Molecular dynamics simulations of the interdiffusion at α-Al_2O_3/AlSi12 interface

Masoud Tahani[1,2], Eligiusz Postek[1], Tomasz Sadowski[3]

[1] Institute of Fundamental Technological Research, Polish Academy of Sciences, Pawińskiego 5B, 02-106 Warsaw, Poland

[2] Department of Mechanical Engineering, Ferdowsi University of Mashhad, Mashhad, Iran

[3] Department of Solid Mechanics, Lublin University of Technology, 20-618 Lublin, Poland

mtahani@ippt.pan.pl, epostek@ippt.pan.pl, t.sadowski@pollub.pl

Keywords: Self-diffusion, Interdiffusion, Metal-ceramic composite, Al_2O_3/AlSi12 interface, Molecular dynamics method

1. Introduction

Metal matrix composites (MMC) are used more and more in the aerospace, automotive, and bio-medical industries because of their high strength-to-weight ratio, high stiffness, and outstanding wear resistance. Aluminium, titanium, and magnesium are the most preferred matrix materials, whereas alumina and silicon carbide are the most used reinforcing elements for these composites. The overall mechanical and failure properties of MMCs depend on the mechanical properties of the constituents and the nature of the interface. The characteristics of the interface must be understood because they have the potential to significantly alter the properties of MMCs. The interface between phases is a fuzzy region because of diffusion. To this end, it is necessary to look into the diffusion between the two phases as the first step for determining the cohesive zone model of the interface.

In this study, AlSi12 metal alloy as matrix material reinforced with α-Al_2O_3 is considered. AlSi12 is an aluminium alloy that contains 12 wt.% silicon with excellent thermal conductivity, good corrosion resistance, and low density. The composite can be used in various high-temperature applications such as furnace linings, engine parts, and aerospace components. It is worth noting that the properties and performance of the composite will depend on the processing conditions, microstructure, the proportion of the components, and the interface's characteristics.

The investigation carried out by Milas et al. [1] regarding the diffusion of Al, O, Pt, Hf, and Y atoms on α-Al_2O_3(0001) can be mentioned as an illustration of research that has been published in the literature. To the authors' knowledge, no studies have been done on α-Al_2O_3/AlSi12 diffusion couple. To this end, the self-diffusion and interdiffusion at the interface are investigated in this research by heating the system to the desired temperature. The effect of annealing temperature and annealing time are studied on the diffusion zone and interdiffusion coefficients. The thickness of the diffusion zone and the interdiffusion coefficients are found to increase as expected with increasing annealing temperature and time.

2. Model and simulation details

Diffusion is the movement of atoms or molecules from an area of higher concentration to an area of lower concentration. Diffusion can occur between the two phases, resulting in the migration

The publication is co-financed from the state budget under the programme of the Minister of Education and Science called "Excellent Science" project no. DNK/SP/548041/2022

Doskonała Nauka

Ministry of Education and Science Republic of Poland

Republic of Poland

of atoms between the two materials. The rate of diffusion between the aluminium oxide and AlSi12 alloy depends on several factors, including the temperature, the chemical composition of the two materials, and the interface between the two phases. At high temperatures, the diffusion rate will be faster, and the atoms will have more energy to move through the material.

The molecular dynamics (MD) method is able to study very basic processes like diffusion by using Newton's second law to describe atomic interactions. The MD simulations in this study are conducted using the open-source MD program LAMMPS and the OVITO software is utilized for visualizing the evolution of the atomic structure. The third-generation many-body COMB potential [2] and the Tersoff potential [3] are used in this research for the Al-O and Si-O interactions, respectively. Furthermore, The Morse potential with parameters $D_0 = 0.4824$ eV, $\alpha = 1.322$ Å^{-1}, and $r_0 = 2.92$ Å [4] is employed for the Al-Si interactions.

