Review
Wetting of rare-earth element oxides by metallic melts

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The results of studies on the wettability and contact interaction of rare-earth (REE) oxides and aluminium oxide (for comparison) with metallic melts are presented. Proceeding from analysis of the data obtained, a mechanism of interaction of metallic melts with rare-earth element oxides is proposed, based on the interaction of the metallic melt components with the cationic and the anionic sublattice of the oxides. The necessity to take into account the interaction of the melt metal with the cationic sublattice of oxide is determined by a high chemical affinity of the REE for quite a number of metals and by specific features of the structure of the surface of the oxides.

1. Introduction
A considerable body of data on the wetting and contact interaction of "classical" high-melting oxides of aluminium, silicon, beryllium, magnesium, etc., with metallic melts has been obtained to date [1–6], making it possible to define the basic physico-chemical laws of the adhesion phenomena in these systems and ways for their use in solving quite a number of technological problems, such as soldering of oxidic materials, fabrication of composites, development of refractories, etc.

According to existing concepts, the surface of such an oxide, due to the large size of an anion as compared to a cation and its higher polarizability, is formed by anions, and the wettability in these systems is governed by the energetics of metal melt interaction with oxygen of the oxide being wetted.

The use of material containing oxides of rare-earth elements (REE) is promising in a number of fields of modern technology, and therefore a knowledge of the adhesive properties of the REE oxides seems to be important.

The REE oxides feature high values of free energy of formation, which are about 1.2 times that for the most stable oxides (aluminium oxide) [7, 8]. From this standpoint it may be expected that the REE oxides as a whole should be less well wetted by liquid metals.

On the other hand, REE have relatively large ionic radii and display a high chemical affinity for a considerable number of metals, which may give rise to a number of specific features in the development of capillary phenomena in the REE oxide-metallic melt contact systems.

This work involved a systematic investigation into the wetting and contact interaction of the REE oxides of the yttrium group (yttrium oxide and oxides of the elements from gadolinium to lutetium) with the metals of the I-b, III-a, and IV-a subgroups of the Periodic system, their alloys with titanium, and comparison of the data with the wettability of aluminium oxide (as reference).

2. Experimental and sample preparation procedures
2.1. Samples and materials used
The polycrystalline samples of REE oxides used for studying the wetting and contact interaction in the metal-oxide systems under consideration were in pellet form, 14 to 18 mm diameter and 3 to 5 mm thick. The compact samples were prepared by isostatic pressing of powders of the corresponding REE oxides followed by sintering and annealing in an air atmosphere and in a vacuum. The content of the basic substance oxide, in the used powders was not less than 99.95 mass %. The metals used in the studies contained over 99.98 mass % of the main element.

2.2. Wetting
The degree of wettability of the oxidic materials by the metallic melts was determined by the sessile drop method in a vacuum of 10^{-3} Pa [1] at a temperature of 1150° C. The concentration dependences of the wettability of oxides by multicomponent titanium-containing melts were studied by changing the melt composition directly in the drop during the course of the experiment [9]. Not less than four independent experiments, whose results were averaged, were conducted for every metal-oxide contact system.

As well as measuring the value of the wetting angle, the increase in diameter of the wetting perimeter was noted in the event of a decrease of the angle. This made it possible to eliminate any error in determining the wetting angle, which might result from a possible decrease of the melt volume during the course of an experiment, because of evaporation of the melt or of its penetration into cracks that sometimes arose in a ceramic sample.
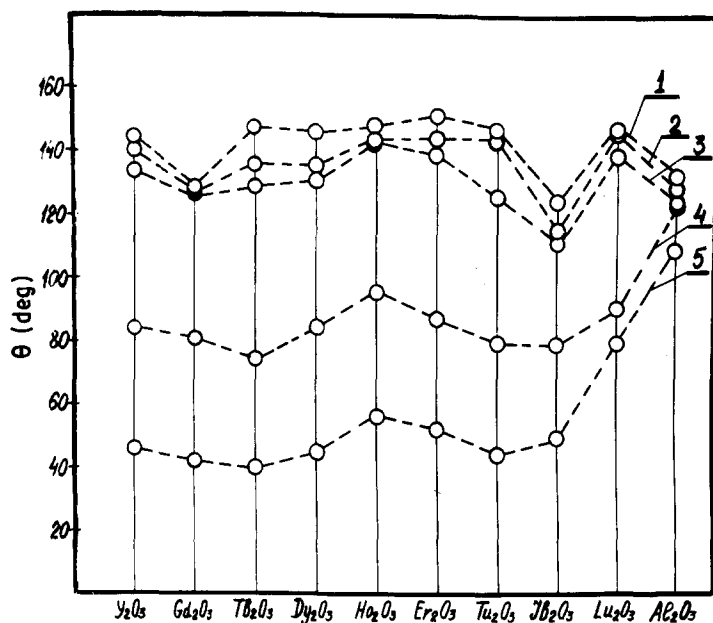
Figure 1 Angles of wetting of REE oxides and aluminium oxide by melts of metals: (1) Cu; (2) Ag; (3) Au; (4) Sn; (5) Ge; $T = 1150°$ C.

