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ФИЗИКО-ТЕХНИЧЕСКИЕ ОСНОВЫ ЭКСПЕРИМЕНТА И ДИАГНОСТИКИ

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Acoustical Investigation of Adhesion in Liquid Metal–Ceramic Interfaces

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In this paper, we briefly review and analyse results of an acoustical investigation of adhesion in metal–ceramic interfaces based on the determination of the slope parameter, which is defined by the linear correlation between the work of adhesion of various liquid metal–ceramic systems and the sound propagation velocity of plate acoustical wave in the corresponding metals. The dependence of values of the work of adhesion slope parameter for several ceramic materials on the acoustic impedances of the corresponding ceramics is examined. The obtained results permit the interpretation of the wave propagation nature in these interfaces according to the existence and the excess of the interfacial bonding.

Key words: interface, adhesion, sound velocity, ceramics, liquid metal.

У статті коротко оглядаються й аналізуються результати акустичного дослідження адгезії в метал-керамічних інтерфейсах на основі визначення танґенса кута нахилу лінійної кореляції між роботою адгезії для різних систем рідкий метал–кераміка та швидкістю поширення поверхневої акустичної хвилі у відповідних металах. Розглядається залежність значень танґенса кута нахилу роботи адгезії для деяких керамічних матеріялів від акустичних імпедансів відповідних керамік. Одержані результати уможливлюють інтерпретувати характер поширення хвиль у цих інтерфейсах, відповідно до наявности та величини міжповерхневого зчеплення.

Ключові слова: поверхня поділу, адгезія, швидкість звуку, кераміка, рі-

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дкий метал.

В статье сделаны краткий обзор и анализ результатов акустического исследования адгезии в металл-керамических интерфейсах на основе определения тангенса угла наклона линейной корреляции между работой адгезии для различных систем жидкий металл–керамика и скоростью распространения поверхностной акустической волны в соответствующих металлах. Рассматривается зависимость значений тангенса угла наклона работы адгезии для некоторых керамических материалов от акустических импедансов соответствующих керамик. Полученные результаты позволяют интерпретировать характер распространения волн в этих интерфейсах в соответствии с наличием и величиной межповерхностного сцепления.

Ключевые слова: поверхность раздела, адгезия, скорость звука, керамика, жидкий металл.

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1. INTRODUCTION

Metalized ceramic are playing a major role in several modern applications [1] such as metal–ceramic joining, metal-matrix composites, thin metal films on ceramic substrates [2], thermal-barrier coatings (TBC) [3], hard TiN coating [4], photovoltaic materials [5] and as functional components in microelectronics [6]. The performance is directly related to the nature of the metal–ceramic interfaces.

The most important characteristics of these materials are their high-impact energy-absorption capacity, dimensional stability, thermal and electrical conductivity, low and controllable density and the large internal surface area. However, they could undergo a severe problem consisting of poor adhesion at metal/ceramic interfaces. This is why an understanding of the adhesion mechanism is needed in order to control the nature of the interfacial bonding and the determination of the reversible work necessary to damage these interfacial bonds [7–12].

Furthermore, the optimization of metal–ceramic interfacial adhesion by non-destructive techniques is crucial to the applications of these materials. In this context, various ultrasonic methods are established for the characterization of the metal–ceramic interfaces [13].

In the liquid metals, the sound propagation is done transversely with a characteristic velocity; there is no matter transfer of but only energy transfer [14, 15]. This acoustic wave does not depend only on the elastic properties of this liquid metal, but it is strongly affected by the properties of the interface with the ceramic substrate. The weakly or strongly adherent regions have different responses. This means that a change in the properties of the adhesion must result in a change of the velocity of the surface waves in the ceramic [15].

In this paper, a new acoustic approach is suggested to interpret the interfacial adhesion in the non-reactive metal–ceramic systems. Discussions will be made on the relation between sound propagation and the nature of bonding of metals with several ceramic materials.

2. METHODOLOGY

The adhesion of the metal–ceramic system is the most important factor of all metal bonds. It is determined by the change in the free energies of two materials when they come into contact [16] (Fig. 1).

