Applied Surface Science 257 (2011) 5077–5082

Contents lists available at ScienceDirect

Applied Surface Science

journal homepage: www.elsevier.com/locate/apsusc

Estimation of the transient interfacial heat flux between substrate/melt at the initiation of magnesium solidification on aluminum substrates using the lumped capacitance method

E. Hajjari [a], M. Divandari [a,*], S.H. Razavi [a], S.M. Emami [a], S. Kamado [b]

[a] Department of Metallurgy and Materials Engineering, Iran University of Science and Technology (IUST), Tehran 16846-13114, Iran
[b] Department of Mechanical Engineering, Nagaoka University of Technology (NUT), Nagaoka 940-2188, Japan

ARTICLE INFO

Article history:
Received 30 October 2010
Received in revised form 5 January 2011
Accepted 6 January 2011
Available online 12 January 2011

Keywords:
Interfacial heat flux
Surface roughness
Aluminum
Magnesium

ABSTRACT

Interfacial heat flux (IHF) between solid pure aluminum/magnesium melt and solid 413 aluminum alloy/magnesium melt couples was evaluated using lumped capacitance method, and the interface microstructures were assessed by scanning electronic microscope. The variation of maximum IHF with surface roughness for these two couples also was evaluated. The results showed that, for both solid aluminum/magnesium melt couples, with increasing the surface roughness, the maximum IHF increases at first and then starts to decrease after reaching a maximum value. In addition the measured maximum IHF for solid 413 aluminum alloy/magnesium melt couples was found to be higher than those measured for solid pure aluminum/magnesium melt couples. That seems to be because of the better wettability of 413 aluminum alloy than pure aluminum, by magnesium melt.

1. Introduction

Heat transfer is an important phenomenon occurring as molten metals begin to freeze on colder substrates. At this early stage, solidification is a short-lived dynamic process occurring in a system out of thermodynamic equilibrium under highly transient heat transfer conditions [1]. The understanding and control of this phenomenon (heat transfer) can find applications in processes having non-equilibrium solidification [2] such as strip casting, thermal spraying, compound casting, permanent mold and direct-chill casting, since the initiation of solidification is also usually rapid. IHF measurement between a solidifying metal and a substrate is also an indirect and qualitative way for estimating the wettability between different melts and metal substrates that has been widely used in recent years. Loulou et al. [3] used a drop-splat set-up to evaluate the heat fluxes during solidification of pure tin, lead and zinc drops on nickel substrate and attempted to correlate wetting with heat transfer data. They reported that metals with lower surface tensions and contact angles provided better heat transfer. Evans and Strezov [4] used a levitation apparatus to melt 0.7 g steel droplets and showed that an increase in the surface tension of steel droplets was associated with a decrease in the maximum heat fluxes. Many other researchers, who have studied the interfacial heat transfer between solid metal substrates and molten metals, have also demonstrated the relationship between IHF or heat transfer coefficient (HTC) with wettability at the interface of solid metal substrates and molten metals. Prabhu and Ravishankar [5] have studied the effect of the sodium modification treatment on casting/chill interfacial heat transfer of Al–13% Si alloy and found that the modification of the melt resulted in an increase in the transient IHF due to improved wetting of the chill surface by the melt containing sodium element. This is in agreement with the results reported by Emadi et al. [6] regarding the effect of sodium on the surface tension of A356 aluminum alloy. Nyamannavar and Prabhu [7] have studied the thermal contact at the interfaces during solidification of Sn–3.5Ag solder alloy against copper and aluminum substrates and found that the IHF for copper substrate is higher than aluminum one. They attributed this to better wetting behavior of copper substrate compared to aluminum one against Sn–3.5Ag solder alloy. Bouchard et al. [1] have examined the relationship between dynamic wetting and heat transfer for aluminum melt and copper substrate in various experimental conditions and reported that an improvement in the dynamic wetting was often accompanied by an increase in the heat transfer, especially in early stages of solidification.

However, studying the interfacial heat transfer between solid metal substrates and molten metals has been a subject of investigation for many years, this phenomenon has not yet been thoroughly studied for light metal couples like magnesium and aluminum. Magnesium and aluminum are the first and second engineering light metals respectively and are attractive in vehicle structure applications for improving energy efficiency, which reduces the emission of greenhouse gases. In many cases, one of these materials

* Corresponding author. Tel.: +98 21 77459151; fax: +98 21 77194208.
E-mail address: Divandari@iust.ac.ir (M. Divandari).