2.3. Contact interaction

The interphase interaction (processes occurring at the oxide–metallic melt interphase boundary) was investigated using mass-spectrometric, metallographic, and X-ray microspectrum analyses. The loss of mass of the samples (rate of evaporation of the wetting metal) was determined by weighing.

2.3.1. Mass-spectrometric studies

The nature and rate of interaction in oxide–metal systems were studied by examining the composition of the vapour phase above the systems when new components (vapours of metals of the oxides being wetted, monoxides of the wetting metals) were recorded in the mass spectrum.

The studies were conducted on a standard MI-1305 mass spectrometer adapted for high-temperature measurements [10].

The sample under study, which was a mix of powders of the corresponding oxide and metal in a 1 : 1 volume ratio, was placed, in the heater of the mass spectrometer. The sensitivity of the instrument was 10^{-4} Pa. The mass spectra were recorded at an ionizing electron energy of 42 eV.

2.3.2. X-ray microspectrum studies

That zone of the interphase interaction of a high-melting oxide with metallic melts was examined using a Superprobe 733 X-ray microspectrum analyser. The distribution of the alloy and oxide elements on metallographic specimens cut perpendicular to the wetting plane, as well as on the surface of oxide plates extracted from the melt in a vacuum at the temperature of the experiment, were determined.

3. Results

3.1. Wettability of REE oxides by metallic metals

The studies of the wettability of REE oxides by melts of metals copper, silver, gold, aluminium, gallium, tin and germanium resulted in establishing the following points.

1. Copper, silver and gold do not wet these oxides (Fig. 1 curves 1, 2, 3), the wetting angles being from 120 to 145°. An insignificant drift (decrease) of the wetting angle with time was observed. The final values of the wetting angle were reached in 5 to 10 min, i.e. in about the same time as on aluminium oxide.

The values of the angles of wetting of REE oxides by these metals turned out to be greater than on aluminium oxide, with the exception of ytterbium oxide. The observed evaporation of metals from drops on substrates of REE oxides appeared to be of a purely thermal origin; no difference in the evaporation rates on various substrates of REE oxides and aluminium oxide was observed.

2. Metals of the III-a and IV-a subgroups of the Periodic system (aluminium, gallium, tin, germanium) wet the REE oxides considerably better than aluminium oxide (Table I, Fig. 1 curves 4, 5). For

TABLE I Wettability of REE oxides by aluminium and gallium ($T = 1150°$ C)