The initial α-Al_2O_3/AlSi12 interface is considered as a single crystal of AlSi12 and a single crystal of α-Al_2O_3 with an initial gap of 2 Å which is near the equilibration atomic distance in the interface. Both Al- and O-terminated interfaces are analysed. The typical dimension of the MD model is approximately 119×58×184 Å with a total of 109,986 atoms. The orientation relationship $(0001)[2\bar{1}\bar{1}0]_{\alpha\text{-Al2O3}} \| (111)[\bar{1}\bar{1}2]_{\text{AlSi12}}$ is considered. The lattice constant of fcc Al is 4.0495 and the lattice parameters of hexagonal α-Al_2O_3 are $a = b = 4.759$ Å, $c = 12.991$ Å, $\alpha = \beta = 90°$, and $\gamma = 120°$. 12 wt.% of Al atoms are replaced by Si atoms to produce AlSi12 single crystal.

The geometric configuration is optimized using the conjugate gradient energy minimization algorithm. The NVT canonical ensemble at a constant temperature of 300 K is imposed on the sample for 10 ps and then the NPT ensemble at a zero pressure and constant 300 K is used for 15 ps to adjust the volume and relax the assembled interface system. Then, the sample is heated to a preset temperature at a heating rate of 10 K/ps. Then, the temperature is maintained at the given temperature for 2 ns to study the interdiffusion and the movements of atoms are recorded during this period. The NPT ensemble with zero pressure with a 0.2 fs time-step is considered for all processes. The temperatures of 1500, 1800, and 2000 K are considered for the simulations.

3. Results and discussions

The elastic constants of α-Al_2O_3 and AlSi12 are obtained with the aforementioned potential functions and compared with the experimental and MD simulations in Table 1. It can be seen that the alumina results agree well with those obtained by other investigators. Next, each system is cut into thin slices of thickness 2 Å parallel to the interface plane and the number of each atom type is counted to obtain the concentration. Figures 1(a) and 1(b) illustrate the initial concentration profiles before diffusion and the concentration profiles after maintaining the system at 2000 K for 2 ns. The self-diffusion coefficients of each atom are determined from the slope of the mean square displacements. For example, the self-diffusion of atoms in an Al-terminated interface at 2000 K are $D_{\text{Al(AlSi12)}} = 53.346$, $D_{\text{Al(Al2O3)}} = 1.165$, $D_{\text{O}} = 0.0282$, $D_{\text{Si}} = 1.246$ (10^{-10} m^2/s).

Table 1. The elastic constants obtained by the present MD simulations and comparison with those obtained by other investigators.

Material	Method	C_{11} (GPa)	C_{12} (GPa)	C_{13} (GPa)	C_{33} (GPa)	C_{44} (GPa)	C_{66} (GPa)
α-Al_2O_3	Present	510	130	138	518	138	165
	Experiment [5]	497	164	111	498	147	167
	ES+ [6]	537	180	106	509	130	179
AlSi12	Present	268	134	154	214	108	105

The activation energy Q and pre-exponential factor D_0 of atoms can be obtained by fitting the self-diffusion coefficients to the Arrhenius equation:

$$D = D_0 \exp\left(-\frac{Q}{RT}\right) \tag{1}$$

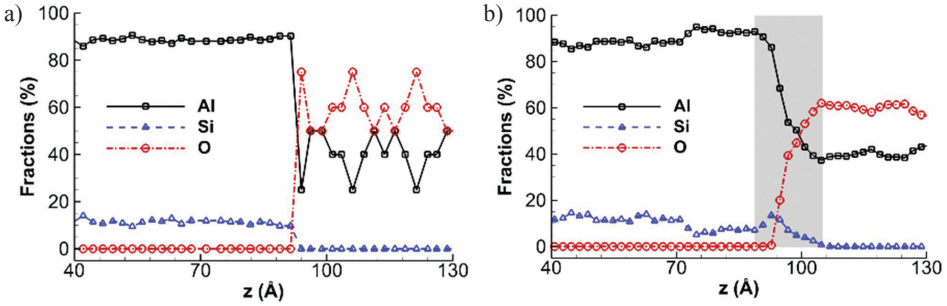

Figure 1. The concentration profiles of Al, Si, and O atoms along the z-direction during interdiffusion of the Al-terminated α-Al_2O_3/AlSi12 interface. (a) Initial system before relaxation and (b) after maintaining it at 2000 K for 2 ns. The grey region shows the diffusion zone.