0169-4332/$ – see front matter © 2011 Elsevier B.V. All rights reserved.
doi:10.1016/j.apsusc.2011.01.024

alone does not satisfy the requirements of lightweight constructions, and dissimilar joining between these two metals through different methods is desired. Therefore, understanding the heat transfer behavior between these light metals is one of the important problems regarding their usage. In the present study, the IHF between solid pure aluminum/magnesium melt and solid 413 aluminum alloy/magnesium melt couples has been estimated using a lumped capacitance method described in detail in the next section.

2. Lumped capacitance method

Lumped capacitance method is a method for analysis of IHF and HTC on surface based on change of internal energy at unit time, assuming uniform temperature distribution throughout the body. This assumption is equivalent to saying that the interface resistance is large in comparison with the internal conduction resistance [8,9]. In case of neglecting temperature gradient within the solid, the problem can no longer be considered from within the framework of the heat transfer equation. Under these circumstances, the transient temperature response is determined from balancing the total energy in solid, instead. This balance must relate the rate of energy entered from the interface to the change rate of internal energy as follow [9]:

$$\dot{E}_{en} = \dot{E}_{st} \quad (1)$$

The subscripts *en* and *st* refer to entered and stored energy respectively. Considering the thermal energy as the only sensible component, Eq. (1) can be expressed as:

$$q = hA(T_\infty - T) = \rho c V \frac{dT}{dt} \quad (2)$$

where ρ, c and V are the density, specific heat and volume of the solid material respectively. A is the interface area and t is time. Also T, T_∞, h and q are the instantaneous temperatures of the solid and liquid materials near the surface, interfacial HTC and heat transferred per unit of time, respectively. Having the temperature history of solid material and the liquid near the solid surface, transient interfacial heat flux $(q/A)_t$ and heat transfer coefficient (h_t) can be calculated easily from Eq. (2). Such an analysis yields reasonable estimations within about 5% error when the Biot number defined as equation below, is less than 0.1. [7]

$$Bi = \frac{h(V/A)}{k} \quad (3)$$

where k is the thermal conductivity of the solid material. Since the thermal conductivity of aluminum and magnesium is relatively high, it seems that the error associated with using the lumped capacitance method for analysis of the IHF in Al/Mg couples is small and negligible [10,11]. More details about principles, applications and applicability of this method can be found in the literatures [8,9].

3. Experimental procedure

3.1. Materials

Commercially pure aluminum, 413 aluminum alloy and commercially pure magnesium were used to estimate the IHF between magnesium melt and aluminum substrates throughout the lumped capacitance method. The chemical composition and thermophysical properties of used materials are listed in Tables 1 and 2 respectively.

Table 1
Chemical composition (wt%) of materials used in this study.

Material	Al	Si	Fe	Cu	Mn	Mg	Zn	Sn
Al*	bal.	0.131	0.171	0.002	0.009	0.027	0	0.076
413 Al	bal.	13.659	0.490	0.215	0.035	0.039	0.07	0.089
Mg**	0	0.029	0.002	0.012	0.017	bal.	0.093	0

* Commercially pure aluminum.
** Commercially pure magnesium.

3.2. Immersion tests and determination of IHF

In order to evaluate the transient IHF between solid substrate and melt in Al/Mg couples, using the lumped capacitance method, immersion tests were conducted. A schematic sketch of the set up used for immersion tests is shown in Fig. 1.

In the immersion tests, a cylindrical solid aluminum insert with 15 mm diameter and 60 mm height was used as substrate, and the magnesium melt was allowed to solidify around it. The insert was instrumented with a K type thermocouple having a diameter of 1 mm. The thermocouple was calibrated at the melting point of pure aluminum exhibiting fluctuations of about 1.0 °C. The temperature of the solid insert during the solidification process was recorded at 0.2 s intervals by a computer-controlled data logger. The thermocouple was selected to have a time response considerably less than the interval between temperature readings. For carrying out the immersion tests, molten magnesium having a temperature of 680 °C was poured into the cylindrical cavity of a CO_2 sand mold with an internal diameter of 30 mm and an external diameter of 60 mm, which was preheated up to about 250 °C to avoid premature solidification of the melt. The cylindrical solid aluminum insert was degreased with acetone and immersed in the melt. The magnesium melt was allowed to solidify around the insert. Using the temperature history of the insert and applying the lumped capacitance method, the transient IHF between solid insert and melt was calculated for different couples of Al/Mg from Eq. (2).

Experiments were carried out for magnesium melt solidifying against pure aluminum and 413 aluminum alloy. In order to study the effect of substrate surface roughness on IHF between substrate and melt, the solid aluminum inserts were ground using 60, 80, 120, 220, 320, 400, 800, 1000 and 1200 silicon carbide abrasive papers and their arithmetical average surface roughnesses (R_a) were measured using a Surtronic 25-Taylor Hobson profilometer. At least five different areas in each sample were used for measurement of surface roughness. Surface morphology of the ground inserts also was observed using a Nanoscope III Digital Instruments atomic force microscope (AFM).

