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High-temperature wetting and interfacial interaction in AlB_{12}–Al system

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The article is dedicated to the study of wetting and specifics of interaction between phases on the "drop-substrate" interface in the AlB_{12}–Al system. Contact angles between an aluminum melt and an aluminum dodecaboride ceramic substrate were studied using the sessile drop method. It was found that upon aluminum melting at 660°C, contact angles are $\Theta > 90$ deg. Intensive spreading of the aluminum melt starts at 1200°C with contact angles $\Theta \approx 15$ deg. The X-ray spectral microanalysis showed that AlB_{12} grains recrystallized through the aluminum melt on the interface. In addition, due to the absence of active chemical interaction, the AlB_{12}–Al system can be recommended for the development of new composite materials.
Keywords: wetting, aluminum dodecaboride, aluminum melt, interphase interaction.

Високотемпературне змочування і міжфазна взаємодія у системі AlB_{12}–Al. *О.П.Уманський, М.С.Стороженко, В.Є.Шелудько, В.Б.Муратов, В.П.Красовський, В.П.Коновал, О.О.Васільєв, О.Є.Терентьев*

Досліджено змочуваність і особливості міжфазної взаємодії на межі розділу "крапля — підкладка" у системі AlB_{12}–Al. Для визначення контактних кутів використано метод "лежачої" краплі у вакуумі. Встановлено, що при температурі 660°C спостерігається плавлення алюмінію, при цьому контактні кути $\Theta > 90°$. Інтенсивне розтікання алюмінію починається при температурі $T = 1200$°C з утворенням контактних кутів $\Theta \approx 15°$. Методом мікрорентгеноспектрального аналізу встановлено, що на міжфазній межі відбувається перекристалізація зерен AlB_{12} у розплаві. При цьому система AlB_{12}–Al характеризується відсутністю активної хімічної взаємодії і може бути рекомендована для створення нових композиційних матеріалів.

Исследованы смачиваемость и особенности межфазного взаимодействия на границе раздела "капля — подложка" в системе AlB_{12}–Al. Для определения контактных углов использован метод "лежащей" капли в вакууме. Установлено, что при температуре 660°C наблюдается плавление алюминия, при этом контактные углы $\Theta > 90°$. Интенсивное растекание алюминия начинается при температуре $T = 1200$°C с образованием контактных углов $\Theta \approx 15°$. Методом микрорентгеноспектрального анализа установлено, что на межфазной границе происходит перекристаллизация зерен AlB_{12} в расплаве. При этом система AlB_{12}–Al характеризуется отсутствием активного химического взаимодействия и может быть рекомендована для создания новых композиционных материалов.

1. Introduction

Modern engineering requires new composite materials capable to ensure high reliability of machinery and equipment in extreme operating conditions such as elevated temperatures, shock load, aggressive chemical environments, and increased wear. Currently,

significant attention is paid to the development of metal-ceramic materials in which a combination of metal and ceramic components may lead to significantly improved performance (high hardness and strength, fracture toughness, wear resistance, etc.) [1–5].

Aluminum dodecaboride AlB_{12} is a promising material lightweight vehicles construction, since it has a low specific weight (2.55 g/cm^3) and high hardness (24 GPa) [6–8]. At the same time, high fragility of AlB_{12} significantly restricts its application [9]; therefore, it is reasonable to use it in a combination with plastic metallic components. Aluminum, having high plasticity, low specific weight (2.7 g/cm^3) and melting temperature (660°C) [10], is a promising matrix metal for composite materials based on aluminum dodecaboride.

There are several key conditions for the successful development of metal-ceramics materials with a high level of physical and chemical properties and high performance, namely:

— wetting contact angles on the "refractory ceramics – metal melt" interface should be close to zero;

— there should be no active chemical interaction between the components leading to the formation of new chemical compounds.

Previously, studies of wetting behavior in the AlB_{12}–Al system in [11] showed that intensive spreading of the aluminum melt over the AlB_{12} substrate begins not at the melting point of aluminum (660°C), but at temperatures above 1000°C. The authors explain this phenomenon by Al_2O_3 oxide film on the surface of the liquid metal which decomposes with increasing temperature, allowing the melt drop to spread. This work, however, does not consider the physical and chemical interactions between liquid and solid phases, which are necessary to predict the specifics of structure formation in AlB_{12}–Al composite materials. Thus, the objective of this work is to study the wetting behavior and the specifics of interphase interaction in the AlB_{12}–Al system using X-ray spectral microanalysis.