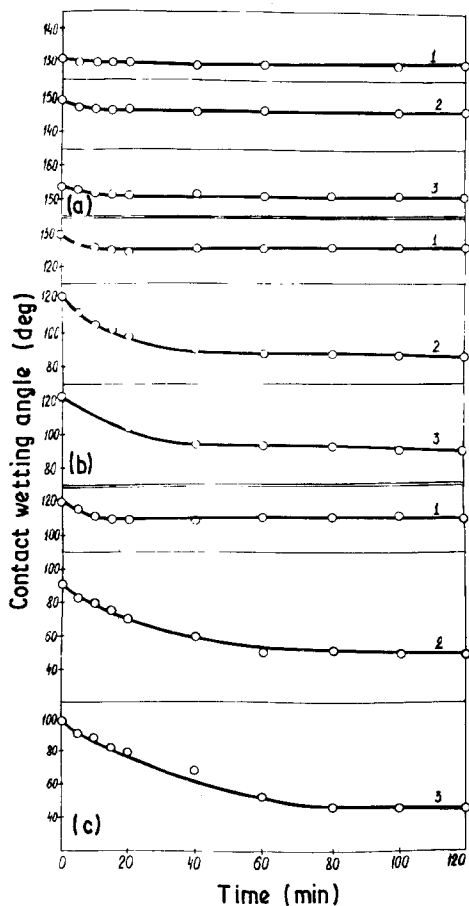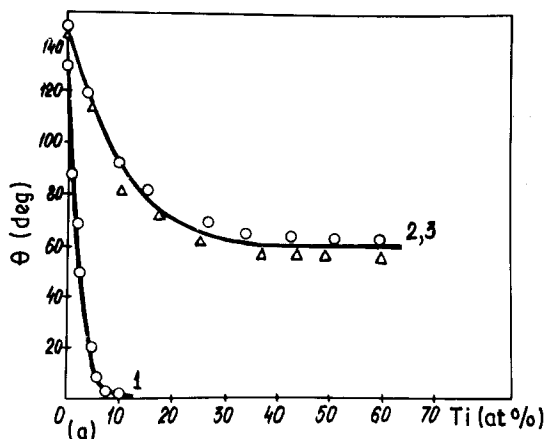| Oxide–metal system	Wetting time (min)						Metal mass loss in 30 min (%)
	0	5	10	15	20	30	
Al_2O_3–Al	92	85	80	80	80	80	4
Y_2O_3–Al	95	87	85	75	60	55	8
Er_2O_3–Al	90	86	84	70	63	58	7
Ho_2O_3–Al	94	84	82	74	66	55	9
Al_2O_3–Ga	129	124	124	124	122	120	15
Y_2O_3–Ga	114	114	114	112	108	102	48
Er_2O_3–Ga	118	118	118	116	112	110	42
Ho_2O_3–Ga	119	119	118	112	105	97	47

Figure 2 Time variation of angles of wetting by (a) Cu, (b) Sn, (c) Ge of oxides (1) Al_2O_3, (2) Y_2O_3, (3) Er_2O_3.

[1, 11–13], the wettability of an oxide by melts can be enhanced by adding a component exhibiting a high affinity for oxygen, such as titanium, into the liquid phase. This method for controlling the wettability proves also to be efficient in the case REE oxides, i.e. the introduction of a metal having a high affinity for oxygen also results in strengthening of the interphase interaction in these systems.

Fig. 3 shows the concentration dependences of the angle of wetting of oxides of aluminium, yttrium and erbium by (Cu–Ti) and (Sn–Ti) melts. The values of wetting angles in REE oxides–(Cu–Ti); (Au–Ti); (Ag–Ti); (Sn–Ti); and (Ge–Ti) melt systems are given in Fig. 4.

The results obtained on the wettability in the studied systems demonstrated that titanium-containing binary alloys of metals of the I-b subgroup of the Periodic system wet REE oxides less than they wet aluminium oxide.

The values of the angles of wetting of REE oxides by titanium-containing melts of metals of the IV-a subgroup of the Periodic system turned out to be less than for aluminium oxide.

3.3. Mass-spectrometric studies

The results of the mass-spectrometric examinations of the composition of the vapour phase over the mix of the oxide and metal (copper, tin, germanium) powders are presented in Table II. The analysis of the data obtained and their comparison with the wettability data show that in the systems characterized by a poor wetting (Er_2O_3–Cu; Al_2O_3–Cu), ions of erbium and aluminium and of their monoxides are observed in the vapour phase within practically the same temperature range as over pure oxides, i.e. no chemical action occurs in these systems (Table II).