The work of adhesion W_{ad} between the liquid metal and the ceramic can be expressed using the Young–Dupré equation relating the surface tension of the liquid metal above the melting temperature γ_{LV} and the measured equilibrium contact angle θ formed by the metal on the ceramic substrate [17]:

$$W_{\mathrm{ad}} = \gamma_{\mathrm{LV}}(1 + \cos\theta). \tag{1}$$

The work of adhesion W_{ad} in metal–ceramic contact is generally written as the sum of different contributions of the interfacial interactions between two phases [17]:

$$W_{\mathrm{ad}} = W_{\mathrm{equil}} + W_{\mathrm{non\text{-}equil}}. \tag{2}$$

$W_{\mathrm{non\text{-}equil}}$ represents the non-equilibrium contribution to the work of adhesion. In the absence of chemical reactions, this term does not appear. W_{equil} represents the equilibrium contribution, which corresponds to non-reactive systems. This later can be expressed by two distinct terms:

$$W_{\mathrm{equil}} = W_{\mathrm{chem\text{-}equil}} + W_{\mathrm{VDW}}, \tag{3}$$

where $W_{\mathrm{chem\text{-}equil}}$ is the adhesion energy between the two contact phases, which results from the establishment of the chemical equilibrium

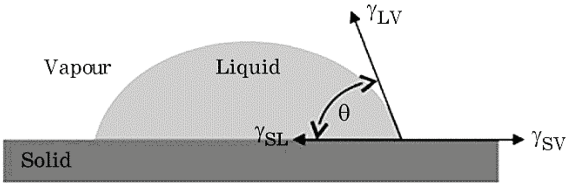

Fig. 1. Schematic profile of a contact angle θ in a solid–liquid–vapour system in equilibrium [16].

bonds obtained by the mutual saturation of the free valences of the surfaces in contact. The formation of these chemical bonds is not accompanied by the rupture of the interatomic bonds in metal–ceramic interface, which takes place in the chemical non-equilibrium systems.

W_{VDW} represents the van der Waals interactions.

During the propagation of the ultrasonic waves, the particles undergo a sinusoidal vibratory displacement around their rest position. Consequently, their density varies by making regions appear denser and others less dense than when they are at rest. The ratio of these deletions and depressions by the acoustic velocity defines the notion of

TABLE 1. Young's moduli E_C and densities ρ_C of ceramics and experimental values work of adhesion W_{ad} in a various metal–ceramic systems.

Ceramic	E_C, GPa [14]	ρ_C, kg/m^3 [14]	Metal	Atmosphere	W_{ad}, mJ/m^2	Refs.
1	2	3	4	5	6	7
AlN	350	3260	Ag	Vacuum	630	[19]
			Al	Vacuum	1136	[19]
			Au	Vacuum	650	[19]
			Co	Vacuum	1270	[19]
			Cu	Vacuum	1060	[20]
			Fe	Vacuum	1320	[19]
			Ga	Vacuum	750	[19]
			Ge	Vacuum	811	[19]
			In	Vacuum	448	[19]
			Ni	Vacuum	1305	[19]
			Pb	Vacuum	203	[19]
			Pd	Vacuum	858	[19]
			Si	Vacuum	1058	[19]
			Sn	Vacuum	461	[20]
Al_2O_3	400	3980	Al	Vacuum	948	[21]
			Au	Vacuum	577	[21]
			Fe	Vacuum	1202	[21]
			Ga	Vacuum	537	[21]
			In	Vacuum	335	[21]
			Ni	Vacuum	1191	[21]
			Pb	Vacuum	218	[21]
			Si	Vacuum	876	[21]
			Sn	Vacuum	305	[21]
BeO	390	3010	Cu	Ar	600	[20]
			Fe	He	717	[20]
			Ni	Vacuum	680	[20]
			Pb	Vacuum	130	[17]
BN	34	3487	Au	Vacuum	205	[22]
			Cu	Vacuum	345	[22]
			Si	Vacuum	364	[22]
			Sn	Vacuum	128	[22]

Continuation of TABLE 1.