2. Methods and materials

The materials used in this study were high-purity Al (99.8 %) and AlB_{12} ceramics prepared by the CVD method at the Frantsevich Institute for Problems of Materials Science (Kyiv, Ukraine), according the reaction of hexagonal boron nitride with aluminum [12]:

$$12BN + 13Al \rightarrow AlB_{12} + 12AlN.$$

The average size of AlB_{12} powder particles was 150÷200 nm. The AlB_{12} solid ceramic specimens (10×10×2) mm^3 were sintered by the hot-pressing technique at 1850°C and applied pressure of 30 MPa for 30 min.

The experimentally measured density of the AlB_{12} ceramics was about 2.52 g/cm^3. The AlB_{12} substrates were grounded by the SiC P1200 sandpaper and subsequently polished using a different clothes and diamond suspensions to achieve a final surface roughness of R_a = 0.02 μm measured 3D profilometer "Optical Profiler Micron alpha" (National Aviation University, Kyiv). Directly before testing, the AlB_{12} substrates and Al pieces of about 0.4 g were mechanically cleaned and washed in an ultrasonic bath.

The structure of the AlB_{12} ceramic substrate was studied using a JEOL-9500FS (SEM) Auger microanalyzer with equipped energy dispersive spectroscopy (EDS) analysis. The structure of the ceramic substrate contains white

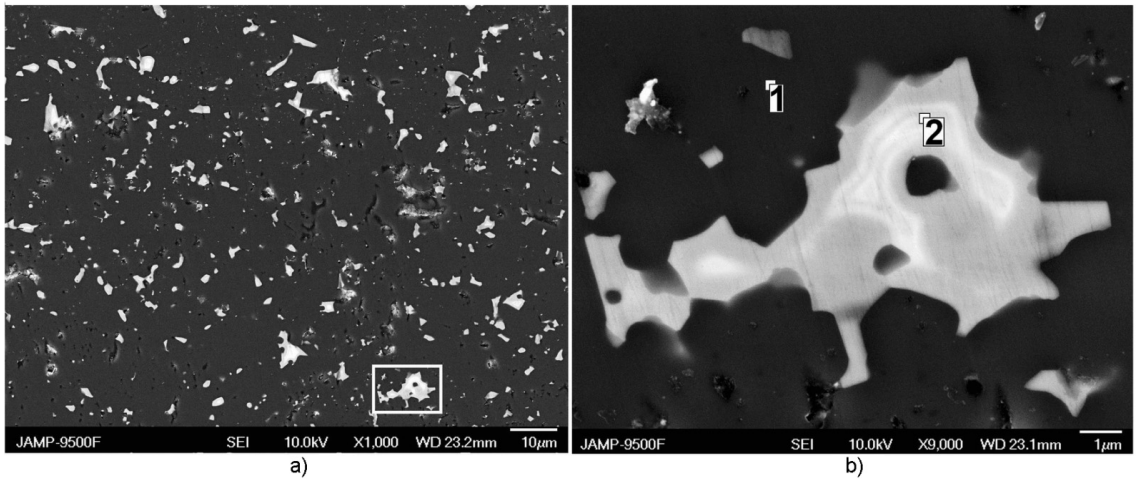

Fig. 1. Structure of hot-pressed solid AlB_{12} ceramic substrate: a — general view; b — detailed region with elemental analysis.