For the REE oxides–tin and REE oxides–germanium contact systems, where wetting angles < 90° were observed, the mass-spectrometric analysis detected a lowering of the temperature of appearance of an REE (erbium) and of its monoxide in the vapour phase compared to a pure oxide, by more than 600° C with Er_2O_3–Ge and by more than 350° C with Er_2O_3–Sn. This result indicates the occurrence of the chemical action in these systems. Apart from the REE and its monoxide, tin and germanium oxides were also observed in the vapour phases at the initial
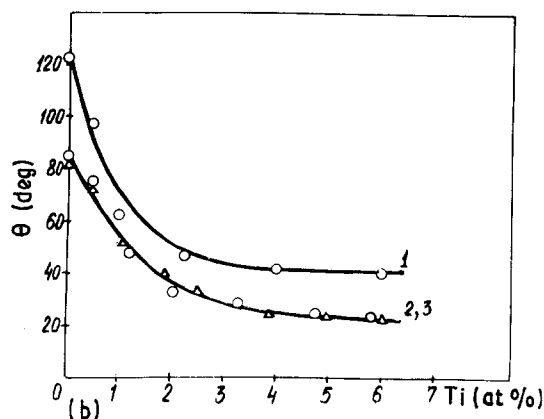
germanium, wetting angles on most oxides amounted to 40 to 50°. In the case of wetting of REE oxides by tin and germanium, the time for the wetting angle to reach a constant value increased considerably compared with that for aluminium oxide (Figs 2b, c). In the process of wetting of REE oxides by aluminium, gallium, tin and germanium, a higher rate of evaporation of the wetting metal from drops located on REE oxide substrates over that with aluminium oxide substrate was observed, e.g. for the germanium–erbium oxide system, 1.5 to 2 times, and for aluminium–REE oxide systems, up to 3 times as high.

3.2. Wetting of oxides by binary titanium-containing melts

According to the concepts presented in the literature

Figure 3 Concentration dependences of wetting by melts (a) Cu–Ti, (b) Sn–Ti of oxides (1) Al_2O_3, (2) Y_2O_3, (3) Er_2O_3.
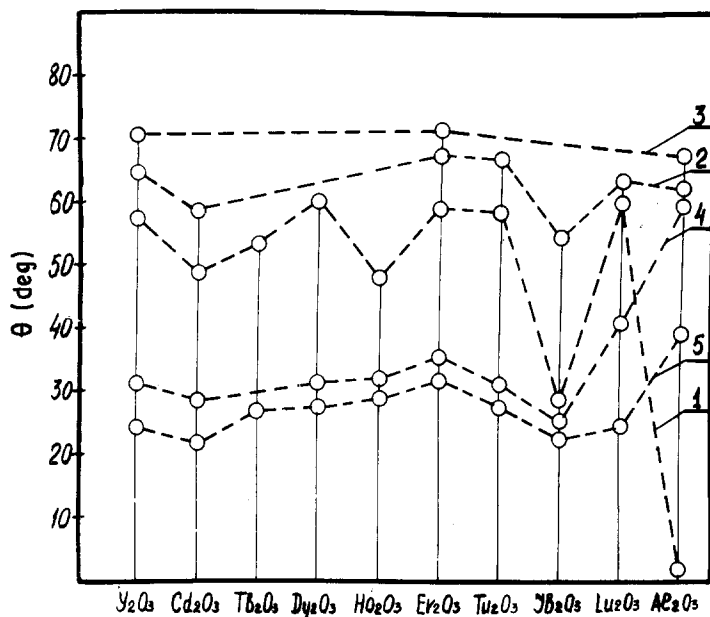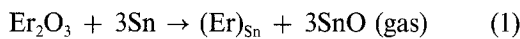
Figure 4 Wetting of REE oxides and aluminium oxide by titanium-containing melts at 1150°C: (1) Cu + 57 at % Ti; (2) Au + 8 at % Ti; (3) Ag + 4 at % Ti; (4) Sn + 4 at % Ti; (5) Ge + 5 at % Ti.

temperatures of interaction, and the probable reaction proceeding at the contact boundary can be written as

$$Er_2O_3 + 3Sn \rightarrow (Er)_{Sn} + 3SnO \text{ (gas)} \qquad (1)$$

A higher rate of evaporation of wetting metals from drops in contact with REE oxides is apparently caused by the formation of volatile monoxides.

In aluminium oxide–tin and aluminium oxide–germanium systems, the temperatures at which aluminium appears in the vapour phase are close to those of its appearance over a pure oxide, while germanium monoxide was detected only at 1570° C.