1	2	3	4	5	6	7
CoO	191	9423	Co	Ar	2526	[2]
			Ni	Ar	2705	[2]
			Sn	Vacuum	994	[2]
MgO	307	3580	Ag	Ar	421	[22]
			Fe	Vacuum	820	[22]
			In	Vacuum	172	[20]
			Ni	He	585	[22]
			Sn	Vacuum	278	[20]
NiO	220	6670	Ag	Ar	1267	[23]
			Cu	Ar	1738	[23]
			Ni	Ar	2652	[23]
			Sn	Vacuum	921	[24]
TiO	387	4950	Au	Vacuum	1858	[20]
			Cu	Vacuum	1581	[20]
			Ni	Vacuum	2652	[20]
SiO_2	72	2600	Au	Vacuum	165	[22]
			Cu	Vacuum	390	[22]
			Sn	Vacuum	253	[24]
ZnO	125	5606	Ag	Ar	747	[23]
			Cu	Ar	1060	[23]
			Sn	Ar	481	[23]
ZrO_2	150	5600	Ag	Vacuum	446	[25]
			Cu	Vacuum	594	[25]
			Pb	Vacuum	114	[25]

impedance. According a normal incidence of the acoustic wave on a flat surface, the acoustic impedance Z expressed by:

$$Z = \rho c. \tag{4}$$

Following the various calculations derived from this equation [18], we will use the general form of the acoustic impedance Z as a function of the density ρ and the Young's modulus E of the transverse acoustic wave:

$$Z = (\rho E)^{1/2}. \tag{5}$$

The stresses imposed by a liquid on the surface of the ceramic are mainly due to the viscosity. The liquid metal–ceramic coupling results in radiation in the liquid of a highly damped transverse wave. The acoustic reflection coefficient at the interface is written as follows:

$$R = (Z_{\mathrm{LM}} - Z_{\mathrm{C}})/(Z_{\mathrm{LM}} + Z_{\mathrm{C}}), \tag{6}$$

where Z_{LM} and Z_C are the impedances of the liquid metal and ceramic. The reflection coefficient R makes it possible to determine the transmitted energy as a function of the impedances at the liquid metal–ceramic interface. This energy is determined by its transmission coefficient, which is defined as follows:

$$T = 1 - R. \tag{7}$$

3. RESULTS AND QUANTIFICATION

A new acoustic model is proposed to interpret the work of adhesion in non-reactive metal–ceramic systems. In this model, the energy transfer by the sound propagation is assured by the existence and the excess of the interfacial bounds between a metal and a ceramic. The relevant parameters determining the work of adhesion of a metal–ceramic system have been found to be the sound propagation velocity in liquid metal and the acoustic impedance of the solid ceramic.

Detailed experimental results of the work of adhesion for various metal–ceramic systems are summarized in Table 1. It should be noted that the criterion of liquid metals and given ceramics selected in this investigation must have the experimental W_{ad} values of at least three different contacting metals available in the literature.

From Figure 2, it can be observed that the work of adhesion of different liquid metal–aluminium nitride (AlN) interfaces increases line-

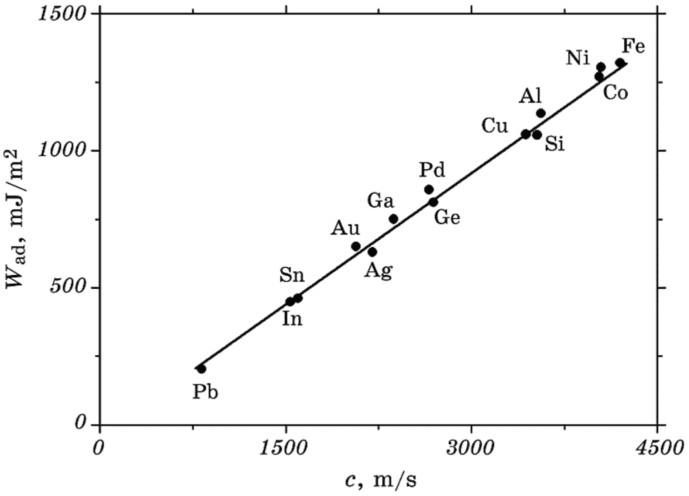

Fig. 2. Correlation between work of adhesion W_{ad} for different liquid metal–AlN systems and sound velocities c of corresponding liquid metals. The sound velocity values of liquid metals are taken from Blairs [27].

arly when the sound propagation velocities c of the corresponding metal increases.

