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Wettability of Ti_3SiC_2 by Ag-Cu and Ag-Cu-Ti melts

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Abstract

Recently, the ternary carbide Ti_3SiC_2 has gained much attention due to its unique characteristics combining the properties of metals and ceramics (i.e., a low density, decent thermal and electrical conductivities, an excellent thermal shock resistance, a good machinability, damage tolerance, low friction and so on). The present study describes an investigation of the wettability in high vacuum of bulk Ti_3SiC_2 by a classical braze alloy based on the Ag-Cu-Ti system. Two techniques, i.e., the sessile drop and dispensed drop methods, were utilized. The results indicated that spreading kinetics is controlled by deoxidation kinetics of Ti_3SiC_2 surface under vacuum. The final contact angle on clean Ti_3SiC_2 is very small ($\sim 10^\circ$), testifying the development of strong, metallic interactions across the liquid-solid interface. The reactivity between the ternary carbide and the liquid phase during isothermal heating at $800^\circ C$ was also considered.

Keywords: Wettability; Reactivity; Ti_3SiC_2 ; Braze alloy; Silver; Copper

Introduction

Recently, nanolaminate ternary ceramics $M_{n+1}AX_n$ (where M corresponds to an early transition metal, A is an A-group element, and X represents either carbon or nitrogen) have attracted extensive attention due to their unique combination of metals and ceramic properties [1]. Among these compounds, titanium silicon carbide (Ti_3SiC_2) has gained much interest as a promising candidate for numerous applications such as interphases in ceramic matrix composites [2, 3], electrical [4, 5] and tribological [6] contact layers or high-temperature structural applications [7, 8]. However, as for other ceramics, the synthesis of bulk Ti_3SiC_2 on a larger scale has proven difficult in practice, and for this reason, studies devoted to joining Ti_3SiC_2 with metals or ceramics are significant for promoting its applications [5].

Among the various joining technologies that exist, brazing is one of the most suitable for ceramic materials, especially for metal-to-ceramic joints [9]. It is thus important to investigate the

wettability of Ti_3SiC_2 by liquid metallic alloys. Although some studies concerning the joining of Ti_3SiC_2 with metals have already been conducted [10, 11], there exist, to the best of the authors' knowledge, no results on fundamental wettability characteristics.

The present work describes an investigation of the wetting of Ti_3SiC_2 by Ag-40at.%Cu eutectic alloy (containing Ti or not) at 800 °C by way of the sessile and dispensed drop techniques under high vacuum. The reactivity at the liquid/solid interface was also considered in order to sustain the analysis of the wetting results.

Experimental section

The classical sessile drop technique was used to study the wetting in the case of a Ti-containing alloy, and the dispensed drop method was employed for Ag-40at.%Cu eutectic alloys. The experiments were carried out in a metallic furnace under a vacuum of about 5×10^{-5} Pa obtained with a rotary vane pump and an oil diffusion pump, each equipped with a liquid nitrogen trap. In the classical sessile drop experiment, copper–silver eutectic alloy droplets of 50 to 100 mg were prepared prior to the wetting experiment by: (i) melting of pure Cu (99.999 wt.%) and Ag (99.999 wt.%) in an alumina crucible, and (ii) their deposition on vitreous carbon substrates, because of the high contact angle and low work of adhesion in this system, under high vacuum. Finally the CuAg–Ti alloy was processed *in situ* during the sessile drop experiment by directly melting a piece of Ti (purity 99.7 wt.%) over the CuAg on the Ti_3SiC_2 substrate. This procedure was used to avoid any direct contact between Ti and the ceramic before melting the alloy.

In the “dispensed drop” technique, the experiment consisted in heating Cu and Ag in an alumina crucible placed above the Ti_3SiC_2 substrate [12]. When employing this method, the melting and spreading processes could be separated, thus permitting fully isothermal tests to be performed. In both experiments, the drop images produced with an optical system fitted with a zoom (30× magnification) were recorded with a video camera connected to a computer, permitting automatic image analysis. This device enabled the linear dimensions (i.e., the drop base radius, R , and height) and the contact angle, θ , of the drop to be measured with an accuracy of $\pm 1\%$ and $\pm 2^\circ$, respectively.