3.4. X-ray microspectral studies of oxide–metal interphase boundary

With copper, no changes in the composition of contacting phases occurred at the contact boundary. With germanium with good wetting of the REE oxides, the formation of a new "needle-like" phase (Fig. 5) at the contact boundary, near the wetted perimeter, was observed. The elementary analysis demonstrated that the "needles"consist of germanium and erbium and contain no oxygen. Their composition is close to that of the ErGe₃ compound. They appear to form during the course of melt crystallization. The formation of this phase near the wetted perimeter results from the most favourable conditions, both for the removal of

gaseous oxides (SnO, GeO) forming during the process of the reaction, and for dissolution of REE cations in the wetting metal.

While examining the zone of interaction between REE oxides and titanium-containing melts of copper, tin and germanium, the formation of transition layers consisting of titanium oxides (TiO, Ti₂O₃, etc.) at the interphase boundary was not to be detected, in contrast to the results reported for the titanium-containing melts with an aluminium oxide contact boundary [11–13].

In the case of interaction of erbium oxide with copper–titanium melts containing a low proportion of titanium (2%), some increase in the titanium concentration at the interphase boundary relative to its concentration in the bulk was found. With a titanium concentration of 15 at % in the melts, no increase in its content at the interphase boundary was observed. Inclusions, apparently of Er₂O₃, resulting from the interaction were found in the melt, their number increasing with the titanium concentration (Fig. 6d). The sizes of some inclusions were of the regions of the destruction (erosion) of the ceramics (Er₂O₃) by the melt. Etching of the ceramics mostly at the grain boundaries is seen (Fig. 6a) at a prepolished surface of an oxide sample extracted from the melt (Cu + 15 at % Ti) at the temperature of the

TABLE II Temperature of appearance of reaction products in vapour phase in Me₂″O₃ (solid) + Me′ (liquid) systems

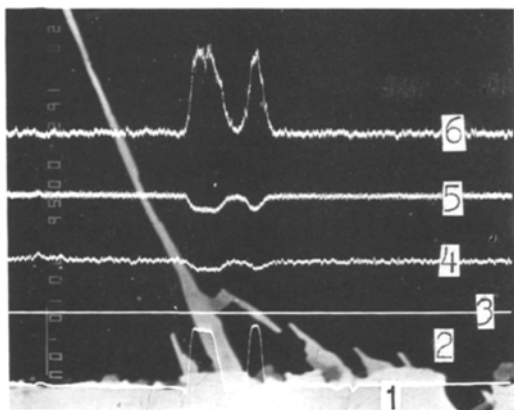| Sample composition	T (° C)	Vapour phase composition (Pa)				
		$P_{Me''}$	$P_{Me''O}$	$P_{Me'}$	$P_{Me'O}$	$P_{Me'O_2}$
Cu	1527	–	–	5.7×10^{-4}	–	–
Sn	1114	–	–	1.3×10^{-1}	–	–
Ge	1575	–	–	7.1×10^{-3}	–	–
Er₂O₃	1922	1.4×10^{-3}	6.7×10^{-4}	–	–	–
Er₂O₃–Cu	1927	1.4×10^{-3}	2.4×10^{-2}	10^{-1}	n/d	n/d
Er₂O₃–Sn	1572	5×10^{-3}	n/d*	2×10^{-1}	1.5×10^{-2}	n/d
Er₂O₃–Ge	1287	2.7×10^{-4}	n/d	1.4×10^{-3}	4.8×10^{-3}	3.3×10^{-3}
Al₂O₃	1727	6.7×10^{-4}	–	–	–	–
Al₂O₃–Ge	1682	7×10^{-2}	–	4.4×10^{-2}	2.3×10^{-2}	3.5×10^{-3}
Al₂O₃–Sn	1422	1×10^{-2}	–	4×10^{-2}	–	–

*n/d - not detected.

Figure 5 Oxygen-free germanium–erbium compounds in erbium oxide (1) –germanium (2) contact zone. Distribution of elements along path (3); oxygen (4); germanium (5); erbium (6).

experiment, the grains dissolving to a depth of 1 to 3 μm. No formation of new phases occurred at the contact surface of erbium oxide during the course of interaction with copper–titanium melts (Figs 6b, c). The process of interaction in these contact systems resulted in dissolution of the REE oxide in the alloy.

