Wetting behaviors of the Ti_2AlC ceramics by nickel and aluminum fillers

Chengjie Lu, Liangbo Sun, Qin Qi, Jie Zhang, Gilles Hug

► To cite this version:

Chengjie Lu, Liangbo Sun, Qin Qi, Jie Zhang, Gilles Hug. Wetting behaviors of the Ti_2AlC ceramics by nickel and aluminum fillers. *Materials Letters*, 2018, 219, pp.240-242. 10.1016/j.matlet.2018.02.077 . hal-02319668

HAL Id: hal-02319668

<https://hal.science/hal-02319668>

Submitted on 2 Nov 2021

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Distributed under a Creative Commons Attribution| 4.0 International License

Wetting behaviors of the Ti₂AlC ceramics by nickel and aluminum fillers

Chengjie Lu^{1,2}, Liangbo Sun¹, Qin Qi¹, Jie Zhang^{1,*}, Gilles Hug²

¹*School of Materials Science and Engineering, Harbin Institute of Technology, Harbin 150001, China*

²*Laboratoire d'Études des Microstructures, CNRS-ONERA, Châtillon 92322, France*

Abstract

In the present work, the behaviors of nickel and aluminum based fillers on the Ti₂AlC ceramics are studied by wetting experiments and characterization of the interface cross-section. The results show that the two kinds of fillers possess different wettability on the Ti₂AlC ceramics: the contact angle of the nickel fillers decreases continuously with rising temperature and reaches less than 20° finally, whereas that of the aluminum fillers remains larger than 135° in the full temperature range investigated. The characterization of the interface from cross-section observations reveals the presence of an interaction layer in the Ti₂AlC ceramic substrate, which promotes the wetting process.

Keywords: MAX phases; wetting; interface; reaction

1. Introduction

In recent years, a group of nanolaminate ternary carbide or nitride ceramics have attracted considerable attention from researchers because of their excellent combination of metal and ceramic properties [1, 2]. They have been systematically

* Corresponding author: Tel/Fax: +86 451 86414234

E-mail address: hitzhangjie@hit.edu.cn (J. Zhang).

introduced by Barsoum *et al.* [3] with the family name ‘MAX’ and the general formula ‘ $M_{n+1}AX_n$ ’, where M is an early transition metal, A is an A-group element and X is either carbon or nitrogen. The n parameters may vary from 1 to 3 and the different structure are often named 211, 312 or 413 after their composition. The Ti_2AlC ceramics, a typical member of the family with $n=1$ (known as the ‘211’ structure), have gained extensive interests since the compound possesses the lowest density [4] and the best oxidation resistance [5] of the MAX family. However, as for other ceramics, the synthesis of bulk Ti_2AlC ceramics has proven to be difficult [6, 7]. Therefore, studies devoted to the joining of Ti_2AlC ceramics are of great significance for applications [8].

Only a few efforts have been carried out to study the joining of Ti_2AlC phases, among which the diffusion technique is the mostly adopted one. These studies revealed that the formation of Ti_2AlC -MAX diffusion couples requires high temperature, along with a long diffusion time [9]. Such conditions are often unfavorable for the thermal stability of the Ti_2AlC ceramic substrates. In addition, mechanical tests of Ti_2AlC -metal diffusion couples [10] exhibit low mechanical strength in general, which is mainly attributed to the large mismatch of the coefficients of thermal expansion of the two materials. Among the various existing joining techniques, brazing is one of the most promising for joining MAX phases, especially concerning the ceramic-metal joints [11]. In the brazing of some traditional ceramic materials, the poor wettability of the molten metallic alloys on the ceramic surface is always a critical problem, which requires to be overcome. Usually, it is done by introducing some reactive elements in

the brazing filler or by including a surface modification of the ceramic substrates. However, there is still no literature reporting on the fundamental wetting behaviors of the liquid brazing filler onto the Ti_2AlC ceramics at this moment. The present work aims at illustrating the wetting behaviors of some fillers on the Ti_2AlC substrate, thereby disclosing the wetting mechanisms of this MAX phase.

2. Materials and methods

Since the elimination of the A element in a MAX compound is considered to be the major factor accounting for its decomposition behavior [12], two kinds of fillers are chosen in this work: Nickel based fillers (including BNi-2 alloy and NiPd eutectic alloy) which are reactive to the A element, and aluminum based fillers (including the pure Al metal filler and AlSi eutectic alloy) which are not. All these fillers are provided in powder state with an average particle size of 50 μm , by Shanghai Simike Welding Materials Limited Company, China. The wetting experiments were carried out in a high vacuum furnace (VAF-30, working at around 10^{-3} Pa). The contact angle during the experiment was recorded with the camera (DV-HV 1303UM) equipped on the furnace, and the cooling speed was 5 $^{\circ}\text{C}/\text{minute}$ after wetting experiments. Finally, the cross-section morphology was investigated using a scanning electron microscope (SEM, Sigma 500, Zeiss).

