Effect of interfacial-active elements addition on the incorporation of micron-sized SiC particles in molten pure aluminum

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Abstract

Ceramic particles generally have poor wettability by liquid metal, leading to a major drawback in fabrication of cast metal matrix composites (MMCs). In this work, the effect of 1 wt. % of Ca, Mg, Si, Ti, Zn and Zr interfacial-active alloying elements was studied on the incorporation of micron-sized SiC particles into the molten pure aluminum using the vortex casting method at 680 1C. The results indicated that Ti, Zr, Zn and Si were not positively effective in improving particulate incorporation, while Ca and especially Mg were very efficient at increasing the incorporation of ceramic particles into the molten Al. Also, it was revealed that Al_3Ti, and Al_3Zr intermetallic phases were formed for samples containing Ti and Zr, making hybrid MMCs with a higher amount of hardness. Finally, it was found that a reaction layer between Al and SiC particles was formed at the Al/SiC interface for all of the samples, expect for the ones containing Si and Ti, indicating that for most of the samples at 680 1C an exothermic reaction took place between the Al and SiC particles.

Keywords: Metal-matrix composites (MMCs); Ceramic particle; Microstructures; Casting

1. Introduction

Aluminum metal matrix composites (AMMCs) have gained significant attention in the recent years. This is primarily due to their lightweight, low coefficient of thermal expansion, machinability, improved mechanical properties such as 0.2% YS, UTS, and hardness. Due to these advantages, they are used in aerospace industries (airframe and aerospace components), automobile industries (engine pistons), and electronic components [1–8].

Many techniques have been developed for producing particulate reinforced AMMCs, such as powder metallurgy and squeeze casting. Besides the fact that each of these methods has its own advantages and disadvantages, they are all relatively expensive. Nowadays, researchers are focusing on producing low-cost composites. Stir casting (vortex technique) is generally accepted as a commercial low-cost method. Its advantages lie in its simplicity, flexibility, and applicability to large volume production. This process is the most economical of all the available routes for AMMCs production and allows very largesized components to be fabricated. However, methods of achieving the following in stir casting are: (i) no chemical reaction between the reinforcement material and matrix alloy, (ii) low porosity in the cast

AMMCs, (iii) wettability between the two main substances, and (iv) achieving a uniform distribution of the reinforcement material [3–17].

It was proposed in Hashim et al. that the percentage of SiC incorporated within the solidified composite is indicative of the success with which wetting was achieved [11]. They used twodimensional micrographs of sections through a composite to compare the SiC content and measure the percentage wetting. This method was used in their study to evaluate the effect of interfacial-active elements on the incorporation and wettability of SiC particles.

The addition of alloying elements can modify the matrix metal alloy by producing a transient layer between the particles and the liquid matrix. This transient layer has a low wetting angle, decreases the surface tension of the liquid, and surrounds the particles with a structure that is similar to both the particle and the matrix alloy [13,14,19,20].

Previous authors [21–27] have used chemically activated wetting method to fabricate aluminum matrix composite reinforced with ceramic particles. They reported that Mg, Ca, Ti, Si, Zr, and Zn could be used as a wetting agent to ease the incorporation of ceramic particles by molten aluminum. However, to the best of our knowledge, no attempt has been made to study the presence of these elements simultaneously on the incorporation of ceramic particle in a molten aluminum in a study with constant conditions for a distinct comparison.

In this study, the effect of 1 wt. % of Ca, Mg, Si, Ti, Zn and Zr alloying elements was studied on the incorporation and wettability of micron-sized SiC particles in the molten aluminum to compare their effect in this regard during AMMCs fabrication.

2. Experimental procedures

Aluminum ingot with 99.8 wt. % commercial purity was used as the matrix. Table 1 tabulates the chemical composition used. As it can be seen, the amount of Si and Fe was almost negligible.

Micron-sized SiC particles with an average particle size of 80 mm and 99.9% purity was supplied as the reinforcement of metal matrix composite. The morphology of silicon carbide particles used in this study is shown in Fig. 1.

