Effect of Ti_3SiC_2 Amount on Microstructure and Properties of TiAl Matrix Composites

Qin Zou

Yanshan University

Zhichao Lou

Yanshan University

Yong Guan

Yanshan University

YanGuo Li (✉ lyg@ysu.edu.cn)

State Key laboratory of Metestable Materials Science and Technology

Jiangbo Xu

Yanshan University

Yuanyuan Li

Yanshan University

Lingyu Bu

Yanshan University

Peng Wang

Yanshan University

Yong'an Luo

Yanshan University

Research Article

Keywords: TiAl matrix composite, Ti_3SiC_2 , Microstructure, Mechanical properties, Tribology

Posted Date: November 17th, 2021

DOI: <https://doi.org/10.21203/rs.3.rs-1050375/v1>

License: This work is licensed under a Creative Commons Attribution 4.0 International License.

[Read Full License](#)

Abstract

The TiAl matrix composites were manufactured using spark plasma sintering under the conditions of 1100 °C/10 min/30 MPa. The effect of Ti_3SiC_2 amount on microstructure and properties of TiAl matrix composites was investigated. Ti_3SiC_2 was homogeneously distributed in the TiAl matrix, and it partly decomposed to form Ti_5Si_3 and TiC. The TiAl matrix with 30 wt.% of Ti_3SiC_2 exhibited the lowest friction coefficient and wear rate of 0.507 and $1.35 \times 10^{-4} \text{ mm}^3\text{N}^{-1}\text{m}^{-1}$ at room temperature and 0.423 and $0.21 \times 10^{-4} \text{ mm}^3\text{N}^{-1}\text{m}^{-1}$ at 550 °C, while the compression strength reached the maximum value of 1080 GPa at room temperature and 640 GPa at 550 °C, respectively. The hardness reached the value of 5.1 GPa. The TiAl matrix composites had a lower friction coefficient and wear rate at 550 °C than at room temperature. A Ti_3SiC_2 lubricating film was formed on the friction surface of the TiAl matrix composites after friction test at room temperature, while a Fe-Ti-Al-Si-oxide lubricating film was formed after friction test at 550 °C. The wear mechanisms of the TiAl matrix composites with the Ti_3SiC_2 addition were mainly abrasive wear and adhesive wear at room temperature and 550 °C, respectively.

Highlights

1. The mechanical and tribological properties of the TiAl matrix composites improved with the Ti_3SiC_2 amount.
2. The addition of Ti_3SiC_2 introduces high density and hardness Ti_3SiC_2 , TiC, and Ti_5Si_3 phases, while TiAl still exists as a major phase of the TiAl matrix composites, which TiC and Ti_5Si_3 are formed by the Ti_3SiC_2 decomposition.
3. Ti_5Si_3 and TiC formed at the interface of the TiAl matrix and Ti_3SiC_2 induce a pinning effect, which is conducive to the stress transfer from the TiAl matrix to Ti_3SiC_2 with higher pressure, and it also impedes grain boundary sliding and increases the resistance to dislocation movement, ultimately increasing the hardness and compression strength.
4. The composition and integrity of the lubricating film play important roles in influencing the tribological properties of the TiAl matrix composites.
5. The composition of the lubricating film is determined by the friction temperature and the composition of the TiAl matrix composites, resulting in different tribological properties after friction tests at room temperature and 550 °C.
6. The integrity of the lubricating film is affected by the synergy of the Ti_3SiC_2 content in the TiAl matrix composites and the interfacial bonding between Ti_3SiC_2 and the TiAl matrix.

1 Introduction

The TiAl matrix composites have attracted great attention due to their excellent properties, such as low density (3.85-4.2 g/cm³) [1], high specific strength [2], high oxidation resistance [3], and high temperature creep resistance [4]. They are widely used in the fields of aeroengine technology [5], automotive

turbochargers [6], and marine ship plates [7]. Many methods are currently used to prepare TiAl matrix composites, including electron beam casting [8], self-propagating high-temperature synthesis [9], hot-pressing sintering [10], and spark plasma sintering (SPS) [11]. The addition of Ti_3SiC_2 could improve tribological properties of the TiAl matrix composite by forming a Ti-Al-Si-oxide lubricating film on the friction surface after friction test above 400 °C [12,13], avoiding the lubrication failure of conventional lubricants (graphite, Ag, etc.) caused by the oxidation at such friction temperatures [14]. The friction coefficient and wear rate of the TiAl matrix composite with the added Ti_3SiC_2 decreased by 4.03-35.09% and 44.78-53.40%, respectively, compared to the pure TiAl matrix composite [15,16].

Although various studies have shown that Ti_3SiC_2 can improve tribological properties of the TiAl matrix composites, only a few studies have reported the relationship between the state of lubricating film and the tribological properties at room and high temperature. In addition, the explanation of the underlying mechanism of the Ti_3SiC_2 beneficial effect requires a more systematic study. In this work, the TiAl matrix composites with added 0-30 wt.% of Ti_3SiC_2 were synthesized by the SPS method. The effect of Ti_3SiC_2 amount on the microstructure and properties of the TiAl matrix composites was systematically studied. In addition, we discussed in detail the relationship between the lubricating film and the tribological properties of the TiAl matrix composites.