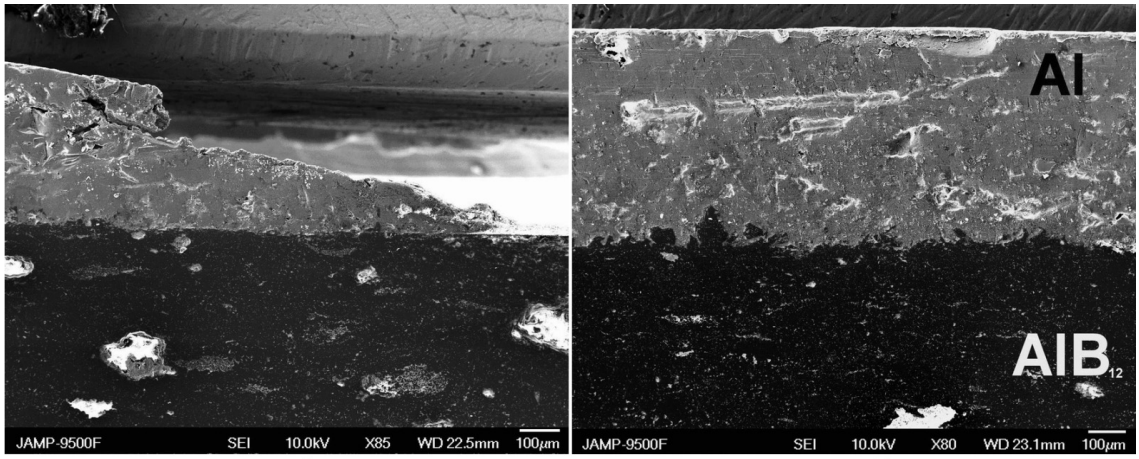

Fig. 2. Microstructure of a cross-section of the AlB_{12}–Al couple showing: a — Al drop on the AlB_{12} substrate; b — general view of the couple AlB_{12}–Al.

Table 1. Chemical composition of phases in the AlB_{12} ceramic substrate (Fig. 1) according to EDS analysis

Spectrum	Elemental content, wt.%			Total
	B	O	Al	
1	83.35	–	16.65	100
2	0.86	45.99	53.15	100

precipitates of about 8–10 μm in size uniformly distributed in the black-colored matrix (Fig. 1). According to the EDS analysis, the black matrix represents the AlB_{12} phase (Fig. 1, Table 1, point 1). The composition of white grains is, on average, 53 wt % Al, 46 wt. % O (Fig. 1, Table 1, point 2). It is reasonable to assume that the Al_2O_3 phase has been formed during hot-pressing the AlB_{12} powder due to the reaction of aluminum with oxygen in air.

The sessile drop method was applied for the investigation of high-temperature wetting and interaction between the AlB_{12} substrate and liquid Al. The alloy-ceramics couple was introduced into the resistance furnace chamber that was pumped out to a vacuum of $1.33 \cdot 10^{-4}$ Pa and then heated using tungsten elements. The temperature in the vacuum chamber was measured by a thermocouple (TPP0679 type). The specimens were maintained at 1200°C for 30 min and then cooled inside the vacuum chamber until the temperature dropped down to 20°C in order to avoid oxidation. The behavior of the Al drop on the AlB_{12} ceramic substrate was registered using a high-resolution digital camera. The contact angles were determined on the left and right sides of the drop. The uncertainty of the averaged contact angle was about ±5 deg.

The structural and chemical characteristics of the solidified couple were determined by the cross-sectional method using a JEOL-9500FS scanning electron microscope (SEM) equipped with an energy-dispersive X-ray spectroscopy (EDS) analyzer.

3. Results and discussion

In the process of wetting, pure Al begins to deform at temperatures around 650–700°C, forming a drop of an irregular shape, which transforms into a sphere upon gradual temperature increasing up to 1000°C. In these temperature ranges, the contact angles remain above 90 deg. This fact can be explained by the presence of an Al_2O_3 oxide film on the drop surface, which prevents the spreading of the aluminum melt. Upon further heating to 1150–1200°C, this film is destroyed, and the melt begins to spread over the solid surface of the ceramics. The final contact angle $\Theta \approx 15$ degrees (Fig. 2a) is formed after 2–3 min of exposure at 1200°C, in accordance with [11].

The contact zone between the liquid and solid phases was studied to determine the specifics of the interphase interaction in the AlB_{12}–Al system. The system is characterized by the absence of a transition zone, but metallographic studies showed a change in the configuration of the profile of the contact interface between the AlB_{12} substrate and the Al drop (Fig. 2b, 3a).

Nevertheless, there is no active chemical interaction with the formation of new compounds between the melt and the substrate is confirmed by the distribution of elements in the characteristic spectra shown in Fig. 3. The microprobe analysis of the AlB_{12}–Al interface showed three main coexisting phases: aluminum,