A total of four independent interdiffusion coefficients are needed to describe the interdiffusion behavior in a ternary system. The interdiffusion coefficients are obtained by computing the interdiffusion flux of atoms using the present single diffusion couple. The Gaussian error function for each component is used to fit the concentration curve (e.g., see Ref. [7] for detail explanation about the method). The average values of the main and cross-interdiffusion coefficients of the Al- and O-terminated α-Al_2O_3/AlSi12 diffusion couple are calculated and listed in Table 2. The diffusion systems are maintained at the annealing temperatures of 1500 and 2000 K for 2 ns.

Table 2. Average ternary interdiffusion coefficients for the Al- and O-terminated α-$Al_2O_3$3/AlSi12 diffusion couple (10^{-11} m^2/s).

Diffusion couple	Temperature (K)	$\bar{\bar{D}}^{Al}_{OO}$	$\bar{\bar{D}}^{Al}_{OSi}$	$\bar{\bar{D}}^{Al}_{SiO}$	$\bar{\bar{D}}^{Al}_{SiSi}$
Al-terminatedα-$Al_2O_3$3/AlSi12	1500	0.584	$-2.6\cdot10^{-9}$	$-6.0\cdot10^{-6}$	3.728
	2000	0.911	$7.5\cdot10^{-8}$	$-3.5\cdot10^{-7}$	14.311
O-terminated α-$Al_2O_3$3/AlSi12	1500	0.575	$-1.3\cdot10^{-6}$	$-2.2\cdot10^{-6}$	3.648
	2000	0.681	$-4.8\cdot10^{-6}$	$1.4\cdot10^{-8}$	13.781

It is seen form Table 2 that the main interdiffusion coefficients increase with increasing the annealing temperature, as expected, and all cross-interdiffusion coefficients are at least five orders of magnitude smaller than the main interdiffusion coefficients. Furthermore, the results in this table indicate that the diffusivity of Si and O atoms is slightly higher for the Al-terminated system compared with the O-terminated one.

4. Conclusions

A molecular dynamics method was employed to investigate the atomistic evolutions during the interdiffusion at α-Al_2O_3/AlSi12 interface. The average main and cross interdiffusion coefficients were obtained using the concentration profiles of atoms during diffusion for the Al- and O-terminated systems. It was found that the interdiffusion coefficients increased with the increasing annealing

temperature and time. Moreover, there was no significant difference between Al- and O-terminated interfaces in terms of the interdiffusion.

References

1. Milas I., Hinnemann B., Carter E.A.: *Diffusion of Al, O, Pt, Hf, and Y atoms on α-Al_2O_3(0001): implications for the role of alloying elements in thermal barrier coatings*. Journal of Materials Chemistry, 21, 2011, 1447–1456.
2. Choudhary K., Liang T., Chernatynskiy A., Phillpot S.R., Sinnott S.B.: *Charge optimized many-body (COMB) potential for Al2O3 materials, interfaces, and nanostructures*. Journal of Physics: Condensed Matter, 27, 2015, 305004.
3. Munetoh S., Motooka T., Moriguchi K., Shintani A.: *Interatomic potential for Si-O systems using Tersoff parameterization*. Computational Materials Science, 39, 2007, 334–339.
4. Zhao H., Chen N.: *An inverse adhesion problem for extracting interfacial pair potentials for the Al(001)/3C-SiC(001) interface*. Inverse Problems, 24, 2008, 035019.
5. Wachtman J.B., Jr.: *Mechanical and thermal properties of ceramics*. National Bureau of Standards: Washington, DC, 1969, 139–188.
6. Streitz F.H., Mintmire J.W.: *Molecular dynamics simulations of elastic response and tensile failure of alumina*. Langmuir, 12, 1996, 4605–4609.
7. Tahani M., Postek E., Motevalizadeh L., Sadowsi T.: *Effect of vacancy defect content on the interdiffusion of cubic and hexagonal SiC/Al interfaces: A molecular dynamics study*. Molecules, 28, 2023, 744.

Acknowledgements. This research was funded by the Polish National Agency for Academic Exchange (NAWA) under grant number [BPN/ULM/2021/1/00115/U/DRAFT/00001]. We acknowledge the National Science Centre, Poland, grant No. UMO 2019/33/B/ST8/01263. The calculations were performed at PLGrid national facilities – Academic Computer Centre Cyfronet in Krakow, Academic Computer Centre in Gdańsk, and Interdisciplinary Centre for Mathematical and Computational Modelling, University of Warsaw, Poland.