2 Experimental Detail

Ti, Al, Cr, and Ti_3SiC_2 powders were used as raw materials. The purity and size of Ti and Al powders were both 99.5% and 38 μm , while the purity and size of Cr powder was 99.7% and 75 μm . Ti_3SiC_2 with a grain size of 80-120 μm and a purity of 91.37% was fabricated according to the production method described in Ref. 17 (see Part 3). The weight fraction of Ti_3SiC_2 in the whole raw powders was fixed at 0, 10, 20, and 30 wt.%, so the corresponding sintered samples were denoted as TA0 (0 wt.% Ti_3SiC_2), TA1 (10 wt.% Ti_3SiC_2), TA2 (20 wt.% Ti_3SiC_2), and TA3 (30 wt.% Ti_3SiC_2). These raw materials were well dispersed via high-energy ball-milling, and then the mixed powders were sintered at a sintering temperature of 1100°C, holding time of 10 min, and pressure of 30 MPa, in a 30 Pa vacuum using a spark plasma sintering system. The heating rate was set to 70°C/min.

An X-ray diffractometer (D/Max-2500PC, Japan) and a field-emission scanning electron microscope (FESEM, S-4800, Japan) equipped with energy-dispersive X-ray spectroscopy (EDS, EX-250, Japan) were used to analyze the phase composition of the TiAl matrix composites and Ti_3SiC_2 . A metallurgical microscope (Axiovert 200 MAT, China) was used to assess the metallographic characteristics of the TiAl matrix composites after immersion in aqua regia for 100 s. The bulk density of the TiAl matrix composites was measured by Archimedes' method. The theoretical density of TA0, TA1, TA2, and TA3 was 3.88, 4, 4.05, and 4.09 g/cm³, calculated as in the method described in Ref. 18. A Vicker's hardness instrument (FM-ARS9000, China) was used to assess the hardness of the TiAl matrix composites with a dwell time of 15 s at a load of 500 gf. A Gleeble thermal simulated test machine (3500, China) was used to determine the compression strength of the TiAl matrix composites with a compression ratio of 20%

and a compression rate of 0.05 mm/min at room temperature (25°C) and 550°C. Friction and wear tests were carried out using a screen display terminal face friction and wear tester (MMU-10G, China) with a pin-on-disc configuration. The size of the disc-specimens of TiAl matrix composites was $\Phi 20.6$ mm. The GCr15 pins were used as the friction counterparts with a hardness of $HRC50 \pm 3$, which the one top of pin-specimen was hemispherical with a diameter of 4 mm. Disc-specimens and GCr15 pins were cleaned by acetone solution before frictional wear tests. All tests were carried out under dry sliding conditions in the air. The wear track diameter was typically 12-14 mm. The friction test was performed at room temperature and 550°C under a 300 N load and at a 0.261 m/s gliding speed for 10 min. The friction coefficient was automatically recorded by the computer system of the friction tester, while the wear rate was calculated using wear volumes described as [19]:

$$W = V / P \cdot L$$

where $V(\text{mm}^3)$ is the wear volume obtained by measuring the weight loss in a microbalance after ultrasonic cleaning and drying and the measured density, $P(\text{N})$ is the applied load, and $L(\text{m})$ is the total sliding distance.

3 Results And Discussion

The Ti_3SiC_2 sample is composed of Ti_3SiC_2 particles (marked by A and B in Fig. 1b), tightly combined with some TiC particles (marked by C in Fig. 1b). The relative content of Ti_3SiC_2 is about 91.37%, calculated by the internal standard method [20] based on Fig. 1a. The Ti_3SiC_2 particles exhibit irregular and lamellar shape with a particle size of 4 to 5 μm . TiAl alloy is composed of TiAl and Ti_3Al . With the Ti_3SiC_2 addition, the XRD patterns of the composites show the presence of Ti_3SiC_2 , TiC, and Ti_5Si_3 in the TiAl matrix composites, while TiAl still exists as a major phase (Fig. 1c). The relative content of each phase in the TiAl matrix composites is calculated by the semi-quantitative method [21]. With the increase of the Ti_3SiC_2 amount, the relative contents of Ti_3SiC_2 , TiC, and Ti_5Si_3 increase, while the relative contents of TiAl and Ti_3Al decrease (Fig. 1d), implying that Ti_5Si_3 and TiC are mainly formed by the Ti_3SiC_2 decomposition. Previous studies have reported that the TiC formation may be attributed to the reaction of TiAl and graphite atmosphere generated by the graphite molds during the sintering process [12, 13, 15, 16]. However, in this work, the molten Al can promote the Ti_3SiC_2 decomposition to form TiC and Ti_5Si_3 [22–24]. The formation of Ti_5Si_3 and TiC improves the mechanical and tribological properties of the TiAl matrix composites in this work. The Ti_3SiC_2 decomposition reaction can be described as follows:

Ti_3SiC_2 (marked by yellow rhombus in Fig. 2c and Fig. 4) is evenly distributed in the TiAl matrix (Figs. 2-3). The distribution and agglomeration of Cr in the TiAl matrix are indicated by the blue triangle in Fig. 2a and the blue ellipse area in Fig. 3. There are few distinct interstices between Ti_3SiC_2 and TiAl matrix (Fig. 4a). Ti from TiAl and Si from Ti_3SiC_2 exhibit mutual diffusion at the interface between Ti_3SiC_2 and the TiAl matrix. The diffusion distance of Si in Ti_3SiC_2 of TA2 is about 2 μm , while that of Ti in TiAl is about 3 μm (Figs. 4b-c). The diffusion of elements yields the formation of TiC and Ti_5Si_3 from the Ti_3SiC_2 decomposition, which can change the interfacial structure between Ti_3SiC_2 and the TiAl matrix, improving the mechanical and tribological properties of the composite [25]. With the increase of the Ti_3SiC_2 amount, the Ti_3SiC_2 agglomeration becomes more pronounced (Fig. 2), deteriorating the mechanical and tribological properties of the TiAl matrix composites. The volume fraction of Ti_3SiC_2 in TA1, TA2, and TA3 is approximately 8.22, 16.8, and 26.2%, respectively, Fig. 2, as determined by using Image-Pro Plus (IPP) software [26]. This corroborates the relative content of Ti_3SiC_2 obtained from the XRD patterns of TA1, TA2, and TA3 (Fig. 1d), which further confirms the result that Ti_3SiC_2 is partly decomposed in the TiAl matrix composite.