About 1 g SiC powders were inserted in an aluminum foil by forming a packet to fabricate a composite with 3 wt. % SiC (E 2.534 vol. % SiC) as reinforcement. The pure aluminum as well as the Al–Ti, Al–Zr, Al–Mg, Al–Si, Al–Zn and Al–Ca mother alloys were separately heated up to 680 ^{0}C using a bottom-pouring system to fabricate six various alloys containing 1 wt. % alloying element. The graphite stirrer was placed below the surface of melt and rotated with a speed of about 500 rpm and simultaneously argon gas with a high purity was blown to the melt surface (see Fig. 2).

Table 1
The chemical composition (in wt. %) of pure aluminum used in this study.

Al (%)	Si (%)	Fe (%)	Cu (%)	Mn (%)	Zn (%0	Ni (%)	Pb (%)	Sn (%)
99.8	0.1	0.04	0.01	0.01	0.02	0.01	0.005	0.004

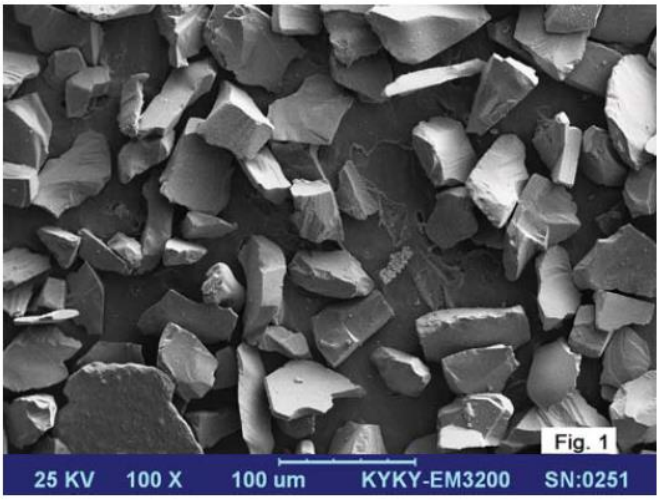


Fig. 1. The morphology of SiC particles that were used as reinforcement.

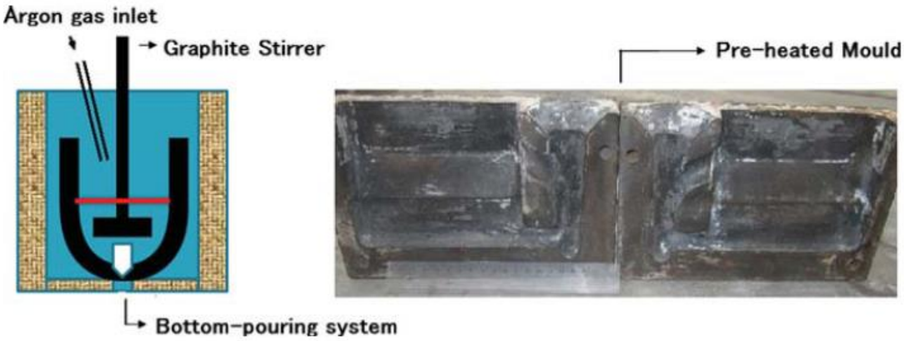


Fig. 2. The experimental set-up used in this study: (a) schematic of the vortex casting set-up and (b) carbon steel mold.

The packets were added to the vortex center and the stirring was continued for about 6 min. Composite slurry was poured into a preheated cast iron mold (at 450 1C). For comparison, a composite was produced containing 3 wt. % SiC without any alloying elements (pure aluminum as the matrix). Microstructural investigations were performed using two kinds of scanning electron microscopes (SEM, Cam Scan Mv2300, equipped with EDAX analysis and SEM, KYKY-EM3200) and an optical microscope. For this purpose, the samples were polished and etched with Keller's reagent (190 ml water, 5 ml HNO3, 3 ml HCl, and 2 ml HF). Microhardness tests were conducted according to ASTM E384 using an applied load of 25 g at 15 s time duration and the results were reported for at least 10 parts of a sample (5 parts in the matrix and 5 parts near the ceramic particles).