For studying the interphase interaction of titanium–containing melts of tin and germanium with REE oxides, alloy compositions were selected which corresponded to the region of minimum values of wetting angles in these contact systems.

Fig. 7a shows a general view of the interphase boundary in the Er_2O_3–Sn + 4 at % Ti system. The distribution of intensities of the characteristic radiation of elements (Figs. 7b, c and d) shows that the "sponge" formed during the course of the interaction is a loose erbium oxide, through the discontinuities of which the tin, containing practically no titanium, penetrates to the unreacted ceramics. Titanium is chemisorbed on erbium oxide as it penetrates through
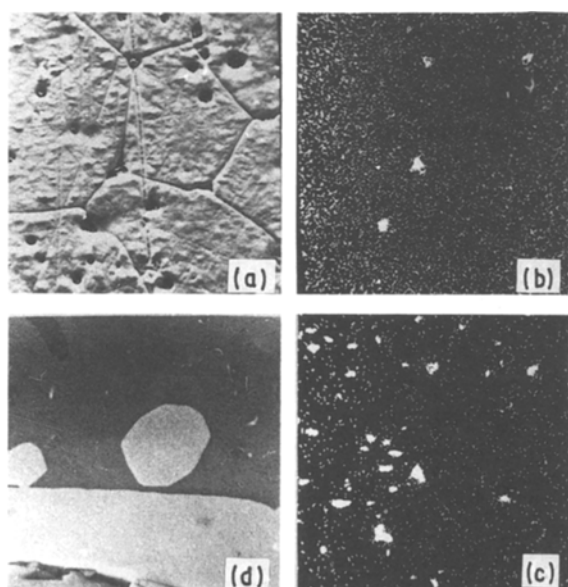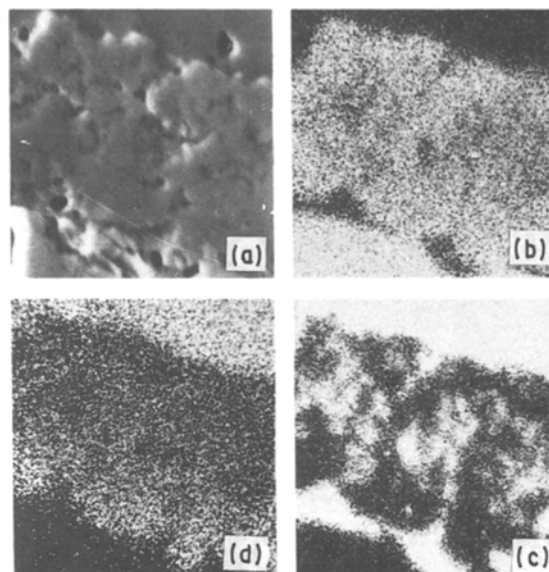

Figure 7 Interphase interaction of erbium oxide with tin–titanium melt: (a) general view: bottom left, ceramics; top right, alloy Sn + 4 at % Ti; centre, reaction zone. Distribution of elements at contact boundary: (b) erbium; (c) tin; (d) titanium. × 400.

the layer of interacted ceramics, but forms no continuous layer of combination with oxygen. The rare-earth element in the course of the interaction passes into the alloy and was found by us as a tin–erbium compound with the composition $ErSn_2$.

An intense interaction was observed at the interphase boundary in the Er_2O_3–$(Ge + Ti)$ melt system, resulting in an active passage of the REE into the melt, the REE subsequently forming oxygen-free compounds with germanium of the same composition as in the case of a pure metal, but in a greater amount. Titanium from the melt becomes chemisorbed on the reacted layer of ceramics, and a practically pure germanium, as in the case of tin, penetrates to the unreacted ceramics.

4. Discussion

According to the chemical theory of wetting, the wettability of high-melting "classical" oxides (oxides where anions are much larger than cations, such as aluminium oxide) by metallic melts is governed by the intensity of metal melt interaction with the oxide oxygen, i.e. is determined by the difference between the value of the free energy of formation of the high-melting oxide and the affinity of the wetting metal for oxygen. From this standpoint, REE oxides should not be wetted by ordinary metals (metals having a low affinity for oxygen).