3.3. Microstructural analysis

To investigate microstructures at the interface of Al/Mg couples, the samples were sectioned in the transverse direction and after grinding and polishing were observed with a JEOL JSM-5600 scanning electron microscope (SEM).

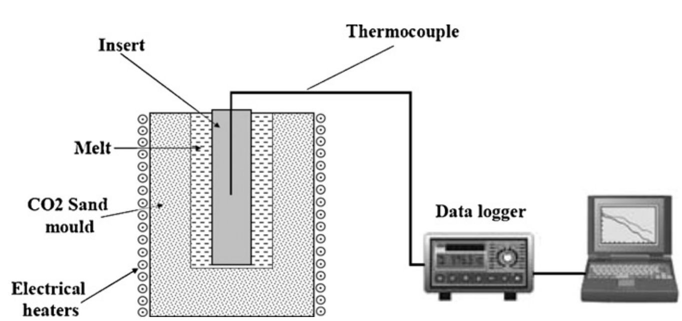

Fig. 1. Schematic sketch of the set up used for immersion tests.

Table 2
Thermophysical properties of materials used in this study [12,13].

Material	Density (kg m^{-3})	Specific heat (J kg^{-1} K^{-1})	Thermal conductivity (W m^{-1} K^{-1})
Al	2698	900	237
413 Al	2657	963	121
Mg	1738	1025	156

4. Results

The results of surface roughness measurements of used pure aluminum and 413 aluminum alloy inserts are presented in Table 3. Due to the different tribological manner of these two materials against silicon carbide abrasive papers, their surface roughnesses are different to some extent.

Fig. 2 shows two different aluminum insert surfaces after grinding by silicon carbide abrasive papers obtained by AFM in contact mode. These surfaces have different roughnesses, but almost exhibit identical grooved texture topographies.

Typical transient IHF measurement results for magnesium solidifying against two pure aluminum inserts with different surface roughnesses are shown in Fig. 3. As it can be seen in this figure, the IHF rises rapidly, reaches a maximum value and then decreases to low values. In these cases, the maximum IHF for pure aluminum inserts with 460 nm and 1040 nm surface roughnesses are 1334 kw/m^2 and 1694 kw/m^2, respectively.

The influence of surface roughness on maximum IHF for magnesium solidifying against pure aluminum and 413 aluminum alloy inserts is shown in Fig. 4. The values for maximum IHF in the diagram shown in this figure are the mean values of at least

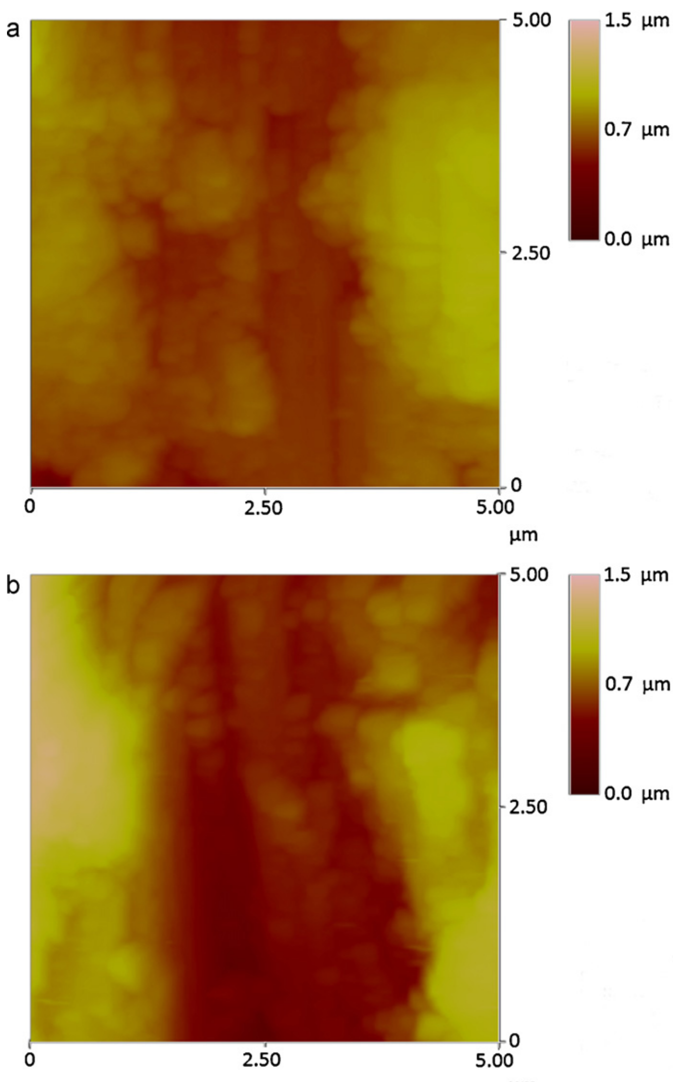

Fig. 2. Topography of the surface of aluminum inserts after grinding by different standard grit silicon carbide abrasive papers obtained by AFM in Contact Mode (a) R_a = 460 nm (b) R_a = 1040 nm.