The bulk density, relative density, and hardness of the TiAl matrix composites are in the range of 3.75-3.88 g/cm^3 , 99.2-99.6%, and 4-5.3 GPa, respectively, while the corresponding values of compression strength, friction coefficient, and wear rate at room temperature and 550°C are in the range of 753-1080 MPa and 298-640 MPa, 0.507-0.581 and 0.423-0.484, $(1.35-2.15) \cdot 10^{-4} \text{ mm}^3\text{N}^{-1}\text{m}^{-1}$ and $(0.21-0.26) \cdot 10^{-4} \text{ mm}^3\text{N}^{-1}\text{m}^{-1}$, respectively (Figs. 5a-e). The values of bulk density and hardness exhibit the same increasing and then decreasing trends with the rise of the Ti_3SiC_2 amount. The compression strength at room temperature and 550°C exhibits an increasing trend, while the relative density, friction coefficient, and wear rate at room temperature and 550°C exhibits a decreasing trend. In the case of 20 wt.% Ti_3SiC_2 addition, the bulk density and hardness of the TiAl matrix composites reach the maximum values of 3.88 g/cm^3 and 5.3 GPa, respectively. At the 30 wt.% Ti_3SiC_2 addition, the compression strength at room temperature and 550°C reach the maximum values of 1080 and 640 GPa, respectively, while the friction coefficient and wear rate reach the minimum values of 0.507 and $1.35 \cdot 10^{-4} \text{ mm}^3\text{N}^{-1}\text{m}^{-1}$, and 0.423 and $0.21 \cdot 10^{-4} \text{ mm}^3\text{N}^{-1}\text{m}^{-1}$ at room temperature and 550°C, respectively. The synthesized Ti_3SiC_2 -TiAl composites exhibit an extremely low wear rate of about $0.21 \cdot 10^{-4} \text{ mm}^3\text{N}^{-1}\text{m}^{-1}$ at 550°C compared with the previous studies (Fig. 5f) [12, 13, 15, 16, 27–29]. In addition, the friction coefficient and wear rate at room temperature and the friction coefficient at 550°C are close to the mean values reported in the previous studies.

The friction coefficient and wear rate of the TiAl matrix composite with the Ti_3SiC_2 addition decrease by about 11.3% and 29.4% at room temperature, and 10.9% and 19.23% at 550°C, respectively, compared with the TiAl alloy itself. The groove (marked by D and F in Fig. 6a and Table 1) exists on the friction surface of TiAl alloy after friction test at room temperature, while abrasive debris (marked by Q in Fig. 6a and Table 1) and plastic deformation (marked by R in Fig. 6e and Table 1) appear after friction test at 550°C. Besides the grooves (marked by G, K, and N in Figs. 6b-d and Table 1), abrasive debris (marked by

U and W in Figs. 6f-g and Table 1) and plastic deformation (marked by T, V, and Z in Figs. 6e-h and Table 1) exist on the friction surface of the TiAl matrix composites with the Ti_3SiC_2 addition after friction test at room temperature and 550°C, while the peeling (marked by H, J, and M in Figs. 6b-d and Table 1) appears on the friction surface after friction test at room temperature. The difference in tribological properties between the TiAl alloy and the TiAl matrix composites with the Ti_3SiC_2 addition is attributed to the formation of a lubricating film on the friction surface of the TiAl matrix composites (Figs. 6-8 and Table 1). The thickness of the lubricating film is about 8 μm (Layer 1 and Layer 2 in Fig. 8). There is a mechanical mixture layer with a thickness of about 18 μm connecting the lubricating film and the TiAl matrix composite surface after friction test both at room temperature and 550°C (Layer 3 in Fig. 8). The mechanical mixture layer is mainly composed of Ti_3SiC_2 and TiAl, which provide Ti, Al, Si that form the lubricating film and prevent the lubrication film from falling off (Fig. 8). The C content distributed in the mechanical mixture layer is higher than that in the TiAl matrix, implying that the lubricating film of the TiAl matrix composites is formed by squeezing the Ti_3SiC_2 additive (Fig. 8). In addition, the plastic flow of the Ti_3SiC_2 lubricant on the friction surface of the TiAl matrix composites also contributes to the formation of the lubricating film. Although the friction test was conducted at room temperature, the instantaneous temperature between the two relatively moving composites in direct contact may reach 3000°C during the friction process [30–32]. Consequently, the lubricating film of the TiAl matrix composites with the added Ti_3SiC_2 originates from the squeezing and plastic flow of Ti_3SiC_2 both at room temperature and 550°C.