3. Results and discussion

3.1. SEM and optical microscopy (OM) studies

Fig. 3a shows a SEM microstructure of Al–3 wt. % SiC without any alloying elements, indicating that a very low amount of ceramic particles were incorporated into the molten metal. This figure indicates that even 80 mm SiC particles do not have enough wettability by molten aluminum.

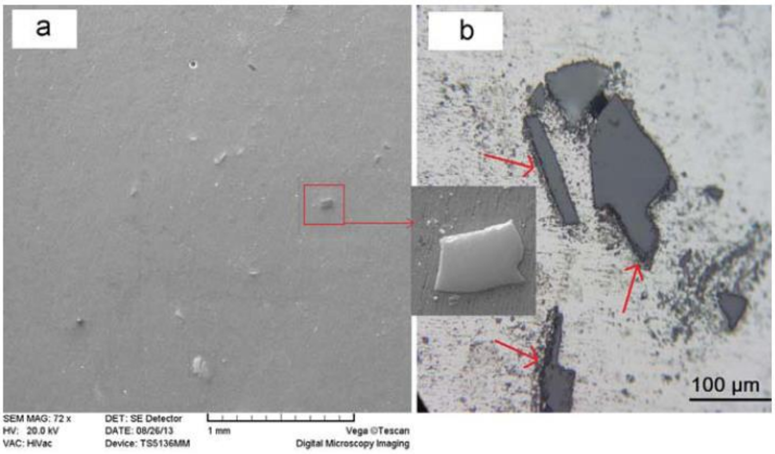


Fig. 3. The microstructures of a composite with the chemical composition of Al–3 wt. % SiC: (a) SEM image and (b) high-magnification optical image.

Fig. 3b is a high-magnification optical image of this sample, revealing that a reaction took place between aluminum and SiC at 680 ^{0}C, although a more or less clean interface without any microvoid formation was obtained. The coefficient of thermal expansion (CTE) mismatch between matrix and reinforcement can cause the formation of microvoids or cracks at the particle–matrix interfaces [18]. However from SEM examination it is clear that a good crack free bonding was formed at discrete locations between the reinforcement and the matrix alloy.

The addition of alloying elements to the Al matrix has been reported to be a suitable method for improving the wettability of ceramic and metal [13]. Fig. 4 shows a microstructure of a composite with a composition of Al–1 wt. % Mg–3 wt. % SiC.

Fig. 4a reveals the incorporation of high amount of ceramic particles into the molten aluminum, indicating that Mg is very effective in wettability improvement of ceramic particles by molten aluminum compared to alloy compositions without Mg. Agglomeration took place in some parts and a reaction layer could be seen around ceramic particles, while a good distribution of ceramic particles was revealed. Fig. 4b shows a SEM image of this sample, revealing that porosity defects were formed around the ceramic particles, between the agglomerated particles and also in the matrix. A weak bonding could be observed at metal/ceramic interface, indicating that an external pressure seems to be needed during solidification for this sample in order to increase interface bond strength. Line EDAX analysis of this sample is shown in Fig. 4c, indicating that Mg is present in the bulk alloy around the ceramic particles.

Fig. 5 shows the microstructure of a composite which contained 1 wt. %. Ca. As can be seen in Fig. 5a, the sampleswith Ca did not show significant increase in re-enforcement, while other literature [13,19,20] has reported that Ca addition is highly effective in this regard. The ceramic agglomeration is obvious and a thin reaction layer could be seen around the ceramic particles. The interface quality (see Fig. 5b) is better than the previous sample, although a shrinkage cavity and air gaps between the agglomerated ceramic particles could be observed for this sample.