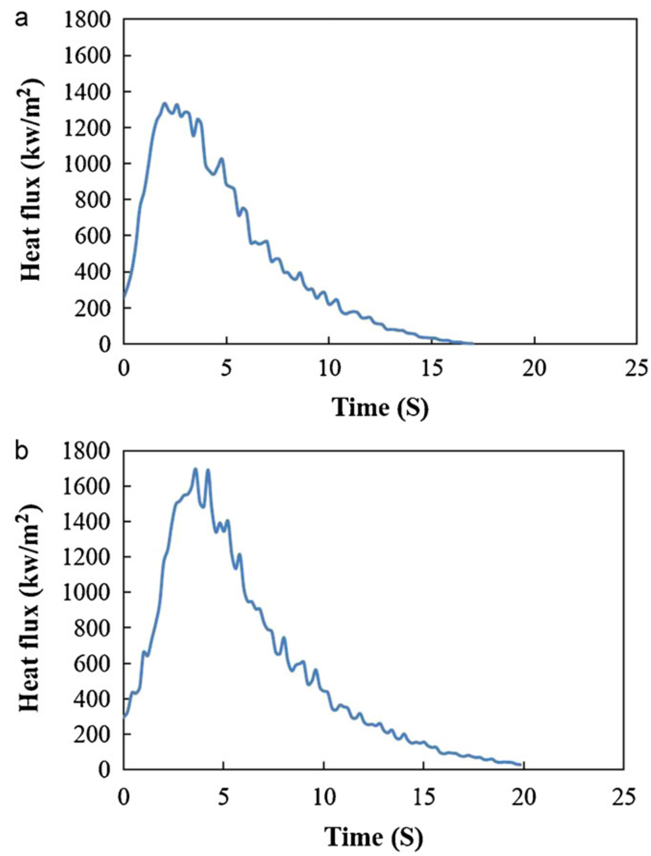

Fig. 3. Variation of IHF with time for magnesium solidifying against pure aluminum inserts for two different values of surface roughnesses. (a) R_a = 460 nm (b) R_a = 1040 nm.

Table 3
Surface roughness of the aluminum inserts after grinding by silicon carbide abrasive papers.

Substrate surface condition (silicon carbide abrasive paper standard grit #)	Surface roughness R_a (nm)	
	Pure Al	413 Al
60	2480	2680
80	2400	2450
120	2180	2070
220	1860	1630
320	1040	950
400	950	680
800	800	580
1000	690	290
1200	460	250

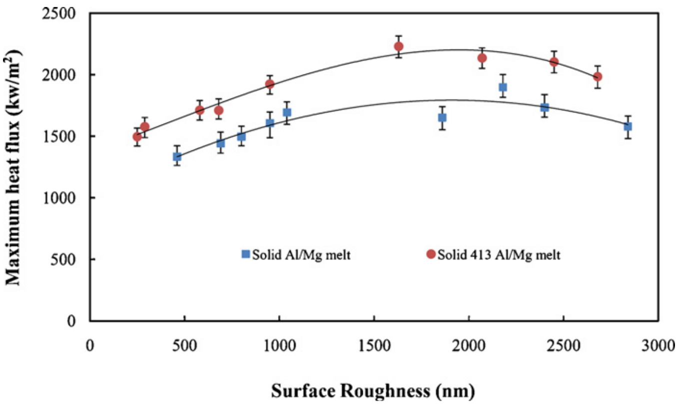

Fig. 4. Variation in maximum IHF with surface roughness for solid aluminum/magnesium melt couples.

three immersion experiments. However, the maximum IHF for solid 413 aluminum alloy/magnesium melt couples are higher than those measured for solid pure aluminum/magnesium melt couples, the variation of maximum IHF with surface roughness for solid pure aluminum/magnesium melt and solid 413 aluminum alloy/magnesium melt couples, almost shows similar trend and in both couples, with increasing surface roughness, the maximum IHF increases at first and after reaching a maximum value, starts to decrease.