The friction coefficient and wear rate values of the TiAl composites at 550°C decrease by about 16.6 and 86.6%, respectively, compared with those at room temperature. The groove and peeling appear on the friction surface of the TiAl matrix composites at room temperature, while the abrasive debris and plastic deformation exist at 550°C (Fig. 6). The wear mechanisms of the TiAl matrix composites at room temperature and 550°C mainly belong to abrasive wear and adhesive wear, respectively. The difference in tribological properties of the TiAl matrix composites at room temperature and 550°C is attributed to the composition of lubricating film on the friction surface of the TiAl matrix composites. The Fe, O, Si, and C contents are about 0.10, 3.62, 3.65, and 6.91%, respectively, at the friction surface of the TiAl matrix composites at room temperature (Figs. 6a-d and Table 1). However, the Fe and O contents on the friction surface of TiAl matrix composite after friction test at 550°C are 639.50 and 9145.45% higher than those at room temperature, respectively, while the Si and C contents are 72.33 and 41.10% lower than those at 550°C, respectively (Figs. 6a-h and Table 1). In addition, the contents of Si and C at the friction surface of the TiAl matrix composite increase at higher temperatures with the Ti_3SiC_2 amount. Fe_2O_3 , Fe_3O_4 , $\text{Fe}_3\text{Ti}_{0.5}\text{O}_{0.3}$, Fe_3Al , and Ti_3SiC_2 appear in the wear debris of the TiAl matrix composites at 550°C (Fig. 7c). According to the analysis of wear debris and elemental distribution of the friction surface of the TiAl matrix, the Ti_3SiC_2 lubricating film forms on the friction surface of the TiAl matrix composite at room temperature, while the Fe-Ti-Al-Si-oxides lubricating film forms at 550°C. The variation in the Fe and O contents exhibits similar trends, opposite to the variation trend of Ti, Al, and Si contents (Figs. 7a-b). It is inferred that the Fe-Ti-Al-Si-oxide lubricating film consists of a Fe-oxide layer and a Ti-Al-Si-oxide layer at

550°C (Layer 1 and Layer 2 Fig. 8), while the Ti_3SiC_2 lubricating film has only a Ti_3SiC_2 lubricating layer at room temperature. The Fe-oxide layer is located at the top of the Fe-Ti-Al-Si-oxide lubricating film (Fig. 8). Fe_2O_3 and Fe_3O_4 exist in wear debris at 550°C, indicating that the Fe-oxide layer on the friction surface of the TiAl matrix composites and the lubricating layer of the counterpart are mainly composed of Fe_2O_3 and Fe_3O_4 (Fig. 7c). In addition, Ti-Al-Si-oxides do not appear in wear debris. The wear of the TiAl matrix composites at 550°C is concentrated in the Fe-oxide layer rather than in the whole lubricating film like that at room temperature, which is consistent with the results of lower friction coefficient and wear rate of the TiAl matrix composites at 550°C than at room temperature. Thus, the phase composition of the lubricating film contributes to better tribological properties of the TiAl matrix composites at 550°C than at room temperature.

The variation of the surface characteristics of the TiAl matrix composites shows three stages with the Ti_3SiC_2 amount (Fig. 6). As the Ti_3SiC_2 amount increases from 0 to 10 wt.%, the variation trend of groove and plastic deformation on the surface of the TiAl matrix composites increases, while the variation trend of peeling and abrasive debris decreases.

As the Ti_3SiC_2 amount increases from 10 to 20 wt.%, the variation trend of groove, peeling, abrasive debris, and plastic deformation decreases. When the Ti_3SiC_2 amount is between 20 and 30 wt.%, the variation trend of groove and peeling increases, while the variation trend of abrasive debris and plastic deformation decreases. On the one hand, the Si and C contents on the friction surface of the TiAl matrix composites after friction test at room temperature increase with the Ti_3SiC_2 amount (Fig. 6 and Table. 1). On the other hand, although the Si and C contents exhibit no distinct comparison trend after friction test at 550°C, the Fe, Ti, Al, O, Si contents exhibit similar values at different regions in the same composite on the friction surface after friction test at 550°C with the Ti_3SiC_2 amount (Fig. 6 and Table 1).

It can be concluded that a higher amount of added Ti_3SiC_2 has a beneficial effect on the integrity of the lubricant film of the TiAl matrix composites on the friction surface both at room temperature and 550°C, decreasing the friction coefficient and wear rate. The peeling region has the lowest content of Si and C compared with other regions at room temperature, indicating that the thin region of the Ti_3SiC_2 lubricating film tends to form peeling (Figs. 6a-d and Table 1). The plastic deformation region has the highest Cr content and the lowest O content compared with other regions at 550°C (Figs. 6e-f and Table 1), indicating that the plastic deformation is liable to form at the thin region of the Fe-Ti-Al-Si-oxide lubricating film. As the Ti_3SiC_2 amount increases from 0 to 10 wt.%, the Ti_3SiC_2 content in the TiAl matrix composite is only 0 - 9% (Figs. 1c-d), so the Ti_3SiC_2 lubricant on the surface of the TiAl matrix composite is in the form of discontinuous islands. Although the integrity of the lubricating film increases with the amount of added Ti_3SiC_2 , the amount of isolated Ti_3SiC_2 lubricant on the friction surface increases, decreasing the variation trend of peeling and abrasive debris. As the Ti_3SiC_2 amount increases from 10 to 20 wt.%, the Ti_3SiC_2 content (Figs. 1c-d) is high enough, 9-17.9%, to form continuous Ti_3SiC_2 lubricant on the TiAl matrix composites' surface. Thus, the peeling and plastic deformation on the friction surface of