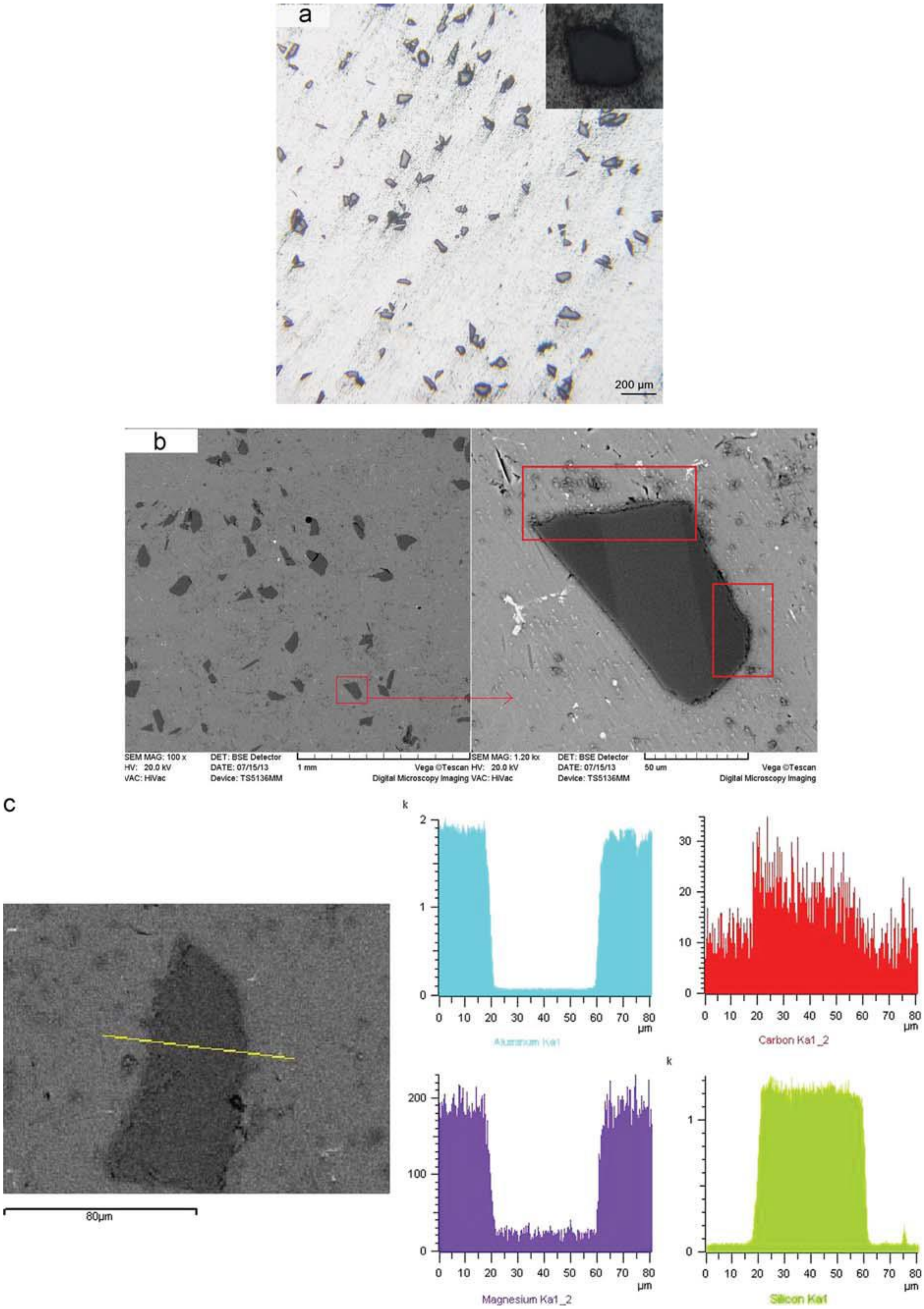


Fig. 4. (a)Microstructures of a composite containing Mg, low and high magnification optical image , low and high magnification SEM image (b), and line EDAX analysis of this composite (c).