All the experiments were performed using Ti_3SiC_2 substrates provided by 3-ONE-2 LLC [13]. The substrates were manufactured by pressureless sintering of a Ti_3SiC_2 powder, which resulted in a dense polycrystalline monolithic ceramic. The Ti_3SiC_2 powder was synthesized by milling of Ti, C and SiC powders followed by cold-pressing and reactive hot-pressing [1]. It should be noted that a significant amount of TiC (approximately 15 wt.%) was detected by XRD analysis. This was due to the unavoidable deviation to homogeneity when large batches of powders are processed. The substrate surface was polished down with a 1- μm diamond paste, thus giving rise to an average roughness value, R_a , of approximately a few hundred nanometers.

The chemistry, morphology and microstructure of the interfacial reaction products were determined by means of SEM (equipped with energy dispersive X-ray spectroscopy - EDXS) and electron probe microanalysis (EPMA).

Results and discussion

Wetting

Ag-40at.%Cu eutectic alloy

Fig. 1 shows the variations in contact angle, θ , and temperature as functions of time for the spreading of two AgCu eutectic alloys over Ti_3SiC_2 in the experiments performed by the dispensed drop technique.

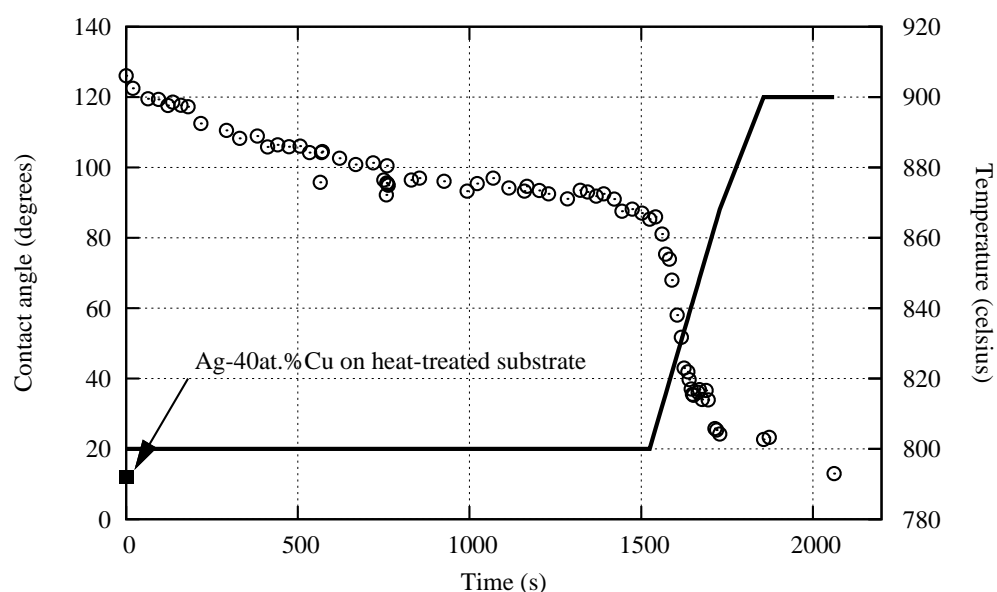


Fig. 1: variations in contact angle, θ , and temperature as functions of time for the spreading of two AgCu eutectic droplets over Ti_3SiC_2 in the experiments performed by the dispensed drop technique: (i) at 800°C after pre-heating of the substrate at 900°C (filled square, indicated by the arrow), (ii) at 800°C during about 1500 s then heated up to 900°C (open circles). Temperature vs. Time is presented by solid line.

In the first case (filled square), the substrate was heated to a temperature of 900°C for 15 min, after which the temperature was decreased to 800°C. Once the latter temperature was attained, the classical dispensed drop procedure (i.e., extrusion and deposition of the droplet) was applied. Under such conditions, an instantaneous and excellent wetting could be observed and the first measurable contact angle after 40 ms of contact was already equal to 12°.

As calculated with a liquid-vapor surface energy for AgCu eutectic alloys of 950 $mJ.m^{-2}$ [14], the work of adhesion at the liquid-solid interface has been found to be close to 1880 $mJ.m^{-2}$. This value represents 99% of the cohesion energy of the liquid, which is a sign of strong chemical interactions between the AgCu alloy and Ti_3SiC_2 across the interface. Moreover, the triple line velocity is higher than 70 $mm.s^{-1}$ (spreading over 5.4 mm for a contact time lower than 40 ms) – a feature characteristic of non-reactive metal/solid (ceramic or metallic) systems. The obtained value

is similar to those of low viscosity liquid metals on solid metals [9, 15, 16]. The results obtained regarding the work of adhesion and triple line velocity thus indicate that strong metal-like interactions has been established with the surface of Ti_3SiC_2 .