Fig. 5 shows typical back scattered SEM micrographs of solid aluminum/magnesium melt interfaces. For both solid pure aluminum/magnesium melt and solid 413 aluminum alloy/magnesium melt couples, a reaction layer without any gap or hole has been formed at the interface of solid/melt during immersion experiments.

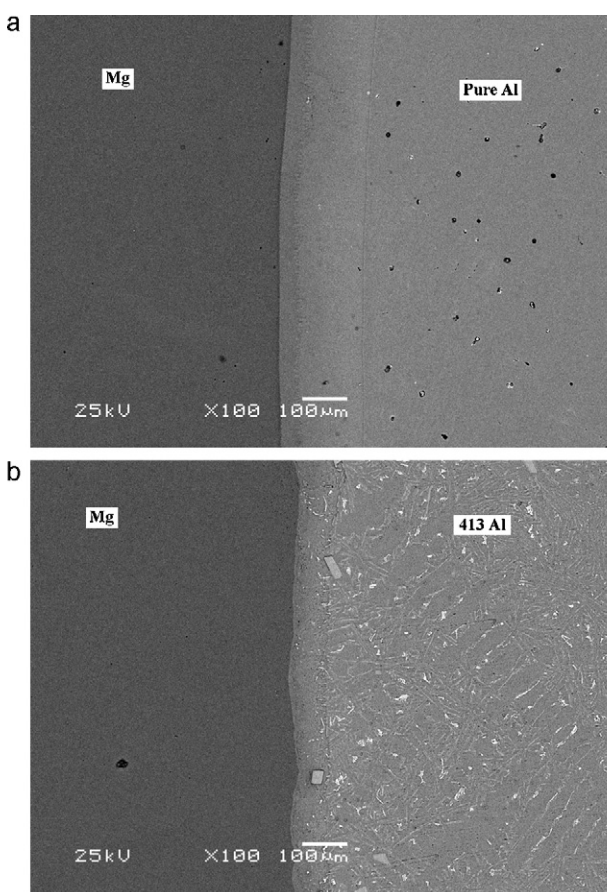

Fig. 5. Typical back scattered SEM micrographs of solid aluminum/magnesium melt interfaces (a) solid pure aluminum/magnesium melt (b) solid 413 aluminum alloy/magnesium melt.

5. Discussion

5.1. Effect of insert surface roughness

According to the graph shown in Fig. 4, the maximum IHFs for solid pure aluminum/magnesium melt and solid 413 aluminum alloy/magnesium melt couples have been affected by solid insert surface roughness. In these couples, with increasing surface roughness, the maximum IHF increases at first and when the surface roughness reaches a threshold value (about 2000 nm), the IHF starts to decrease. Many researchers have studied the effect of solid substrate surface roughness on IHF between substrate and melt for various solid/melt couples and have reported that in most of these couples, increasing the surface roughness has led to a decrease in IHF due to increasing the contact resistance at the interface of solid and melt [1,14–16]. The different tendency obtained in this research may be related to the wetting characteristics of aluminum inserts by magnesium melt. During the contact between magnesium melt and aluminum inserts, magnesium melt is expected to easily react with the thin aluminum oxide layer on the surface of aluminum inserts and reduce it due to negative standard free Gibbs energy of equation below [17]:

$$Mg_{(l)} + \tfrac{1}{3}Al_2O_3 \rightarrow MgO_{(s)} + \tfrac{2}{3}Al_{(l)} \quad \Delta G^0 = -39\,\text{kJ mol}^{-1}\ \text{of Mg at 1000 K} \qquad (4)$$

This can bring about straight contact between magnesium melt and fresh aluminum insert surface, followed by reaction between them and finally formation of intermetallic compounds like $Al_{12}Mg_{17}$ and Al_3Mg_2 [18] at the interface. The reaction layers between magnesium melt and aluminum inserts shown in Fig. 4, confirm the direct contact and the interaction between magnesium melt and aluminum insert surfaces. In this situation, due to the reaction between magnesium melt and fresh aluminum surface, magnesium melt can wet the surface of aluminum insert in good conditions and enter into its surface roughnesses and spread on it completely. Because of complete contact, IHF can be considered as a function of real area of contact (RAC) between melt and substrate. So if increasing the surface roughness leads to increasing the RAC, it can cause an increase in IHF; but if further increase in surface roughness leads to a decrease in RAC, it can cause a decrease in IHF. Some of researchers believe that in addition to surface roughness, the RAC is also a function of the surface texture [19]. Donoso et al. [20] have modeled the effect of surface roughness on RAC of the surfaces with grooved roughness morphologies (Fig. 6), and have reported that for these kinds of surface morphologies, with increasing surface roughness, RAC will increase at first, and then starts to decrease after reaching a maximum value.