3. Results and discussion

Fig. 1 shows the variations of the contact angle θ as a function of temperature T . It suggests that the two kinds of fillers possess different wetting behaviors on the

Ti₂AlC ceramics. The contact angle of the two nickel based filler alloys, as displayed in Fig. 1(a), both stay lower than 90° during the whole wetting process, and can even reach as small as 20° finally. Although the presence of the additive Cr and B elements in the BNi-2 filler alloy is believed to be favorable for the wetting process, it is still reasonable to deduce that the nickel melts possess excellent wettability on the Ti₂AlC ceramics. On the contrary, the two aluminum fillers, shown in Fig. 1(b), are not wettable on the ceramic surface, whose contact angle keeps larger than 135°. It is worthwhile pointing out that the presence of the Si element in the AlSi eutectic filler contributes to a smaller contact angle at the initial wetting stage, however, it does not alter the non-wetting behavior of the filler on the Ti₂AlC ceramics. Therefore, it can be safely assumed that the aluminum melts possess poor wettability on the Ti₂AlC ceramics.

Fig. 1. Variations in the contact angle θ as a function of temperature T for the Ti₂AlC ceramics wetted by (a). Nickel based fillers; (b). Aluminum based fillers.

The cross-section of the BNi-2/Ti₂AlC wetting sample is displayed in Fig. 2. In the left side of the figure the original separation surface is clearly apparent.

Accordingly, the reaction layer can be divided into two regions: beneath the original surface, there exists a diffusion area, which is supposed to be a solid state reaction during the wetting experiments since a highly ordered microstructure can be observed; above the original interface, there exists the solidified filler melt with the typical isothermal solidification microstructure of BNi-2 filler alloy [13], which is considered to be liquid state during the wetting experiments. Fig. 2(b) shows the magnified morphology of the cross-section area. Beneath the original surface, the formation of the interaction area is likely to be related to the diffusion behavior of the Ni element in the Ti_2AlC substrate. In addition, some modified Ti_2AlC (with lighter contrast in the figure when compared to the unmodified Ti_2AlC substrate) can be observed, which is assumed to be the original grain boundary according to the distribution morphology. Therefore, it is suggested that the nickel element from the brazing filler diffuses into the Ti_2AlC substrate mainly along the grain boundary, and then contributes to the formation of the interaction layer with the increasing nickel content. The average chemical composition of the interlayer is determined from EDS analysis to be 28.1 at.%, 11.8 at.%, 31.7 at.% and 28.4 at.% for Ti, Al, C and Ni elements, respectively. Thus it is assumed to be a mixture of $\text{Ni}_3(\text{Al, Ti})$ and TiC_x . It can be concluded from the above analysis that the diffusion of Ni element from the filler in the Ti_2AlC substrate accounts for the formation of a thick interaction layer. In addition, the presence of the additive elements (The B element for example) in the BNi-2 filler contributes to a strong interaction between the melt and the substrate, thereby leading

to a quick reduction in the contact angle. However, for the NiPd filler, without the assistance of the reactive elements, the formation of the interaction layer by diffusion of Ni and Pd elements in the Ti_2AlC substrate requires some time, which accounts for the slow reduction rate in the contact angle at the initial stage. Afterwards, the following wetting process is promoted with the formation of interaction layer, then the contact angle decreases rapidly. In conclusion, the formation of the interaction layer is assumed to be the major wetting mechanism of the nickel fillers on the Ti_2AlC ceramics.

Fig. 2. Cross-section morphology of the BNi-2/ Ti_2AlC wetting sample: (a). morphology of the edge region; (b). morphology of the middle region.