It is already known that Ti_3SiC_2 has various metal-like properties: at room temperature it demonstrates a thermal conductivity similar to pure Ti ($37 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ as opposed to $30 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ for pure Ti) and an electrical conductivity higher than that of pure Ti or TiC ($4.5 \times 10^6 \text{ V}^{-1}\cdot\text{m}^{-1}$ as opposed to $2.3 \times 10^6 \text{ V}^{-1}\cdot\text{m}^{-1}$ for Ti and $1.6 \times 10^6 \text{ V}^{-1}\cdot\text{m}^{-1}$ for TiC) [1, 17]. The apparent combination of ceramic and metallic properties has been explained by the existence of two types of Ti bonding in the crystal structure [18]. Indeed, the Ti_3SiC_2 phase consists of a layered structure with a double Ti–C block, each block made up of two edge-sharing CTi_6 octahedra, separated by a square planar Si sheet. It should be noted that the titanium carbide structure can also be described as a stacking of such octahedra. The stacking sequence of Ti_3SiC_2 ($\text{Ti}_{\text{II}}\text{–C–Ti}_{\text{I}}\text{–C–Ti}_{\text{II}}\text{–Si}$) contains two unique sites for Ti, and there consequently also exist two unique chemical states for the Ti atom.

The charge density of Ti in the first C– Ti_{I} –C layer has been shown to be similar to that of Ti in Ti–C but with a lower average spacing (0.213 nm instead of 0.216) [19, 20, 21]. In contrast, the charge density of the second type of Ti site (i.e. C– Ti_{II} –Si sequence) contains a higher proportion of metallic bonds and hence contributes more to the metallic properties of Ti_3SiC_2 . This is due to a slight distortion of the CTi_6 octahedra and a $\text{Ti}_{\text{II}}\text{–Si}$ bond distance that is somewhat larger than that of a normal covalent bond [19, 21].

In the second case (open circles in Fig. 1), the liquid droplet was extruded from the crucible and dispensed onto the Ti_3SiC_2 substrate once the experimental temperature reached 800°C . The spreading proceeded from the first measurable contact angle value of 126° with a triple line velocity of about $1 \mu\text{m}\cdot\text{s}^{-1}$. After 1250 s, the temperature was raised to 900°C with a heating rate of $20^\circ\text{C}\cdot\text{min}^{-1}$, which was followed by an increase in the spreading velocity up to a maximum of about $20 \mu\text{m}\cdot\text{s}^{-1}$. Finally, the capillary equilibrium was rapidly obtained, with a contact angle value of 20° , before the temperature of 900°C was reached. One should note that this final contact angle value could not be compared to the one from the first experiments. Indeed, during spreading, the surface chemistry of the substrate was modified by the reactivity between the liquid and Ti_3SiC_2 . This is described further in the section concerning the reactivity.

The low spreading rate and the high activation energy of the spreading process are typical for reactive metal/ceramic systems when the reaction at the solid/liquid/vapor triple line controls the spreading rate [12]. In the Ag-40at.%Cu/ Ti_3SiC_2 system, the long spreading time is attributed to the deoxidation of the Ti_3SiC_2 surface under very low oxygen partial pressure (approximately 10^{-20} mbar). Indeed, it has been established that the formation of the native oxide layer on a pure polycrystalline Ti_3SiC_2 sample is the result of a chemical interaction in two systems, i.e., in Ti–C–O for the C– Ti_{I} –C bonds and in Si–Ti–C–O for the C– Ti_{II} –Si bonds [21].

The obtained experimental results, especially the high spreading rate observed after the heat treatment at 900°C ($70 \text{ mm}\cdot\text{s}^{-1}$), are attributed to the fact that the Ti_3SiC_2 surface has been deoxidized at 900°C , without any contact with the liquid, and thus has acquired a metallic character (see Fig. 1). It is interesting to note that Kozlova et al. observed a similar behavior for Ti

containing stainless steel substrates [22]. According to their results the deoxidation of the solid surface is mainly due to reduction by residual carbon present in solid steel to form carbon monoxide (CO). In the present study, the same atmosphere is used and the C activity in the solid should also be approximately the same because some TiC is present both in stainless steels and in Ti_3SiC_2 substrates. Therefore, the same mechanism, reduction of oxide film by C leading to CO formation, can be responsible of deoxidation of the surface under vacuum. At 800°C, this process can remain efficient but its kinetics is significantly reduced, and instantaneous wetting cannot be observed.