Thus, it is important to define a new interfacial characteristic, which defines the energy transfer strength by the interfacial bounds to the ceramic phase, depending on the stability and the strength of the interfacial adhesion between different metals in contact with the ceramic that is the slope parameter of the work of adhesion $\xi = dW_{ad}/dc$. Similar idea was used by Li [26] for the same systems but for the dependence of the work of adhesion and the electron density n_{ws}, which is responsible of the electronic transfer between the metallic phases and the ceramics. Therefore, the electron density allows the formation of the interfacial bonds whether in excess or in deficit depending on the type of the ceramic.

The linear correlation of the points presented in Fig. 2 yields a ξ value of 0.312 mJ·s/m^3 for aluminium nitride. These results, together with the ξ values, obtained for other solid ceramic materials are given in Table 2.

It is important to note that, the W_{ad} values for the selected liquid metal–ceramic systems exhibit a good convergence as a function of the sound velocity of several liquid metals on a ceramic, as indicated by the values of the regression coefficients given in Table 2.

In Figure 3, the slope parameter ξ values for work of adhesion for various ceramics are plotted as a function of the acoustic impedance Z_C of the corresponding ceramics. It can be seen that the slope parameter ξ stabilizes at about 0.185 mJ·s/m^3; however, then it increases sharply. For $Z_C > 38 \cdot 10^6$ kg/m^2·s, ξ seems to stabilize again, but at about 0.600 mJ·s/m^3.

The excellent correlation between ξ and Z_C as presented in Fig. 3 demonstrates that the work of adhesion slope parameter ξ of liquid

TABLE 2. Slope parameter ξ of work of adhesion for various solid ceramic materials and the coefficient of the linear regression R.

Ceramic	ξ	R
AlN	0.312	0.9951
Al_2O_3	0.270	0.9780
BeO	0.200	0.9991
BN	0.184	0.9846
CoO	0.606	0.9954
MgO	0.203	0.9389
NiO	0.604	0.9526
TiO	0.603	0.9066
SiO_2	0.183	0.9464
ZnO	0.585	0.9880
ZrO_2	0.183	0.9764

metal–ceramic interfaces depends only on the nature of the ceramic and not on the contacting liquid metals.

From Figure 3, we can distinguish two different cases of the variation of ξ as a function of Z_C. The first part of this figure reveals that the reflection mode R of the propagating acoustic wave in liquid metal becomes the most dominant according to the equation (6), for the impedance acoustic values of ceramics lower than that of AlN ($Z_C < Z_{AlN}$). Therefore, the acoustic wave will be reflected in this middle and only a small part of the energy will be transmitted from the metal–ceramic interface because of a low interracial adhesion. The work of adhesion W_{ad}, in this part, is only resulting from the van der Waals interactions W_{VDW}, and the chemical equilibrium contribution $W_{chem\text{-}equil}$ is negligible [20, 22, 23].

The determination of W_{VDW} values for different metal–ceramic systems have been reported in different previous researches. For example, Naidich [17] found a W_{VDW} value of 350 ± 150 mJ/m^2 for metal–oxide ceramic systems.