the TiAl matrix composites increase. When the Ti_3SiC_2 amount is from 20 to 30 wt.%, Ti_3SiC_2 gradually agglomerates (Fig. 2). It induces detaching of the Ti_3SiC_2 lubricant, enhancing the peeling region on the friction surface of the TiAl matrix composite (Figs. 6a-d and Table 1). However, the Ti_3SiC_2 agglomeration has little influence on the formation of plastic deformation on the friction surface at 550°C. It is attributed to the phase composition of the Fe-Ti-Al-Si-oxide film and is affected by the double oxide layer of Fe-Ti-Al-Si-oxides film (Figs. 6e-f and Fig. 8). Consequently, the peeling on the friction surface of the TiAl matrix composites increases with the Ti_3SiC_2 amount (20-30 wt.%), while the plastic deformation decreases. Fe exists on the groove, while other regions do not exhibit the Fe presence at room temperature (Figs. 6a-d and Table 1). Moreover, the variation trend of groove depth is consistent with the value variation trend of the hardness of the TiAl matrix composites. Thus, the hard phase of the counterpart sinks into the friction surface due to the applied load and then, as the counterpart moves along the surface, forms a groove on the friction surface at room temperature [33]. However, the groove does not appear on the friction surface at 550°C since the direct friction between the TiAl matrix composites and the counterpart is transformed into the friction between the Ti-Al-Si-oxide film of the TiAl matrix composites and the oxide film of the counterpart. The abrasive debris region has the lowest Ti, Al, and Si contents compared with other regions at 550°C, while the O and Fe contents are higher than in the other regions (Figs. 6e-h and Figs. 7a-b). In addition, the Fe content in the abrasive debris region decreases with the Ti_3SiC_2 amount. It can be inferred that the abrasive debris tends to form at the thick region of the Fe-oxide layer located at the top of the Fe-Ti-Al-Si-oxide film, which can be inhibited by the increase in the amount of added Ti_3SiC_2 (Figs. 6e-h and marked by Layer 1 in Fig. 8).

Table 1
EDS elemental analysis (wt.%) of the regions marked in Fig. 6.

EDS measured points	Ti	Al	O	Fe	Cr	Si	C
D	62.41	31.73	4.42	0.2	1.24	–	–
E	61.91	32.38	4.16	–	1.55	–	–
F	62.72	31.78	3.78	0.11	1.61	–	–
G	58.37	29.41	3.65	0.05	1.93	2.47	4.12
H	60.28	30.16	4.13	–	1.90	1.14	2.39
I	58.68	29.39	3.66	–	1.88	2.38	4.01
J	58.09	29.63	3.52	–	0.98	2.00	5.78
K	55.07	29.23	3.24	0.02	1.21	4.55	6.68
L	55.38	28.62	3.18	–	0.89	4.52	7.41
M	55.58	28.49	3.29	–	0.87	4.18	7.59
N	51.85	26.29	3.24	0.13	0.92	5.73	11.84
O	49.68	28.02	3.11	0.1	0.85	5.89	12.35
P	46.41	18.78	25.5	8.93	0.38	–	–
Q	12.18	10.12	37.44	40.13	0.13	–	–
R	40.63	24.51	25.01	9.37	0.48	–	–
S	42.24	22.04	22.4	8.57	0.34	0.83	3.58
T	39.61	24.83	21.46	8.74	0.51	0.91	3.94
U	22.81	15.13	30.46	29.73	0.4	0.34	1.13
V	40.21	20.61	23.7	8.51	1.08	1.27	4.62
W	32.02	18.06	30.74	15.38	0.22	0.48	3.10
X	42.38	15.49	28.31	8.11	0.25	1.39	4.07
Y	44.07	22.34	20.3	6.51	1.15	1.43	4.20
Z	37.38	14.06	29.16	8.05	2.02	1.45	7.88

As mentioned above, the mechanical and tribological properties of the TiAl matrix composites improve with the Ti_3SiC_2 amount. The change in the density, hardness, and compression strength values with the amount of added Ti_3SiC_2 is achieved by the joint contribution of three factors. Firstly, the addition of Ti_3SiC_2 introduces high density and hardness Ti_3SiC_2 (4.53 g/cm³ and 4 GPa), TiC (4.93 g/cm³ and 28.5 GPa), and Ti_5Si_3 (4.32 g/cm³ and 9.5 GPa) phases (Fig. 1c) into the TiAl matrix composites, yielding the

increase of bulk density and hardness of the TiAl matrix composites. Secondly, Ti_5Si_3 and TiC formed at the interface of the TiAl matrix and Ti_3SiC_2 induce a pinning effect (Fig. 4b), which is conducive to the stress transfer from the TiAl matrix to Ti_3SiC_2 with higher pressure, and it also impedes grain boundary sliding and increases the resistance to dislocation movement, ultimately increasing the hardness and compression strength. Finally, the Ti_3SiC_2 agglomeration is especially aggravated when the Ti_3SiC_2 amount exceeds 20 wt.% (Fig. 2). It leads to the stress concentration and the increase of number of pores, resulting in a notable decrease of relative density, bulk density, and hardness (Figs. 5a-c). However, the Ti_3SiC_2 agglomeration has little effect on the compression strength with 0-30 wt.% of added Ti_3SiC_2 . The compression strength is mainly affected by the increase in the relative content of Ti_3SiC_2 , TiC, and Ti_5Si_3 hard phases. Although these factors also affect the trends of friction coefficient and wear rate, they are mediated by the lubricant film formed on the friction surface of the TiAl matrix composites. The composition and integrity of the lubricating film play important roles in influencing the tribological properties of the TiAl matrix composites. The composition of the lubricating film is determined by the friction temperature and the composition of the TiAl matrix composites, resulting in different tribological properties after friction tests at room temperature and 550°C. Ti_3SiC_2 in the TiAl matrix composites gradually accumulates on the composite's surface during the friction process due to the squeezing and plastic flow of Ti_3SiC_2 , and then it forms a Ti_3SiC_2 lubricating film at room temperature or a Fe-Ti-Al-Si-oxide film at 550°C (Figs. 6-8). The direct friction between the Ti_3SiC_2 lubricating film of the TiAl matrix composites and the counterpart at room temperature is transformed to the friction between the Ti-Al-Si-oxides film of the TiAl matrix composites and the oxide film of the counterpart. In addition, the wear of the TiAl matrix composites at 550°C is mainly concentrated on the Fe-oxide layer of the Fe-Ti-Al-Si-oxide film rather than on the whole lubricating film at room temperature during the friction process (Figs. 6-8). Thus, the Fe-Ti-Al-Si-oxide film yields better tribological properties of the TiAl matrix composites than the Ti_3SiC_2 lubricating film (Figs. 5-6). The integrity of the lubricating film is affected by the synergy of the Ti_3SiC_2 content in the TiAl matrix composites and the interfacial bonding between Ti_3SiC_2 and the TiAl matrix. The more compact the lubricating film, the better tribological properties of the TiAl matrix composites. The increase of the Ti_3SiC_2 content in the TiAl matrix yields a higher content of the Ti_3SiC_2 lubricant on the surface of the TiAl matrix composites, resulting in better integrity of the lubricating film (Figs. 1c-d and Figs. 5d-e). However, in excess of Ti_3SiC_2 (20-30 wt.%) in the TiAl matrix, the Ti_3SiC_2 agglomeration is aggravated, decreasing the interfacial bonding between Ti_3SiC_2 and the TiAl matrix (Figs. 1c-d and Fig. 2-4). The decrease in the interfacial bonding strength between Ti_3SiC_2 and the TiAl matrix leads to higher consumption than the production of the lubricating film on the friction surface of the TiAl matrix composites, decreasing the integrity of the lubricating film. This is why the decreasing trends of the TiAl matrix's friction coefficient and wear rate moderate when the relative Ti_3SiC_2 content in the TiAl matrix composites increases from 17.9 to 26.7% (Figs. 1c-d and Figs. 5d-e). Moreover, an unprecedented low wear rate of the TiAl matrix composites at 550°C compared with previous studies (Fig. 5f) is attributed to the Ti_3SiC_2 decomposition. The formation of TiC and Ti_5Si_3 hard phases effectively pin Ti_3SiC_2 so that it prevents falling off and improves the integrity of the lubricating film [34].