Because of structural similarities (i.e., the presence of Ti_6C edge-sharing octahedra), it is interesting at this point to compare the present results with contact angle values reported for TiC_x in the literature [21], even if deoxidation at low temperatures of that carbide is difficult [23]. For TiC_x with the same ratio between C and Ti as in Ti_3SiC_2 (i.e., $x = 0.67$), contact angles are much higher than on Ti_3SiC_2 (from 60° to 80° for pure Cu and Ag as opposed to 10° in the present study for Ag-40at%Cu) [24]. Therefore, the metallic character of the MAX phase Ti_3SiC_2 is more pronounced than for TiC_x , even when sub-stoichiometric.

Ti-containing Ag-40at.%Cu eutectic alloys

Ti is commonly added to the Ag-40at.%Cu eutectic alloy to produce the most widely used active braze family. Moreover, the dissolution of Ti from a Ti_3SiC_2 substrate in the liquid alloy can be expected during the spreading process. For this reason, various experiments have been performed on AgCu-2,5wt.%Ti by the classical sessile drop technique. It is worth noting that pre-heating to higher temperatures is not allowed with this procedure.

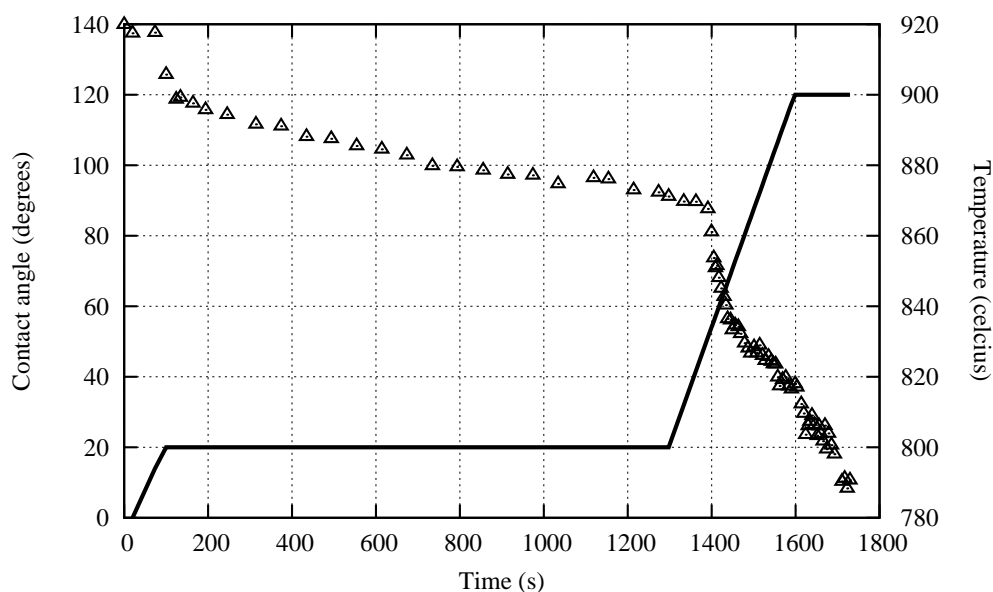


Fig. 2: Contact angle measured by the classical sessile drop technique as a function of time for AgCu-2,5wt.%Ti alloy on Ti_3SiC_2 at 800°C during about 1300 s and then after heating up to 900°C

Figure 2 presents the variation in contact angle θ and temperature as functions of time for the spreading of AgCu-2,5wt.%Ti over Ti_3SiC_2 (open triangles) with the associated temperature evolution (solid line). The first measurable contact angle under isothermal conditions of 800°C was about 125° , i.e., nearly the same as for dispensed drop experiments performed without Ti. Subsequently, spreading occurred with a triple line velocity of about $1 \mu\text{m}\cdot\text{s}^{-1}$, which was similar to the value observed for the AgCu eutectic alloy without Ti (cf. filled square in Fig. 1). After 1300s, the temperature was raised to 900°C with a heating rate of $20^\circ\text{C}\cdot\text{min}^{-1}$, which led to an increase in the spreading velocity by one order of magnitude and, finally, to capillary equilibrium with a contact angle value of approximately 10° .

It is worth noting that the addition of Ti has no effect on the initial contact angle value, nor on the triple line velocity. Concerning the final contact angle values (12° against 8°), they can be considered as nearly the same.

It can be concluded that Ti has no significant effect on the process of deoxidation of the Ti_3SiC_2 substrate. This conclusion is counter-intuitive since Ti is recognized as an element promoting wetting on numerous solids such as oxides [25], nitrides [26, 27] or carbides [28, 29]. This effect of Ti is usually attributed to the formation by reaction with the solid substrate of a better wetted compound with a metallic character, e.g., TiC_{1-x} or TiO_{1-x} [30]. With Ti_3SiC_2 , a possible reaction would be the transformation of the ionic-covalent native oxide film to a thin continuous layer of the wettable oxide TiO_{1-x} .

