generally, the α-Al_2O_3 coating improved the interface between aluminum matrix and SCFs greatly.

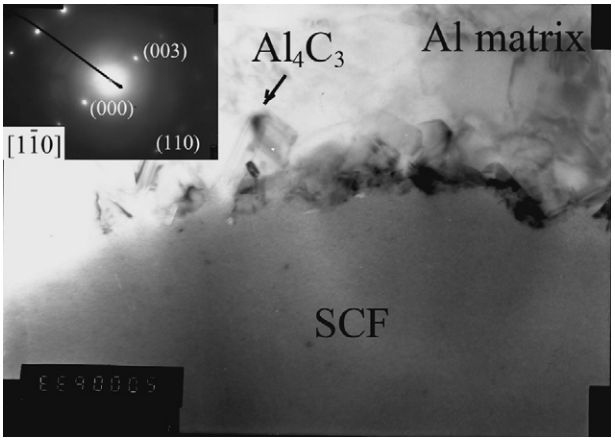

Fig. 5. TEM image and SAD of the interface between aluminum matrix and uncoated SCF.

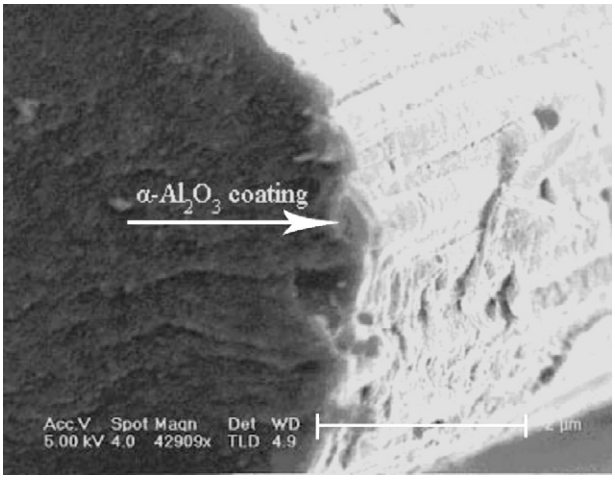

Fig. 4. FESEM micrograph of α-Al_2O_3 coating on SCF surface.

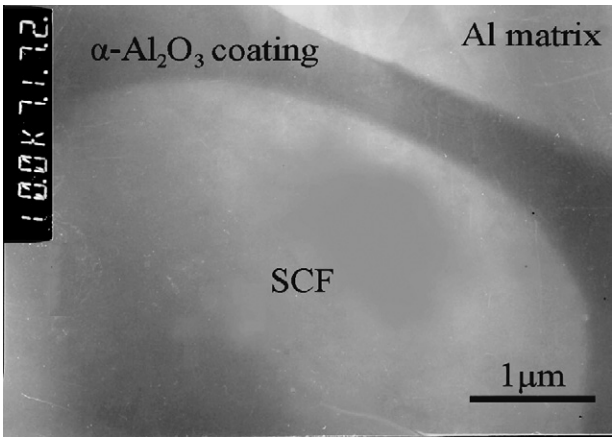

Fig. 6. TEM micrograph of the interface between α-Al_2O_3-coated SCF and aluminum matrix.

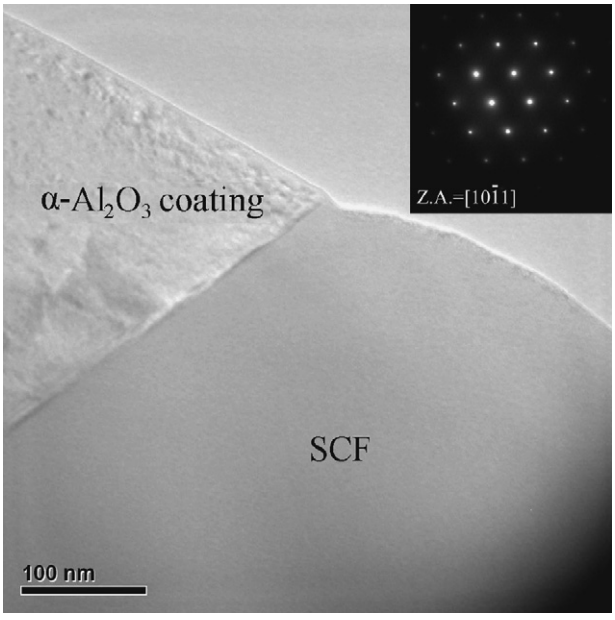

Fig. 7. High magnified TEM image of the interface zone and SAD of the α-Al_2O_3 coating.