4 Conclusions

The TiAl matrix composites with the Ti_3SiC_2 addition were composed of TiAl, Ti_3Al , Ti_3SiC_2 , Ti_5Si_3 , and TiC synthesized via SPS at 1100°C for 10 min. With the increase of the Ti_3SiC_2 amount, the relative contents of TiAl and Ti_3Al decreased, while the relative contents of Ti_3SiC_2 , TiC, and Ti_5Si_3 increased. Ti_3SiC_2 gradually agglomerated with the Ti_3SiC_2 amount. The mechanical and tribological properties of the TiAl matrix composites improved with the Ti_3SiC_2 amount. They exhibited a lower friction coefficient and wear rate at 550°C than at room temperature. A Ti_3SiC_2 lubricating film was formed on the friction surface of the TiAl matrix composites after friction test at room temperature, while a Fe-Ti-Al-Si-oxides film was formed after friction test at 550°C . With 30 wt.% of added Ti_3SiC_2 , the TiAl matrix composites demonstrated the optimal comprehensive properties, having the hardness of 5.1 GPa. The compression strength, friction coefficient, and wear rate at room temperature and 550°C were 640 and 1080 MPa, 0.423 and 0.507, $0.21 \cdot 10^{-4}$ and $1.35 \cdot 10^{-4} \text{ mm}^3\text{N}^{-1}\text{m}^{-1}$, respectively.

This work provides a theoretical basis for the research of other lubricating materials and also expands the tribological applications of the TiAl matrix composites under severe environments.

Declarations

Acknowledgments

This work was supported by the Key Program of Scientific Research of Higher Education of Hebei Province (Grant No. ZD2021099).

Conflict of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

References

1. Bewlay, B.P., Nag, S., Suzuki, A., Weimer, M.J.: TiAl alloys in commercial aircraft engines. *Mater. High Temp.* **33**, 549–559 (2016). <https://doi.org/10.1080/09603409.2016.1183068>
2. Cui, S., Cui, C.X., Xie, J.Q., Liu, S.J., Shi, J.J.: Carbon fibers coated with graphene reinforced TiAl alloy composite with high strength and toughness[J]. *Sci. Rep.* **8**, 2364 (2018). <https://doi.org/10.1038/s41598-018-20799-y>
3. Pan, Y., Lu, X., Hayat, M., Yang, F., Liu, C.C., Li, Y., Li, X.Y., Xu, W., Qu, X.H.: P. Cao. Effect of Sn addition on the high-temperature oxidation behavior of high Nb-containing TiAl alloys. *Corros. Sci.* **116**, 108449 (2020). <https://doi.org/10.1016/j.corsci.2020.108449>
4. Tian, S.G., Lv, X.X., Yu, H.C., Wang, Q., Jiao, Z.H., Sun, H.F.: Creep Behavior and Deformation Feature of TiAl-Nb Alloy with Various States at High Temperature. *Materials Science & Engineering A.* **651**,