Considering the similarities between the surface morphology of aluminum inserts used in this study (Fig. 2) and the supposed samples on their model (Fig. 6), it seems to be possible to generalize the results of this model for estimating the change in RAC of the used aluminum inserts with different surface roughnesses. So it seems that for the used pure aluminum and 413 aluminum alloy inserts, increasing the surface roughness up to about 2000 nm has led to an increment in RAC and IHF, but increasing too much more than this value on the surface roughness has led to a decrease both in RAC and IHF. The effect of surface roughness on RAC and IHF for two good wetting and poor wetting conditions between melt and substrate is shown schematically in Fig. 7.

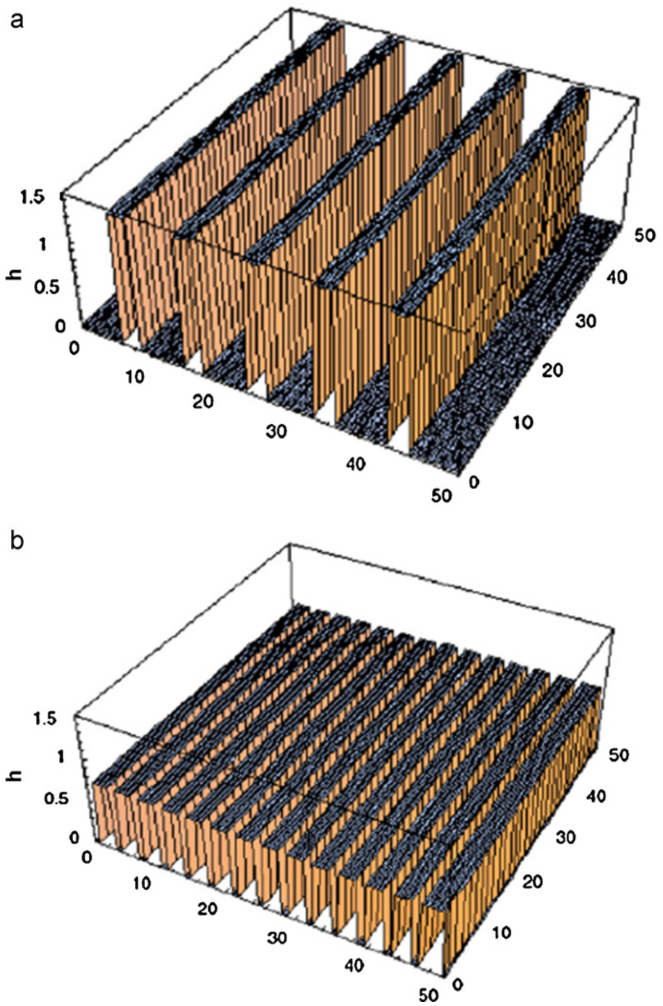

Fig. 6. Three-dimensional representation of the model used by Donoso et al. for determination of RAC (a) Roughness = 0.649, RAC = 65 (b) Roughness = 0.344, RAC = 71 [20].

5.2. Effect of substrate composition

As it can be seen in Fig. 4, at almost all the ranges of the surface roughnesses, the maximum IHFs for solid 413 aluminum alloy/magnesium melt couples are higher than those for solid pure aluminum/magnesium melt couples. This may have arisen from the different surface oxide characteristics of pure aluminum and 413 aluminum alloy. Upon exposure of pure aluminum to the atmosphere at room temperature, a film of about 2–3 nm thick of amorphous aluminum oxide (alumina) is formed on the aluminum surface [21], whereas for 413 aluminum alloy due to existence of a considerable amount of silicon element (about 13 wt%) within the alloy composition, in addition to alumina, silicon oxide also is formed on the surface of the inserts [22]. Besides that, due to the negative standard free Gibbs energy, magnesium melt can react with the silicon oxide on the surface of 413 aluminum alloy inserts, similar to reaction with aluminum oxide, and reduce it according to Eq. (5) [17]:

$$Mg_{(l)} + \tfrac{1}{2}SiO_2 \rightarrow MgO_{(s)} + \tfrac{1}{2}Si_{(s)} \quad \Delta G^0 = -128\,\text{kJ mol}^{-1}\ \text{of Mg at 1000 K} \qquad (5)$$

Considering Eqs. (5) and (4), it seems that, from the thermodynamic viewpoint, magnesium melt has higher tendency to react with silicon oxide than aluminum oxide due to more negative standard free Gibbs energy (-128 kJ mol^{-1} against -39 KJ mol^{-1}). Therefore considering the existence of some amounts of silicon oxide on the surface of 413 aluminum alloy, one can deduce that magnesium melt has a higher tendency to react with the surface of 413 aluminum alloy than pure aluminum, which can result in higher IHF for solid 413 aluminum alloy/magnesium melt couples in comparison with solid pure aluminum/magnesium melt couples.