In the present case, the thermodynamic activity of Ti is too low to promote the formation of TiO_{1-x} with decent wetting. Indeed, during temperature rise to 900°C the kinetics of dissolution of Ti in Ag-40at.%Cu eutectic is significantly reduced by the formation at solid Ti/liquid AgCu interface of different Cu_xTi_y compounds such as Cu_4Ti , Cu_3Ti_4 or CuTi [31]. As a consequence the effective activity of Ti in the drop is lower than the nominal one.

Reactivity

Ag-40at.%Cu eutectic alloy

Figure 3 shows an SEM micrograph representing a cross-section of the interface between an AgCu eutectic and a Ti_3SiC_2 substrate after a prior heat treatment of the substrate at 900°C and a subsequent isothermal spreading at 800°C (filled square in Figure 1). No interfacial reaction layer was formed at the substrate surface. However the spreading was followed by a deep penetration of the liquid into the grain boundaries of the substrate, as revealed by the presence of bright solidified silver channels in Fig. 3. This penetration phenomenon was associated with solid state diffusion of Cu into the grains of Ti_3SiC_2 . As a consequence the substrate was transformed up to 40 micrometers below the liquid/solid interface after 420 s contact at 800°C . Quantitative electron probe micro analyses have pointed out a composition of the transformed light grey grains that is

likely $\text{Ti}_{47}\text{Cu}_8\text{Si}_{13}\text{C}_{32}$. At the present time, it is not possible to determine whether this composition is that of a quaternary phase or of an intimate mixture of fine-grained binary or ternary phases. Note that Si was barely detectable by EPMA in the solidified liquid with a mean amount lower than 1 wt%.

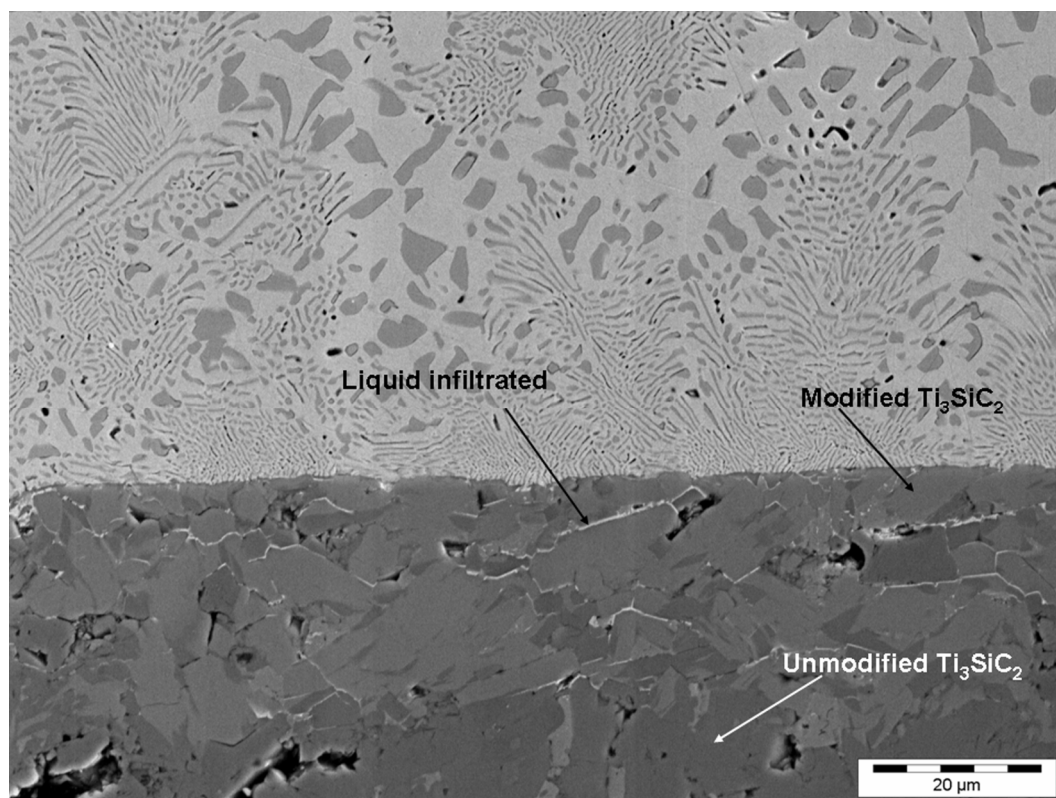


Fig. 3: SEM micrograph representing a cross-section of the interface between an AgCu eutectic and a Ti_3SiC_2 substrate after a prior heat treatment of the substrate at 900°C and a subsequent contact time of 420 s at 800°C (filled square in Figure 1).