- 490–498 (2015). <http://dx.doi.org/10.1016/j.msea.2015.10.096>
5. Klocke, F., Herrig, T., Zeis, M., Klink, A.: Experimental Research on the Electrochemical Machinability of selected γ -TiAl alloys for the Manufacture of Future Aero Engine Components. *Procedia CIRP*. **35**, 50–54 (2015). <https://doi.org/10.1016/j.procir.2015.08.050>
 6. Jung, J.Y., Park, C., Bae, C., Effects of High-Response TiAl Turbine Wheel on Engine Performance under Transient Conditions. *JSAE*: 2015, 01: 1881. <https://doi.org/10.4271/2015-01-1881>
 7. Cheng, J., Li, F., Zhu, S.Y., Yu, Y., Qiao, Z.H., Yang, J.: Electrochemical corrosion and tribological evaluation of TiAl alloy for marine application. *Tribol. Int.* **115**, 483–492 (2017). <https://doi.org/10.1016/j.triboint.2017.06.027>
 8. Korotitskiy, A.V., Asnis, E.A., Piskun, N.V., Statkevich, I.I., Gorshenkov, M.V., Korotitski, A.V.: A promising microstructure/deformability adjustment of β -stabilized γ -TiAl intermetallics. *Mater. Lett.* **162**, 180–184 (2016). <https://doi.org/10.1016/j.matlet.2015.09.139>
 9. Yeh, C.L., Sun, W.E.: Use of TiH_2 as a reactant in combustion synthesis of porous Ti_5Si_3 and $\text{Ti}_5\text{Si}_3/\text{TiAl}$ intermetallics. *Journal of Alloys & Compounds*. **669**, 66–71 (2016). <https://doi.org/10.1016/j.jallcom.2016.01.236>
 10. Tian, S.W., Jiang, H.T., Guo, W.Q., Zhang, G.H., Zeng, S.W.: Hot deformation and dynamic recrystallization behavior of TiAl-based alloy. *Intermetallics*. **112**, 106521 (2019). <https://doi.org/10.1016/j.intermet.2019.106521>
 11. Sun, Y.J., Lin, Y.F., Zhang, N., Zhang, D.L.: Microstructures and mechanical properties of TiAl alloy fabricated by spark plasma sintering. *Int. J. Mod. Phys. B*. **34**, 2040036 (2020). <https://doi.org/10.1142/S0217979220400366>
 12. Xu, Z.S., Shi, X.L., Zhang, Q.X., Zhai, W.Z., Yao, J., Chen, L., Zhu, Q.S., Xiao, Y.C.: High-Temperature Tribological Performance of $\text{Ti}_3\text{SiC}_2/\text{TiAl}$ Self-Lubricating Composite Against Si_3N_4 in Air. *J. Mater. Eng. Perform.* **23**, 2255–2264 (2014). <https://doi.org/10.1007/s11665-014-0969-9>
 13. Xu, Z.S., Xue, B., Shi, X.L., Zhang, Q.X., Zhai, W.Z., Yao, J., Wang, Y.F.: Sliding Speed and Load Dependence of Tribological Properties of $\text{Ti}_3\text{SiC}_2/\text{TiAl}$ Composite. *Tribol. Trans.* **58**, 87–96 (2014). <https://doi.org/10.1080/10402004.2014.951748>
 14. Shen, Q., Shi, X.L., Zou, J.L., Yang, K., Huang, C., Zhang, A., Ibrahim, M.M., Wang, Y.F.: Tribological Performance and Self-Lubricating Film Formation Mechanism of TiAl-Based Composites at Elevated Temperatures. *Journal of Materials Engineering & Performance*. **26**, 268–276 (2017). <https://doi.org/10.1007/s11665-016-2433-5>
 15. Xu, Z.S., Shi, X.L., Wang, M., Zhai, W.Z., Yao, J., Song, S.Y., Zhang, Q.X.: Effect of Ag and Ti_3SiC_2 on Tribological Properties of TiAl Matrix Self-lubricating Composites at Room and Increased Temperatures. *Tribol Lett.* **53**, 617–629 (2014). <https://doi.org/10.1007/s11249-014-0299-y>
 16. Xu, Z.S., Zhang, Q.X., Shi, X.L., Zhai, W.Z., Yang, K.: Tribological Properties of TiAl Matrix Self-Lubricating Composites Containing Multilayer Graphene and Ti_3SiC_2 at High Temperatures. *Tribol. Trans.* **58**(6), 1131–1141 (2015). <https://doi.org/10.1080/10402004.2015.1046007>