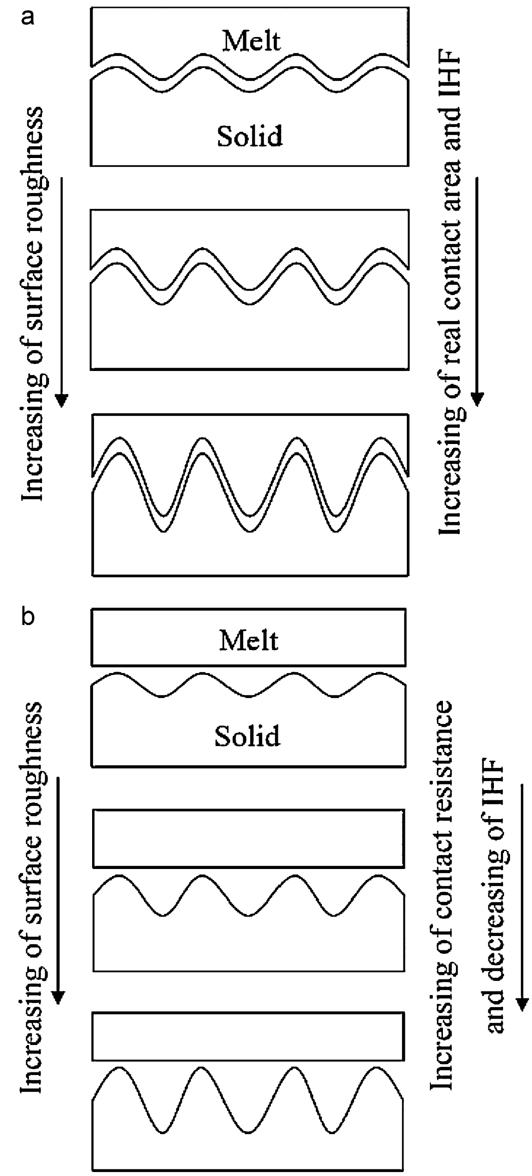

Fig. 7. Schematic representation of contact between melt and substrate for two different wetting conditions (a) good wetting (b) poor wetting.

Furthermore, the minimum contact angle between melt and substrate in reactive wetting conditions (θ_{min}), can be calculated using equation below [23]:

$$\cos\theta_{\min} = \cos\theta_0 - \frac{\Delta\gamma_r}{\gamma_{lv}} - \frac{\Delta G_r}{\gamma_{lv}} \qquad (6)$$

where γ_{lv} is the surface tension of the melt, θ_0 is the contact angle of the melt on the substrate in the absence of any reaction, and $\Delta\gamma_r$ takes into account the change in interfacial energies brought about by the interfacial reaction. ΔG_r is the change in free energy per unit area released by the reaction of the material contained in the immediate vicinity of the substrate/melt interface. Considering more negative standard free Gibbs energy of Eq. (5) than Eq. (4), it can be concluded that the term ΔG_r in Eq. (6) is also more negative for contact between 413 aluminum alloy and magnesium melt than between pure aluminum and magnesium melt. Therefore the contact angle in reactive wetting condition (θ_{min}) for solid

413 aluminum alloy/magnesium melt couples is predicted to be less than that for solid pure aluminum/magnesium melt couples. This means better wettability for 413 aluminum alloy by magnesium melt than pure aluminum. Consequently, higher IHF for solid 413 aluminum alloy/magnesium melt couples than solid pure aluminum/magnesium melt couples (Fig. 4), can be the result of better wetting conditions for solid 413 aluminum alloy/magnesium melt couples than solid pure aluminum/magnesium melt couples.

6. Conclusions

a) IHF for solid pure aluminum/magnesium melt and solid 413 aluminum alloy/magnesium melt couples is a function of surface roughness.
b) For both solid pure aluminum/magnesium melt and solid 413 aluminum alloy/magnesium melt couples, by increasing the surface roughness, the maximum IHF increases at first and after reaching a maximum value, starts to decrease.
c) Almost for all ranges of surface roughness, the maximum IHFs for solid 413 aluminum alloy/magnesium melt couples are higher than those for solid pure aluminum/magnesium melt couples. This seems to be due to the better wettability of 413 aluminum alloy than pure aluminum, by magnesium melt.

Acknowledgements

The authors would like to thank Dr. M. Reza Naimi-Jamal from the department of chemistry at Iran University of Science & Technology for AFM studies. The first author also acknowledges the Iran Ministry of Science, Research and Technology and Shahid Chamran University of Ahwaz for financial support.