The second case, which corresponds to the values of ceramic acoustic impedance Z_C greater than that of AlN ($Z_{AlN} < Z_C$), shows that the transmission T becomes the most dominant mode. The energy of the propagating wave in the liquid will be transmitted almost totally to the ceramic from metal–ceramic interface. In this limiting case, the work of adhesion W_{ad} is approximately determined by the surface tension γ_{LV} of liquid metals as indicated by equation (1), and the work of adhesion slope parameter ξ is proportional to $d\gamma_{LV}/dc$, that is the linear dependence of the surface tension of liquid metals on the sound propagation

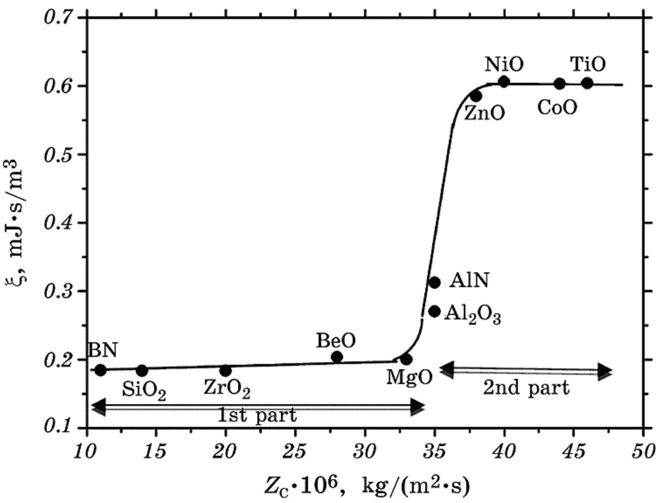

Fig. 3. Work of adhesion slope parameter ξ values of various ceramic materials as a function of the acoustic impedance Z_C of the corresponding ceramics.

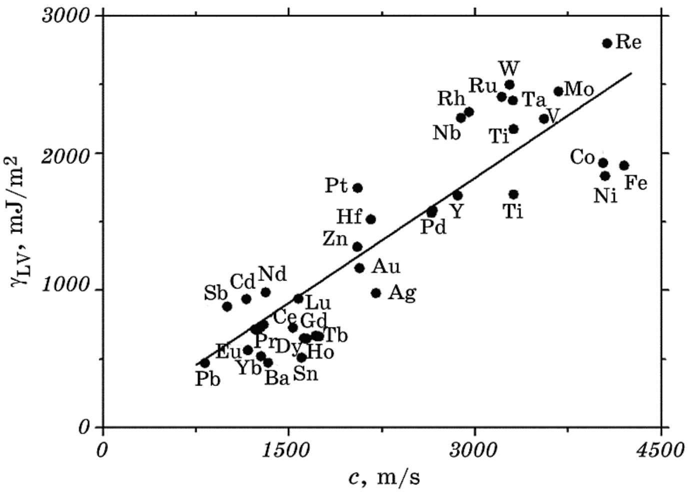

Fig. 4. Correlation between the surface tension γ_{LV} of various liquid metals and the sound velocity c of the corresponding metals. The surface tensions of liquid metals are taken from Keene [28].

velocity of corresponding metal.

A linear correlation between γ_{LV} values of various liquid metals against c is shown in Fig. 4. An important point that can be interpreted from this figure is the possibility of determining the unknown surface tension as function of a known sound velocity of liquid metals and *vice versa*.

The points presented in Fig.4 yields a surface tension slope parameter equal to 600 mJ·s/m^3, which corresponds exactly to the upper limit of Fig. 3. It is noted that this limit of has been already saturated for solid ceramic materials characterized by a dominant transmission energy mode, which explain the good interfacial adhesion.

4. CONCLUSIONS

In this paper, the work of adhesion of different liquid metals on a given ceramic was investigated. A new approach of the interfacial phenomenon was introduced. This novel interfacial phenomenon investigation was deduced after the study of W_{ad} *versus* the sound velocity propagation of plate acoustical wave in corresponding metals that shows a linear correlation. Moreover, the work of adhesion slope parameter for several ceramic materials shows a strong dependence on the acoustic impedance of the corresponding ceramics. This result proves that this slope parameter depends only on the ceramic properties. Hence, the determined correlation between the slope parameter values and the acoustic impedance of various ceramic materials has a deep effect on the nature of the acoustical wave propagation (reflective or transitive) in metal–ceramic interfaces according to the existence and the excess of the interfacial bonding.

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