It is worth noting that the existence of reactivity between Cu and Ti_3SiC_2 has already been reported on by Guo et al. in the case of Cu magnetron sputtering layers heat-treated at temperatures ranging from 900°C to 1100°C [32]. The authors identified Cu_3Si and substoichiometric TiC_{1-x} as the reaction products. Moreover, other researchers have already reported on topotactic transformation of Ti_3SiC_2 by outward diffusion of Si along the layers of the structure that can be partially balanced by inward diffusion of certain other elements [33, 34, 35]. In the present case, the partial exchange of Si atoms for Cu in the structure between two layers of CTi_6 octahedra could be considered. A complete reactivity study of Ag-Cu(Ti) melts on Ti_3SiC_2 is currently in progress.

Ti-containing AgCu eutectic alloys

As demonstrated in Figure 4, an AgCu eutectic to which Ti is added differs from a pure AgCu eutectic in that a secondary phase has appeared in the liquid. Crystals with hexagonal shape were

identified by EDXS analysis as $Ti_5Si_3C_x$, signifying that some limited dissolution of Ti_3SiC_2 had occurred. This, in turn, led to an increase in the amount of Si in the liquid and finally to the precipitation of $Ti_5Si_3C_x$. It was also observed that, after 20 min at 800°C and 10 min at 900°C, the Ti_3SiC_2 substrate was penetrated and transformed up to a depth of 100 μm .

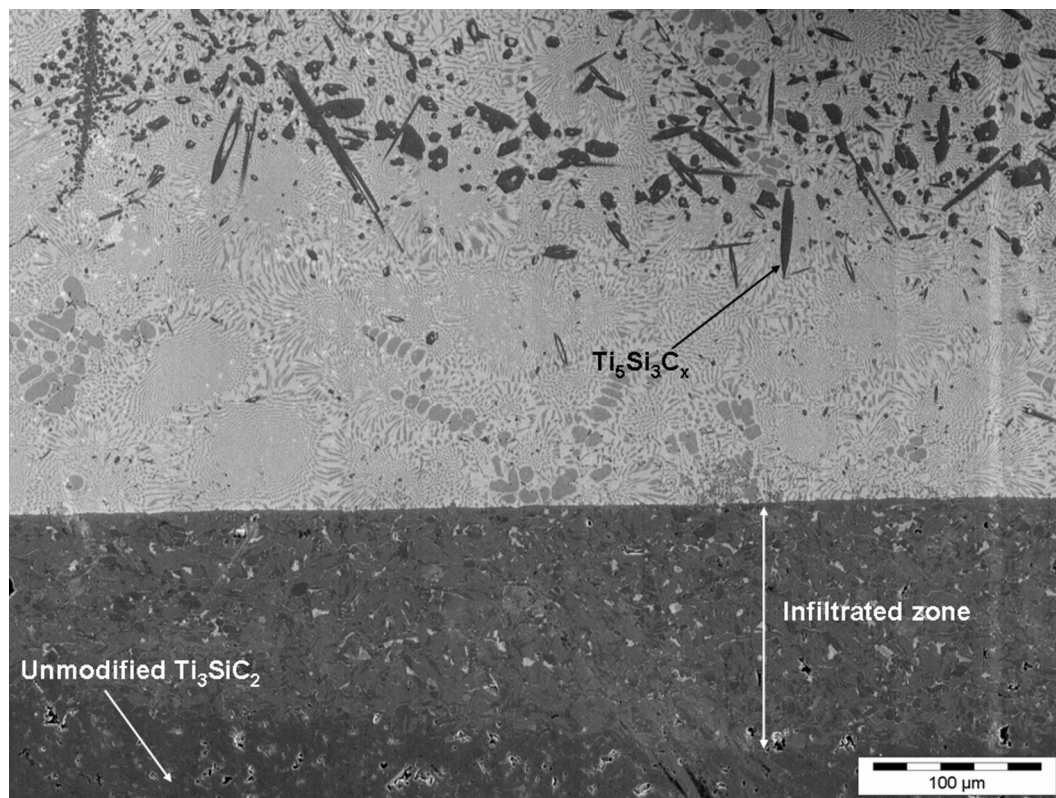


Fig. 4: SEM micrograph representing a cross-section of the interface between an AgCu-2,5wt.%Ti melt and a Ti_3SiC_2 substrate after 7 min of contact at 800°C and 10 min at 900°C (experiment presented by open triangles in Figure 2)