17. Liang, B.Y., Xin, H., Zou, Q., Wang, M.Z.: Fabrication of SiC/Ti₃SiC₂/TiC Ceramic Composites by Pressureless Sintering of 3TiC/2.2 Si/0.2 Al. *Adv. Mater. Res.* **105**, 31–33 (2010). <https://doi.org/10.4028/www.scientific.net/AMR.105-106.31>
18. Rahmani, K., Majzoubi, G., Compos, J.: The effect of particle size on microstructure, relative density and indentation load of Mg-B₄C composites fabricated at different loading rates. *J. Compos. Mater.* **54**(17), 1–15 (2020). <https://doi.org/10.1177/0021998319896009>
19. Gürbüz, M., Mutuk, T., Uyan, P.: Wear and Thermal Behaviors of Graphene Reinforced Titanium Composites. *Met. Mater. Int.* **3**(18), 1–9 (2020). <https://doi.org/10.1007/s12540-020-00673-1>
20. Zhang, Z.F., Sun, Z.M., Hashimoto, H.: Rapid synthesis of ternary carbide Ti₃SiC₂ through pulse-discharge sintering technique from Ti/Si/TiC powders. *Metall. Mater. Trans. A.* **33**, 3321–3328 (2002). <https://doi.org/10.1007/s11661-002-0320-1>
21. Talero, R., Trusilewicz, L., Delgado, A., Pedrajas, C., Lannegrand, R., Rahhal, V., Mejía, R., Delvastoe, S., Ramírezf, F.A.: Comparative and semi-quantitative XRD analysis of Friedel's salt originating from pozzolan and Portland cement. *Constr. Build. Mater.* **25**, 2370–2380 (2011). <https://doi.org/10.1016/j.conbuildmat.2010.11.037>
22. El-Raghy, T., Barsoum, M.W., Sika, M.: Reaction of Al with Ti₃SiC₂ in the 800–1000°C temperature range. *Materials Science and Engineering: A.* **298**, 174–178 (2001). [https://doi.org/10.1016/S0921-5093\(00\)01281-8](https://doi.org/10.1016/S0921-5093(00)01281-8)
23. Gu, W.L., Yan, C.K., Zhou, Y.C.: Reactions between Al and Ti₃SiC₂ in temperature range of 600–650°C. *Scripta Mater.* **49**, 1075–1080 (2003). <https://doi.org/10.1016/j.scriptamat.2003.08.016>
24. Wang, Q., Chen, G.Q., Wang, K., Fu, X.S., Zhou, W.L.: Microstructural evolution and growth kinetics of interfacial compounds in TiAl/Ti₃SiC₂ diffusion bonding joints. *Materials Science & Engineering A.* **756**, 149–155 (2019). <https://doi.org/10.1016/j.msea.2019.04.034>
25. Dang, W.T., Ren, S.F., Zhou, J.S., Yu, Y.J., Wang, L.Q.: The tribological properties of Ti₃SiC₂/Cu/Al/SiC composite at elevated temperatures. *Tribol. Int.* **104**, 294–302 (2016). <https://doi.org/10.1016/j.triboint.2016.09.008>
26. Zuo, L.J., Ye, B., Feng, J., Zhang, H.X., Kong, X.Y., Jiang, H.Y.: Effect of ε-Al₃Ni phase on mechanical properties of Al–Si–Cu–Mg–Ni alloys at elevated temperature. *Materials Science & Engineering A.* **772**, 138794 (2020). <https://doi.org/10.1016/j.msea.2019.138794>
27. Shi, X.L., Yao, J., Xu, Z.S., Zhai, W.Z., Song, S.Y., Wang, M., Zhang, Q.X.: Tribological performance of TiAl matrix self-lubricating composites containing Ag, Ti₃SiC₂ and BaF₂/CaF₂ tested from room temperature to 600°C. *Materials & Design.* **53**, 620–633 (2014). <https://doi.org/10.1016/j.matdes.2013.07.053>
28. Xu, Z.S., Shi, X.L., Zhang, Q.X., Zhai, W.Z., Li, X.X., Yao, J., Song, S.Y.: Wear and Friction of TiAl Matrix Self-Lubricating Composites against Si₃N₄ in Air at Room and Elevated Temperatures. *Tribol. Trans.* **57**(6), 1017–1027 (2014). <https://doi.org/10.1080/10402004.2014.931497>

29. Zhai, W.Z., Shi, X.L., Wang, M., Xu, Z.S., Yao, J., Song, S.Y., Zhang, Q.X.: Friction and Wear Properties of TiAl-Ti₃SiC₂-MoS₂ Composites Prepared by Spark Plasma Sintering. *Tribol. Trans.* **57**(3), 416–424 (2014). <https://doi.org/10.1080/10402004.2014.880539>
30. Barsoum, M.W.: The M_{N+1}AX_N phases: A new class of solids: Thermodynamically stable nanolaminates. *Prog. Solid State Chem.* **28**, 201–281 (2000). [https://doi.org/10.1016/S0079-6786\(00\)00006-6](https://doi.org/10.1016/S0079-6786(00)00006-6)
31. Gupta, S., Barsoum, S.M.W.: On the tribology of the MAX phases and their composites during dry sliding: A review. *Wear.* **271**, 1878–1894 (2011). <https://doi.org/10.1016/j.wear.2011.01.043>
32. Kalin, M.: Influence of flash temperatures on the tribological behaviour in low-speed sliding: a review. *Materials Science & Engineering A.* **374**, 390–397 (2004). <https://doi.org/10.1016/j.msea.2004.03.031>
33. An, Q., Huang, L.J., Bao, T., Zhang, R., Jiang, S., Gen, L., Xiao, M.M.: Microstructure and tribological behavior of in situ synthesized (TiB+TiC)/Ti6Al4V (TiB/TiC=1/1) composites. *Tribol. Int.* **121**, 252–259 (2018). <https://doi.org/10.1016/j.triboint.2020.106177>
34. Yang, J., Gu, W., Pan, L.M., Song, K., Chen, X., Qiu, T.: Friction and wear properties of in situ (TiB₂ + TiC)/Ti₃SiC₂ composites. *Wear.* **271**, 2940–2946 (2011). <https://doi.org/10.1016/j.wear.2011.06.017>

Figures

Figure 1

(a) XRD pattern of Ti_3SiC_2 ; (b) FESEM cross-sectional micrograph of Ti_3SiC_2 ; (c) XRD patterns of the TiAl matrix composites; (d) relative content of each phase in the TiAl matrix composites.

Figure 2

Metallographic images of (a) TA0, (b) TA1, (c) TA2, and (d) TA3.

Figure 3

Elemental distribution of TA2.

Figure 4

Interfacial analysis of Ti_3SiC_2 and TiAl of TA2. (a) FESEM micrograph of TA2 and (b, c) EDS line-scanning analysis of the interface between Ti_3SiC_2 and the TiAl matrix in TA2.

Figure 5

Mechanical and tribological properties of the TiAl matrix composites: (a) density, (b) hardness, (c) compression strength, (d) friction coefficient, and (e) wear rate; (e) comparison of friction coefficient and wear rate between as-prepared Ti₃SiC₂-TiAl composites and those reported in the literature.

Figure 6

FESEM micrographs of the friction surface of (a, e) TA0, (b, f) TA1, (c, g) TA2, and (d, h) TA3 after frictional testing at (a-d) room temperature and (e-h) 550 °C.

Figure 7

Analysis of the friction surface and wear debris of TA2 after friction testing at 550 °C. (a) FESEM micrograph of the friction surface, (b) EDS line-scan elemental analysis at the plastic deformation region marked by Line 1 in Fig. 7(a), and (c) XRD pattern of wear debris of TA2 after friction test at 550 °C.

Figure 8

FESEM micrographs and elemental distributions of the TA3 cross-section.