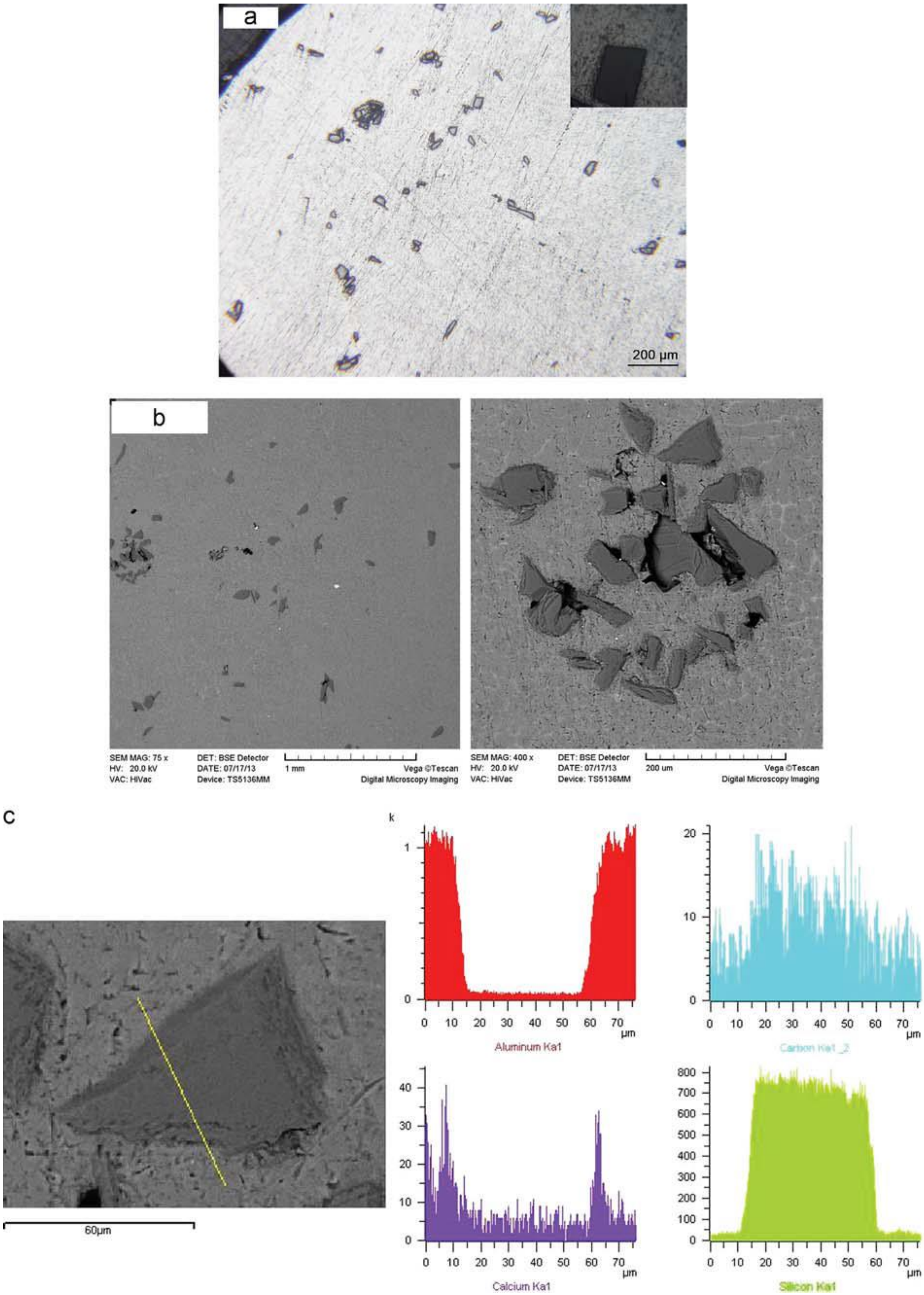


Fig. 5. The microstructures of a composite containing Ca, optical image (a), SEM image (b), and line EDAX analysis (c).