Conclusions

Wetting experiments performed by the dispensed drop technique have demonstrated that a Ti_3SiC_2 surface can be deoxidized under high vacuum above 850°C leading to a strong metallic character. The results indicated that spreading kinetics is controlled by deoxidation kinetics of Ti_3SiC_2 surface under vacuum. The final contact angle on clean Ti_3SiC_2 is very small ($\sim 10^\circ$), testifying the development of strong, metallic interactions across the liquid-solid interface. The wetting experiments also confirmed the existence of a complex reactivity between Cu and Ti_3SiC_2 and a study dedicated to this reactivity is on-going.

References

- [1] Barsoum MW, El-Raghy T. Synthesis and characterization of a remarkable ceramic: Ti_3SiC_2 (1996), *J Am Ceram Soc* 79(7):1953. doi: 10.1111/j.1151-2916.1996.tb08018.x.
- [2] Lan F, Li K, Li H, He Y, Shen X, Cao W. Joining of carbon/carbon composites for nuclear applications (2009), *J Mater Sci* 44(14):3747. doi: 10.1007/s10853-009-3501-0.
- [3] Robin-Brosse C, Rocher JP, Naslain R. Composite material with a ceramic matrix with lamellar interphase between refractory reinforcing fibres and matrix, and process for its manufacture, Patent n°FR2675141 (6/08/1993).
- [4] Tuller H, Spears M, Mlcak R. Stable electrical contact for silicon carbide devices, Patent n°US6544674 (06/06/2002).
- [5] Wenzel R, Goesmann F, Schmid-Fetzer R. Bulk diffusion studies of metallizations on titanium- based contacts at 600°C (1998), *Mater Sci Eng B-Solid* 52(2-3):175. doi: 10.1016/S0921-5107(97)00284-5.
- [6] Myhra S, Summers JWB, Kisi EH. Ti_3SiC_2 –a layered ceramic exhibiting ultra-low friction (1999), *Mater Lett* 39(1):6. doi: 10.1016/S0167-577X(98)00206-7.
- [7] Barnes L, Rago ND, Leibowitz L. Corrosion of ternary carbides by molten lead (2008), *J Nucl Mater* 373(1-3):424. doi: 10.1016/j.jnucmat.2007.04.054.
- [8] Nappé J, Grosseau P, Audubert F, Guilhot B, Beauvy M, Benabdesselam M, Monnet I. Damages induced by heavy ions in titanium silicon carbide: Effects of nuclear and electronic interactions at room temperature (2009), *J Nucl Mater* 385(2):304. doi: 10.1016/j.jnucmat.2008.12.018.
- [9] Eustathopoulos N, Nicholas M, Drevet B. Wettability at High Temperatures. Pergamon, Pergamon Materials Science Series (1999) vol. 3.
- [10] Gao N, Miyamoto Y. Joining of Ti_3SiC_2 with Ti-6Al-4V alloy (2002), *J Mater Res* 17(1):52. doi: 10.1557/JMR.2002.0010.
- [11] Gu W-L., Zhou C-Y.. Reactions between Ti and Ti_3SiC_2 in temperature range of 1273-1573 K (2006), *Transactions of Nonferrous Metals Society of China* 16(6):1281. doi: 10.1016/S1003-6326(07)60007-7.
- [12] Dezellus O, Hodaj F, Rado C, Barbier JN, Eustathopoulos N. Spreading of Cu-Si alloys on oxidized SiC in vacuum: experimental results and modelling. (2002), *Acta Mater* 50:979.
- [13] 3-ONE-2 LLC, Voorhees NJ, USA.
- [14] Bricard A, Eustathopoulos N, Joud JC, Desré P (1972), *Cr Acad Sci II C* 276:1613.
- [15] Naidich Y, Zabuga V, Perevertailo V. Temperature effect on the kinetics of spreading and wetting in systems with different character of interaction of contacting phases (1992), *Adgeziya rasplavov i paika materialov* 27:23.
- [16] Saiz E, Tomsia AP. Atomic dynamics and marangoni films during liquid-metal spreading (2004), *Nat Mater* 3(12):903. doi: 10.1038/nmat1252.
- [17] Barsoum MW, El-Raghy T, Rawn CJ, Porter WD, Wang H, Payzant EA, Hubbard CR. Thermal properties of Ti_3SiC_2 (1999), *J Phys Chem Solids* 60(4):429. doi: 10.1016/S0022-3697(98)00313-8.
- [18] Zhou Y, Sun Z. Electronic structure and bonding properties in layered ternary carbide Ti_3SiC_2 (2000), *J Phys: Condens Matter* 12(28):L457.
- [19] Kisi EH, Crossley JAA, Myhra S, Barsoum MW. Structure and crystal chemistry of Ti_3SiC_2 (1998), *J Phys Chem Solids* 59(9):1437. doi: 10.1016/S0022-3697(98)00226-1.
- [20] Medvedeva NI, Novikov DL, Ivanovsky AL, Kuznetsov MV, Freeman AJ. Electronic properties of Ti_3SiC_2 -based solid solutions (1998), *Physical Review B* 58(24):16042. doi: 10.1103/PhysRevB.58.16042.
- [21] Riley DP, O'Connor DJ, Dastoor P, Brack N, Pigram PJ. Comparative analysis of Ti_3SiC_2 and associated compounds using x-ray diffraction and x-ray photoelectron spectroscopy (2002), *J Phys D: Appl Phys* 35(13):1603.
- [22] Kozlova O, Voytovych R, Devismes MF, Eustathopoulos N. Wetting and brazing of stainless steels by copper-silver eutectic (2008), *Mater Sci Eng A* 495(1-2):96. doi: 10.1016/j.msea.2007.10.101.
- [23] Li JG. Influence of oxygen partial pressure on the wetting behaviour of titanium carbide by molten copper and other metals (1993), *Mater Lett* 17(1-2):74. doi: 10.1016/0167-577X(93)90151-M.
- [24] Frage N, Froumin N, Dariel MP. Wetting of TiC by non-reactive liquid metals (2002), *Acta Mater* 50(2):237. doi: 10.1016/S1359-6454(01)00349-4.