The cross-section morphology of the Al/ Ti_2AlC wetting sample is displayed in Fig. 3(a) and (b). No interaction area in the Ti_2AlC substrate can be observed around the wetting surface: the flat morphology remains unchanged, and the chemical composition ratio of Ti: Al: C stays 2: 1: 1 in the Ti_2AlC grains nearby according to the EDS results. In order to reveal the functionalization mechanisms of the pure Al filler metal on the Ti_2AlC ceramics, a supplementary wetting experiment has been carried

out, using an Al foil with a thickness of 20 μm . Instead of gathering together to form a liquid sphere when the Al powders are heated, the molten Al foil spills on the surface, and then interacts with the Ti_2AlC ceramics during the wetting process. It finally contributes to the formation of the surface with the morphology displayed in Fig. 3 (c) and (d). It can be observed from the Fig. 3(c) that a mass of grey phase with sheet morphology forms on the surface. It is identified as the intermetallic compound TiAl_3 according to the EDS result. Fig. 3(d) shows the magnified morphology of the fine particles, and the corresponding chemical composition is found to be 4.9 at.% and 85.1 at.% for Ti and Al elements, respectively, suggesting to be a solidified Al melt on the Ti_2AlC substrate. The formation of the TiAl_3 compound is most probably considered to be due to the decomposition of the Ti_2AlC ceramics thereby providing a source of Ti element. However, this reaction does not improve the wettability of the Al filler on the Ti_2AlC ceramics, because the reaction product, the TiAl_3 compound, comes into during the solidification period. It is worthwhile pointing out that the additive Si element is able to diffuse into the Ti_2AlC substrate during the wetting experiment. The diffusion behavior is likely to be favorable for promoting the interaction between the melt and the Ti_2AlC substrate, which accounts for the smaller contact angle at the initial wetting stage. However, the formation of the $\text{Ti}_2(\text{Al}, \text{Si})\text{C}$ promotes little to the wetting behavior of the AlSi filler on the Ti_2AlC ceramics, and fails to alter the non-wetting phenomenon of the Al melt on the ceramic surface, whose contact angle remains around 135° during the whole wetting experiment.

Fig. 3. Morphology of the Al/Ti₂AlC sample: (a). cross-section; (b). triple line; (c). surface morphology of the reacted area; (d). magnified morphology of the particles.

4. Conclusions

In summary, the two kinds of brazing fillers examined in this present work possess different wettability on the Ti₂AlC ceramics. The nickel based filler alloys are wettable, with the contact angle reaches as low as 20° finally, which is mainly ascribed to the interaction between Ni filler and Ti₂AlC substrate. On the contrary, the aluminum based filler alloys are non-wettable on the ceramic surface, whose contact angle remains higher than 135° during the whole wetting experiments. The weak interaction between Al filler and Ti₂AlC substrate should be blamed for the poor wettability. Therefore, the active Al atoms in the Ti₂AlC structure are considered to be

the major factor promoting the wetting process, as a result, neither the active brazing elements nor the surface pretreatment are required for the wetting and brazing of Ti_2AlC ceramics in most cases, and those fillers which are reactive to the Ti_2AlC substrate, for example, the nickel based fillers in this work, are more suitable for the brazing of Ti_2AlC ceramics.

Acknowledgements

This work was financially supported by the National Natural Science Foundation of China (NSFC) under Grant Nos. 51372049, 51621091 and U1537206. The author gratefully acknowledges the funding from China Scholarship Council (CSC).

References

- [1] M.W. Barsoum, D. Brodtkin, T. El-Raghy, Scripta Mater. 36(5) (1997) 535-541.
- [2] M. Radovic, M.W. Barsoum, Am. Ceram. Soc. Bull. 92(3) (2013) 20-27.
- [3] M.W. Barsoum, Prog. Solid State Chem. 28(1-4) (2000) 201-281.
- [4] Z.M. Sun, Int. Mater. Rev. 56(3) (2011) 143-166.
- [5] J.L. Smialek, Oxid. Met. 83(3-4) (2015) 351-366.
- [6] S.R. Kulkarni, A. Wu, J. Alloy. Compd. 490(1-2) (2010) 155-159.
- [7] Y.L. Bai, H.X. Zhang, X.D. He, et al, Int. J. Refract. Met. Hard Mat. 45 (2014) 58-63.
- [8] R.E. Loehman, A.P. Tomsia, Am. Ceram. Soc. Bull. 67(2) (1988) 375-380.
- [9] A. Ganguly, M.W. Barsoum, R.D. Doherty, J. Am. Ceram. Soc. 90(7) (2007) 2200-2204.

- [10] D.J. Tallman, J. Yang, L. Pan, et al, J. Nucl. Mater. 460 (2015) 122-129.
- [11] N. Eustathopoulos, M.G. Nicholas, B. Drevet, Wettability at high temperatures, Elsevier 1999.
- [12] W.K. Pang, I.M. Low, B.H. O'Connor, et al, J. Alloy. Compd. 509(1) (2011) 172-176.
- [13] S.K. Tung, L.C. Lim, M.O. Lai, Scripta Mater. 34(5) (1996) 763-769.