However, the strength of all the evidence obtained on wetting and contact interaction of REE oxides with metals demonstrates that in a number of systems with more thermodynamically stable REE oxides (in contact with tin, germanium, aluminium) the wettability proves to be higher than for aluminium oxide, the thermodynamic potential of whose formation is somewhat lower than that for REE oxides; thus $-\Delta G^0_{f1423} Ln_2O_3$ are within 1481 to 1571 kJ mol^{-1} [7] while for aluminium oxide $-\Delta G^0_{f1423} Al_2O_3$ is

Figure 6 Erbium oxide interaction with copper–titanium melt: × 500. (a) Surface of erbium oxide plate extracted from the melt in vacuum at 1150° C after hourly isothermal holding; distribution of copper (b) and titanium (c) over plate surface; (d) formation of Er_2O_3 inclusions in melt, × 1500.
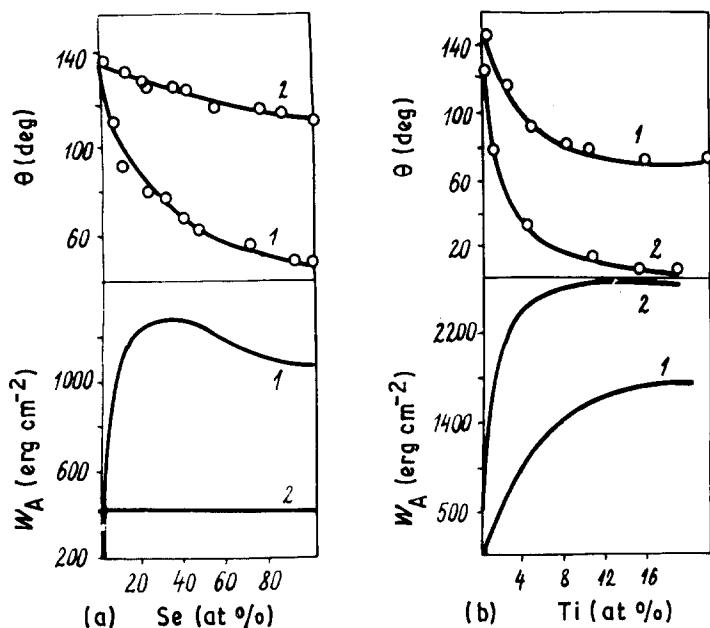
Figure 8 Wettability and work of adhesion of (a) Cu–Ge and (b) Cu–Ti melts to (1) yttrium oxide and (2) aluminium oxide; $T = 1150°$C.

1248 kJ mol^{-1} [8], Table III (where Ln = Y and elements from Gd to Lu).

Thus, there exists an additional (apart from the metal–oxygen interaction) energy factor which contributes considerably to the work of adhesion. It was suggested that such a factor in the interaction of liquid metal with the cationic sublattice of the REE oxide. The contribution appears to be determined by the intensity of interaction of the REE itself with the wetting metals. Proceeding from the ratio of the ionic radii of oxygen and rare-earth elements ($R_{Ln}^{3+}/R_O^{2-} = 0.6$ to 0.7) [7] as compared with this ratio for classical oxides (0.22 to 0.4) (Table III), it can be supposed that metallic ions are as well represented at the surface, i.e. the surface of REE oxides is formed by two types of ions, oxygen and metal. Thus, a sufficient "geometric permissiveness" of cationic sublattice interaction with the melt metal apparently exists in these systems, whereas for oxides of aluminium, beryllium, silicon, magnesium having a small R_{Me}^{n+}/R_O^{2-} ratio, the interaction of the metal melt with the oxide cation is spatially difficult. It can be supposed that the oxide surface is, in general, heterogeneous and its heterogeneity can be characterized by the proportion of area occupied by oxide cations and anions, respectively, the portion of the surface area formed by cations being in the case of REE oxides much greater than that for "classical" aluminium, silicon, magnesium and beryllium oxides.

Rare-earth elements also exhibit high energies of interaction with quite a number of metals, which is evidenced by the values of free energies of formation

of intermetallic compounds REE–tin: $-\Delta G_{fLnSn_3}^0$ (Ln–Y, Er, Gd, Lu) are respectively 209, 197, 178, 157 kJ mol^{-1} [16] (the enthalpy of formation of an alloy corresponding to a composition of AlSn$_3$ is as little as 8 kJ mol^{-1} [17], Table IV lists the partial enthalpies of dissolution for systems of REE with tin, germanium, copper and of aluminium with the same metals. In systems with REE, their values increase from copper to tin and germanium, while in systems with aluminium their values are low.