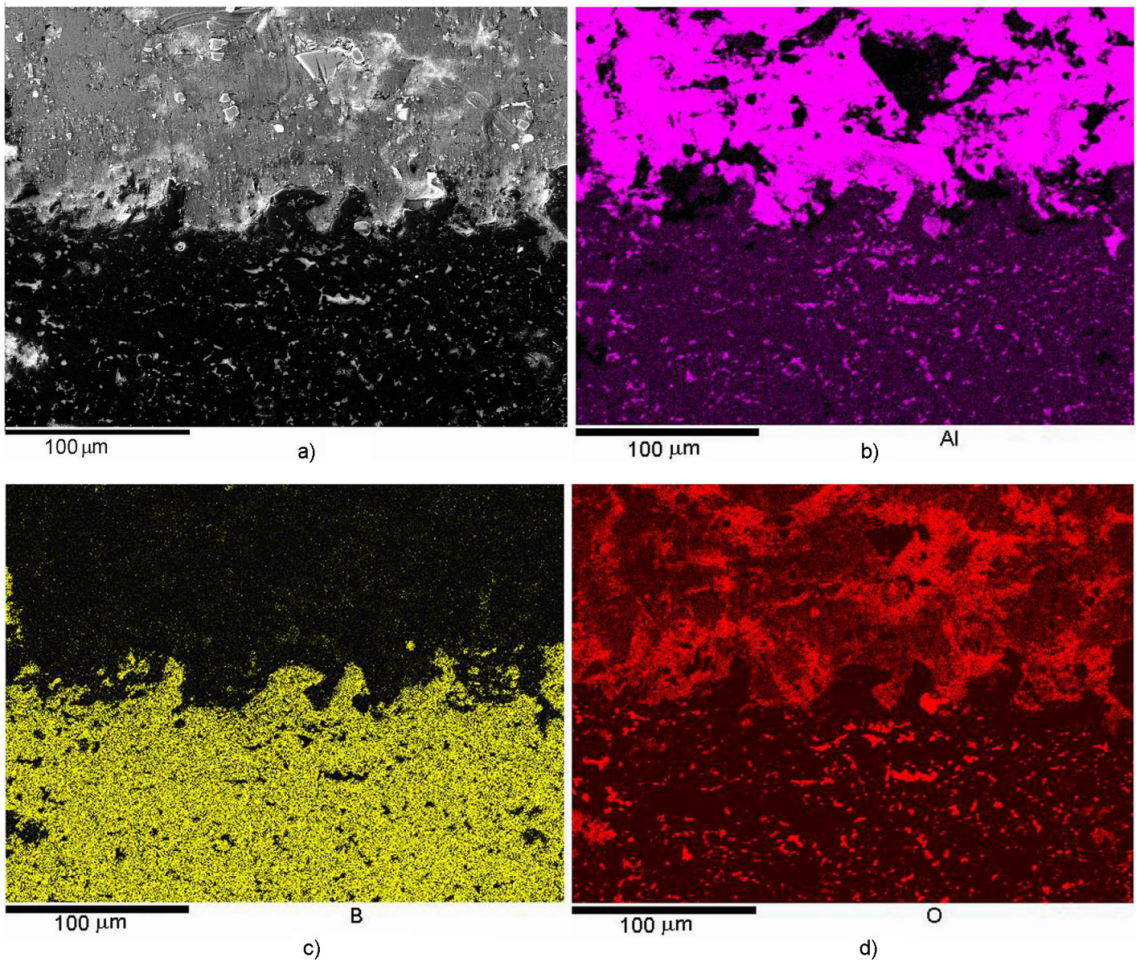

Fig. 3. BSE image of microstructure of an AlB_{12}–Al cross-section (a) and EDS elemental maps showing Al (b), B (c), O (d) distribution within the interface region.

aluminum dodecaboride, and alumina (Fig. 4). As was shown before, the ceramic substrate AlB_{12} contains the Al_2O_3 phase due to the technological specifics the hot pressing process in graphite molds in air of aluminum dodecaboride.

However, the droplet zone along the interface contained AlB_{12} grains devoid of Al_2O_3 inclusions (Fig. 5, 1–3, Table 2). It can be assumed that in the zone of contact between the liquid and solid phases, the components of the substrate dissolve in molten aluminum with subsequent recrystallization and growth of AlB_{12} grains at the interface. As a result, the interface configuration becomes more developed, increasing contact area between the metal and ceramic phases and, as a consequence, the work of adhesion in the AlB_{12}–Al system.

Thus, the results of this work have demonstrated that aluminum satisfies the requirements to the metal matrix and can be recommended for the development of new composite materials based on aluminum dodecaboride.

4. Conclusions

Thus, studies of the features of wetting and contact interaction in the AlB_{12}–Al system lead to the following conclusions:

Liquid Al wets the AlB_{12} substrate at temperatures of 1100–1250°C, which is significantly higher than aluminum melting temperature (660°C), forming the contact angle $\Theta \approx 15$ deg.

The interaction zone in the AlB_{12}–Al system is characterized by the formation of aluminum dodecaboride grains due to recrystallization through the liquid phase. This can be regarded as a positive factor, since it leads to an increase in the contact area of the liquid and solid phases and an increase in the work of adhesion in the system "refractory ceramics — metal melt".