- [25] Voytovych R, Robaut F, Eustathopoulos N. The relation between wetting and interfacial chemistry in the CuAgTi/alumina system (2006), *Acta Mater* 54(8):2205. doi: 10.1016/j.actamat.2005.11.048.
- [26] Loehman RE, Tomsia AP. Reactions of Ti and Zr with AlN and Al₂O₃ (1992), *Acta Met Mat* 40(Supplement 1):S75. doi: 10.1016/0956-7151(92)90266-H.
- [27] Nicholas MG, Mortimer DA, Jones LM, Crispin RM. Some observations on the wetting and bonding of nitride ceramics (1990), *J Mater Sci* 25(6):2679. doi: 10.1007/BF00584866.
- [28] Dezellus O, Hodaj F, Mortensen A, Eustathopoulos N. Diffusion-limited reactive wetting: spreading of Cu-Sn-Ti alloys on vitreous carbon (2001), *Scr Mater* 44(11):2543. doi: 10.1016/S1359-6462(01)00946-0.
- [29] Naidich Y, Kolesnichenko G (1968), *Russ Metall* 4:141.
- [30] Voytovych R, Ljungberg LY, Eustathopoulos N. The role of adsorption and reaction in wetting in the CuAg-Ti/alumina system (2004), *Scr Mater* 51(5):431. doi: 10.1016/j.scriptamat.2004.05.002.
- [31] Andrieux J, Dezellus O, Bosselet F, Viala J. Low-temperature interface reaction between titanium and the eutectic silver-copper brazing alloy (2009), *J Phase Equilib Diffus* 30(1):40. doi: 10.1007/s11669-008-9424-7.
- [32] Guo H, Zhang J, Li F, Liu Y, Yin J, Zhou Y. Surface strengthening of Ti₃SiC₂ through magnetron sputtering Cu and subsequent annealing (2008), *J Eur Ceram Soc* 28(10):2099. doi: 10.1016/j.jeurceramsoc.2008.02.011.
- [33] Barsoum M, El-Raghy T, Ogbuji U. Oxidation of Ti₃SiC₂ in air (1997), *J Electrochem Soc* 144(7):2508. doi: 10.1149/1.1837846.
- [34] Barsoum MW. Comment on "reaction layers around SiC particles in Ti: an electron microscopy study" (2000), *Scr Mater* 43(3):285. doi: 10.1016/S1359-6462(00)00404-8.
- [35] El-Raghy T, Barsoum MW, Sika M. Reaction of Al with Ti₃SiC₂ in the 800-1000°C temperature range (2001), *Mater Sci Eng A* 298(1-2):174. doi: 10.1016/S0921-5093(00)01281-8.