4. Conclusions

The α-Al_2O_3 coatings with 200–250 nm thickness on SCFs surfaces were successfully fabricated by sol–gel method using aluminum nitrate and ammonia solution. The analyses of the calcinations of precursor reveal that the sol is composed of boehmite. As the temperature increase, the amorphous transit to γ-Al_2O_3 at 700 °C, and then γ-Al_2O_3 to α-Al_2O_3 at 1000 °C. The uncoated Al/SCFs composite has interfacial reactions between Al matrix and SCF, and brittle phase of Al_4C_3 formed in the interface zone. The interface of Al/Al_2O_3-coated SCFs composite was improved by the α-Al_2O_3 coating protecting fibers from the reaction with Al matrix.

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References

[1] J.B. Donnet, R.C. Bansal, M.J. Wang, Carbon Fibers, 3rd ed., Dekker, New York, 1990.
[2] S.J. Park, M.S. Cho, Effect of anti-oxidative filler on the interfacial mechanical properties of carbon–carbon composites measured at high temperature, Carbon 38 (2000) 1053–1058.
[3] S. Chand, Carbon fibers for composites, J. Mater. Sci. 35 (2000) 1303–1313.
[4] O. Perez, G. Patriarche, M. Lancin, et al., Interphase and mechanical properties in carbon fibers/Al matrix composites, Journal De Physique 3 (7) (1993) 1693–1698.
[5] K. Raghukandan, K. Hokamoto, J.S. Lee, An investigation on underwater shock consolidated carbon fiber reinforced Al composites, J. Mater. Process. Technol. 134 (3) (2003) 329–337.
[6] R.B. Bhaqat, M.F. Amateau, E.C. Smith, Damping behavior of squeeze cast planar random carbon fiber reinforced 6061 Al matrix composites, in: Proceedings of the International Symposium on Advances in Cast, 1988, p. 407.
[7] Y.M. Ryu, E.P. Yoon, M.H. Rhee, NCG reinforced MMC fabricated by the squeeze casting method, Appl. Composite Mater. 7 (4) (2000) 251–267.
[8] J.F. Silvain, A. Proult, M. Lahaye, J. Douin, Microstructure and chemical analysis of C/Cu/Al interfacial zones, Composites: Part A 34 (2004) 1143–1149.
[9] A. Urena, J. Rams, M.D. Escalera, M. Sánchez, Characterization of interfacial mechanical properties in carbon fiber/aluminum matrix composites by the nanoindentation technique, Composites Sci. Technol. 65 (2005) 2025–2038.
[10] K. Shiro, M. Norio, I. Yoshifumi, TiC coating on carbon fiber and the compatibility of TiC coated carbon fiber with aluminum, J. Jpn. Inst. Metals 43 (7) (1979) 589–594.
[11] Z. Xingui, Z. Changrui, H. Xinbo, B4C coating-containing carbon fiber reinforced SiC composites by hot pressing, High Technol. Lett. 10 (6) (2000) 68–70 (Chinese).
[12] G. Hackl, H. Gerhard, N. Popovska, Coating of carbon short fibers with thin ceramic layers by chemical vapor deposition, Thin Solid Films 513 (2006) 217–222.
[13] R.W. Jones, History, in: Fundamental Principles of Sol–Gel Technology, The Institute of Metals, USA, 1989, pp. 1–4.
[14] M. Guglielmi, Sol–gel coatings on metals, J. Sol–Gel Sci. Technol. 8 (1997) 443–449.
[15] D.R. Uhlmann, G. Teowee, Sol–gel science technology: current state and future prospects, J. Sol–Gel Sci. Technol. 13 (1998) 153–162.
[16] Z. Qingbing, Fabrication of Al_2O_3-coated carbon fiber-reinforced Al-matrix composites, J. Appl. Polym. Sci. 70 (1998) 177–183.
[17] W. Jiwen, H. Tao, L. Geyang, A combined process of coating and hybridizing for the fabrication of carbon fiber reinforced aluminum matrix composites, Composites: Part A 28A (1997) 943–948.
[18] L. Jiang, P. Yubai, X. Changshu, Low temperature synthesis of ultrafine a-Al_2O_3 powder by a simple aqueous sol–gel process, Ceram. Int. 32 (2006) 587–591.
[19] E. Saiz, A.P. Tomsia, K. Suganuma, Wetting and strength issues at Al/α-alumina interfaces, J. Eur. Ceram. Soc. 23 (2003) 2787–2796.
[20] C. McCullough, H.E. Deve, T.E. Channal, Mechanical response of continuous fiber reinforced Al_2O_3–Al composites produced by pressure infiltration casting, Mater. Sci. Eng. 189A (1) (1994) 147–154.