The AlB_{12}–Al system does not demonstrate any active chemical interaction between the components of the solid and liquid phases. Aluminum may be recommended as a metal matrix for composite materials based on aluminum dodecaboride.

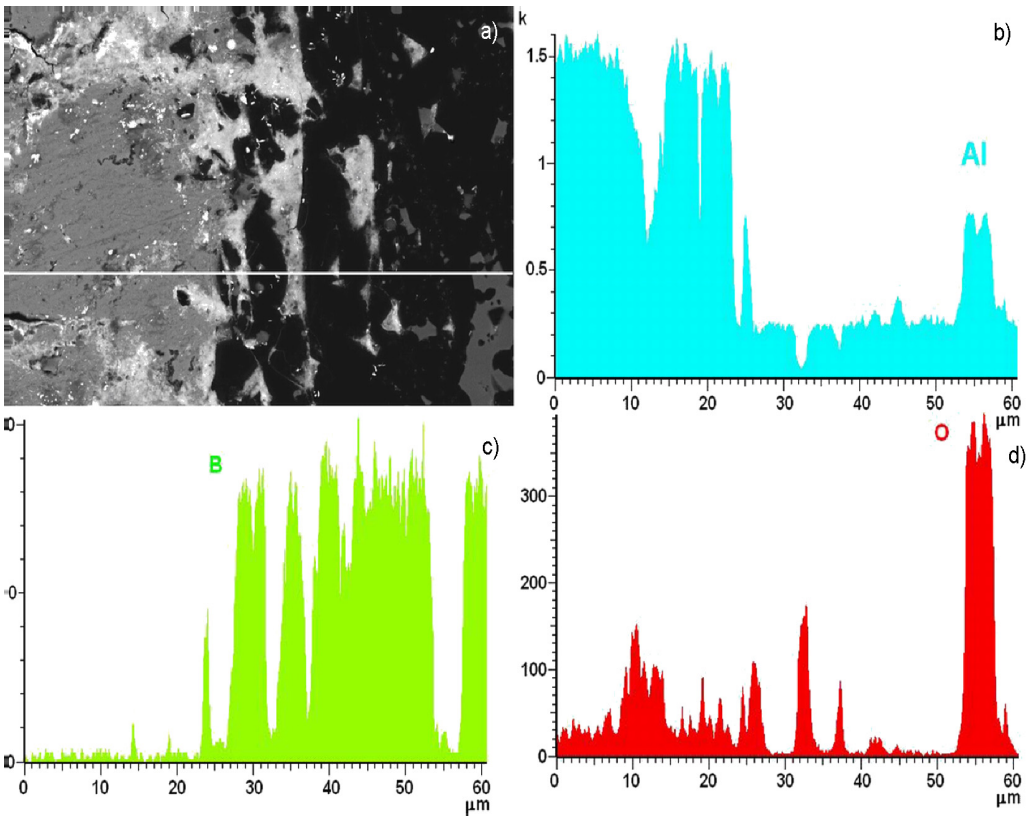

Fig. 4. Microstructure of an AlB_{12}–Al interface cross-section with scanning line (a) and distribution of Al (b), B (c), O (d) within the interface region in the AlB_{12}–Al system.

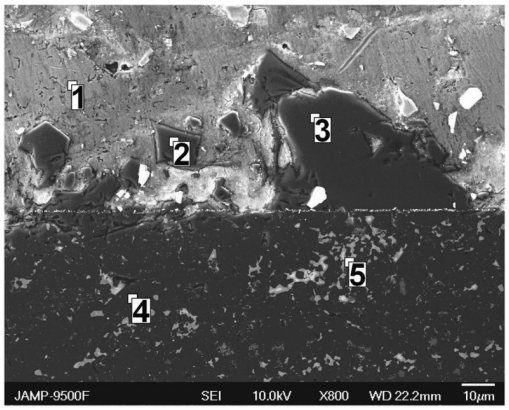

Fig. 5. Microstructure of an AlB_{12}–Al interface cross-section with elemental analysis in separate points.

Table 2. Microprobe analysis of AlB_{12}–Al interface (Fig. 5)

Spectrum	Elemental composition, wt. %			Total
	B	O	Al	
1	–	3.83	96.17	100
2	82.90	0.41	16.69	100
3	82.75	0.38	16.87	100
4	83.12	0.26	16.62	100
5	–	48.66	51.34	100

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