An analysis of the features of the interaction of REE oxides with metals suggests the following mechanism for the interphase reaction: the melt metal interacts with both oxygen and the oxide metal, with the result that the REE partly passes into the liquid phase. For a number of systems, oxygen is brought out of the reaction zone in the form of gaseous oxides of the wetting metal, which results in shifting the equilibrium of Reaction 1 to the right. The rare-earth element which has passed into the liquid phase participates in the formation of the melt–oxide transition zone. These processes appear to be the cause of the increase in spreading time (hundreds of minutes) in REE oxide–tin, germanium metals contact systems.

To attain a high degree of wetting and adhesion of metallic melts to oxides, components exhibiting a high affinity for oxygen, such as titanium, should be added to the melt. Titanium also exhibits an interphase activity in REE oxide–titanium-containing melt systems. The proposition stated above, on the contribution of the metal–cation interaction to interphase processes, makes it possible to suggest that

TABLE III Ratio of ionic radii of metal and oxygen and free energies of formation of oxides Me$_2$O$_3$

	Be^{2+}	Mg^{2+}	Al^{3+}	Si^{4+}	Y^{3+}	Gd^{3+}	Tb^{3+}	Dy^{3+}	Ho^{3+}	Er^{3+}	Tu^{3+}	Yb^{3+}	Lu^{3+}
$R_{Me^{n+}}$ (nm) [8]	0.31	0.65	0.41	0.50	0.93	0.94	0.92	0.91	0.89	0.87	0.86	0.85	0.84
$R_{Me^{n+}}/R_{O^{2-}}$	0.22	0.46	0.29	0.36	0.66	0.67	0.66	0.65	0.63	0.62	0.61	0.61	0.60
$-\Delta G_{f1423}^0$ Me$_2$O$_3$ (kJ mol^{-1})	–	–	1248	–	1571	1489	1535	1529	1546	1556	1553	1481	1539

TABLE IV Partial enthalpies of oxide metal dissolution in melts

	Melt					
	Er–Cu	Er–Sn	Er–Ge	Al–Cu	Al–Sn	Al–Ge
$\Delta \bar{H}$ (kJ mol^{-1})	−92 [15]	−129 [16]	−213 [18]	−4 [14]	+8 [14]	−20.5 [17]

elements featuring a high affinity for the cation will also be interphase-active in such contact systems.

To substantiate this point, compare the wettability and adhesion of a REE oxide by copper alloyed in one case with germanium and in the other case with titanium (Fig. 8). The work of adhesion was in all cases calculated from Equation 2.

$$W_A = \gamma_{LV}(1 + \cos\theta) \qquad (2)$$

For Cu–Ga and Cu–Ti alloys the values of γ_{LV} given in [18] were used. A possible decrease in γ_{LV} of alloys through oxygen liberated in the interaction between the melt and oxides, was not taken into account.

Pure copper does not wet REE oxides (Fig. 1a). When germanium, which intensely interacts with REE, or titanium, which exhibits a high affinity for oxygen, is added to the melt, an increase in the wettability and adhesion is observed in either case. At the same time, experiments on the wetting of aluminium oxide by copper–titanium and copper–germanium melts (Fig. 8) demonstrate that only titanium-containing melts exhibit an adhesion activity in these systems. No increase in the work of adhesion is observed in aluminium oxide–germanium alloy systems (the energy of aluminium interaction with germanium is low) [17].

5. Conclusions

Based on the results obtained and their analysis, the interaction of metallic melts with oxides of rare-earth elements has been discussed and its mechanism proposed, characterized by the formation of a bond between the melt components and both the anionic and cationic sublattice of the oxide, which is determined by a high chemical affinity of the REE for quite a number of metals and by specific features of the oxide surface structure (large ionic radius, greater area occupied by cations). The wettability in metal–oxide systems of this type can be predicted by using the data on the energies of formation of the wetted oxide and energies of chemical reactions proceeding at the interaction of the liquid metal with oxygen and the rare-earth element.

Acknowledgement

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