References

[1] D. Bouchard, S. Leboeuf, J.P. Nadeau, R.I.L. Guthrie, M. Isac, Dynamic wetting and heat transfer at the initiation of aluminum solidification on copper substrates, J. Mater. Sci. 44 (2009) 1923–1933.
[2] L.A. Jacobson, J. McKittrick, Rapid solidification processing, Mater. Sci. Eng. R 11 (1994) 355–408.
[3] T. Loulou, E.A. Artyukhin, J.P. Bardon, Estimation of thermal contract resistance during the first stages of metal solidification process: II – experimental setup and results, Int. J. Heat Mass Trans. 42 (1999) 2129–2142.
[4] T. Evans, L. Strezov, Interfacial heat transfer and nucleation of steel on metallic substrates, Metall. Mater. Trans. B 31 (2000) 1081–1089.
[5] K.N. Prabhu, B.N. Ravishankar, Effect of modification melt treatment on casting/chill interfacial heat transfer and electrical conductivity of Al-13% Si alloy, Mater. Sci. Eng. A 360 (2003) 293–298.
[6] D. Emadi, J.E. Gruzleski, J.M. Toguri, The effect of Na and Sr modification on surface tension and volumetric shrinkage of A356 alloy and their influence on porosity formation, Metall. Trans. B 24 (1993) 1055–1063.
[7] S. Nyamannavar, K.N. Prabhu, Thermal contact at solder/substrate interfaces during solidification, Mater. Sci. Tech. 25 (2009) 707–710.
[8] J.P. Holman, Heat Transfer, ninth ed., McGraw-Hill, New York, 2002.
[9] F.P. Incropera, D.P. Dewitt, Fundamentals of Mass and Heat Transfer, sixth ed., John Wiley & Sons, New York, 2007.
[10] C.S. Hsien, S.C. Long, Analysis of interfacial heat transfer coefficient of green sand mold casting for aluminum and tin-lead alloys by using a lump capacitance method, J. Heat Trans. 129 (2007) 595–600.
[11] I.O. Mohamed, An inverse lumped capacitance method for determination of heat transfer coefficients for industrial air blast chillers, Food Res. Int. 41 (2008) 404–410.
[12] J.R. Davis, Aluminum and aluminum alloys, in: ASM Specialty Handbook, first ed., ASM International, Materials Park, Ohio, 1993.
[13] M.M. Avedesian, H. Baker, Magnesium and magnesium alloys, in: ASM Specialty Handbook, first ed., ASM International, Materials Park, Ohio, 1998.
[14] G.X. Wang, E.F. Matthys, Experimental determination of the interfacial heat transfer during cooling and solidification of molten metal droplets impacting on a metallic substrate: effect of roughness and superheat, Int. J. Heat Mass Trans. 45 (2002) 4967–4981.
[15] W.D. Griffiths, R. Kayikci, The effect of varying chill surface roughness on interfacial heat transfer during casting solidification, J. Mater. Sci. 42 (2007) 4036–4043.
[16] B. Coates, S.A. Argyropoulos, The effects of surface roughness and metal temperature on the heat-transfer coefficient at the metal mold interface, Metall. Mater. Trans. B 38 (2007) 243–255.
[17] Reaction-Web, Fact-Web Programs, http://www.crct.polymtl.ca/factweb.php.
[18] Alloy phase diagrams, ASM Handbook, vol. 3, ninth ed., ASM International, Materials Park, Ohio, 1995.
[19] P.J. Ramon-Torregrosa, M.A. Rodriguez-Valverde, A. Amirfazli, M.A. Cabrerizo-Vilchez, Factors affecting the measurement of roughness factor of surfaces and its implications for wetting studies, Colloids Surf. A 323 (2008) 83–93.
[20] M.G. Donoso, A. Mendez-Vilas, J.M. Bruque, M.L. Gonzalez-Martin, On the relationship between common amplitude surface roughness parameters and surface area: Implications for the study of cell-material interactions, Int. Biodeterior. Biodegrad. 59 (2007) 245–251.
[21] Y. Li, L. Wang, Study of oxidized layer formed on aluminium alloy by plasma oxidation, Thin Solid Films 517 (2009) 3208–3210.
[22] C.R. Werrett, D.R. Pyke, A.K. Bhattacharya, XPS studies of oxide growth and segregation in aluminium-silicon alloys, Surf. Interface Anal. 25 (1997) 809–816.
[23] L. Espie, B. Drevet, N. Eustathopoulos, Experimental study of the influence of interfacial energies and reactivity on wetting in metal/oxide systems, Metall. Trans. A 25 (1994) 599–605.