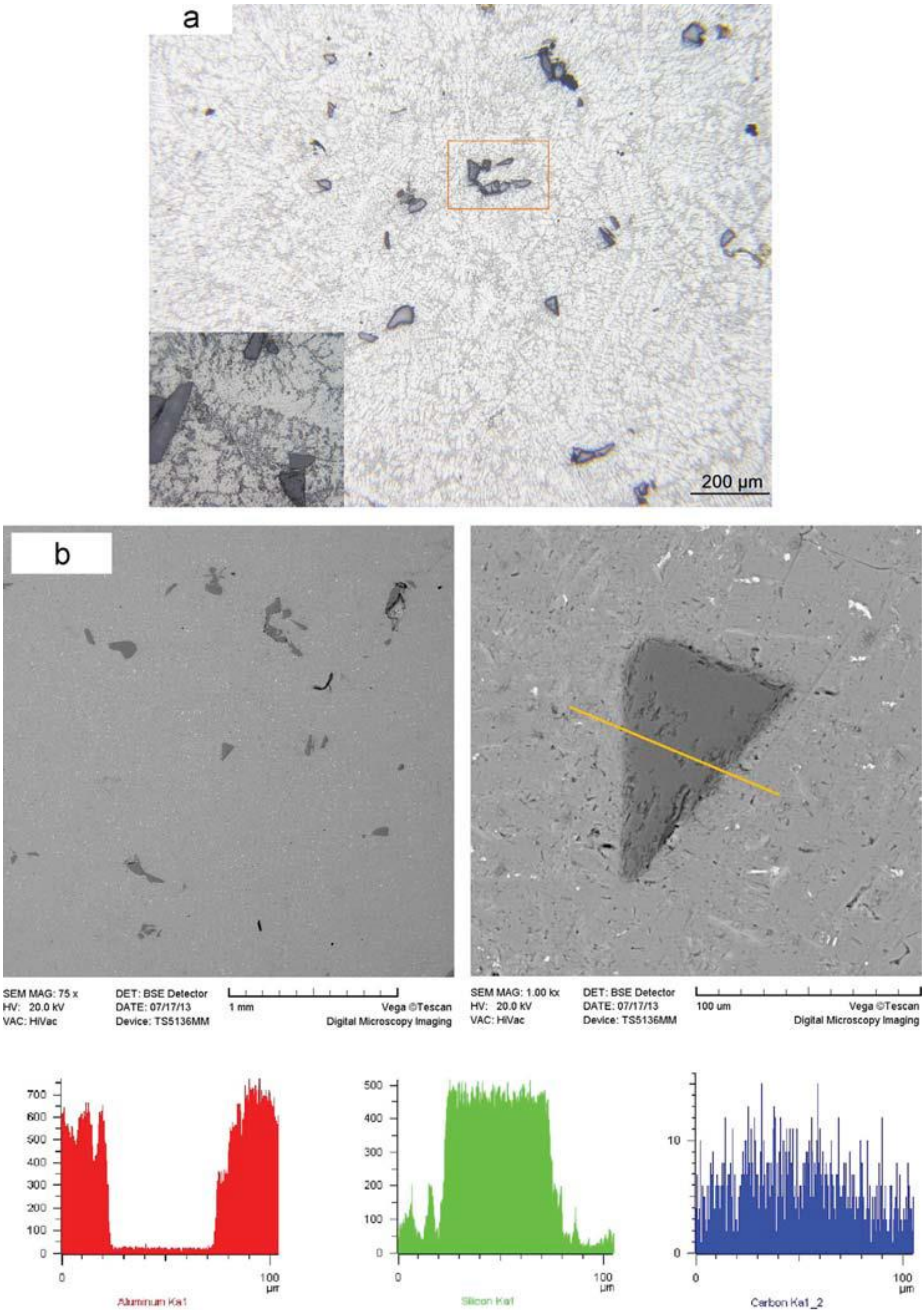
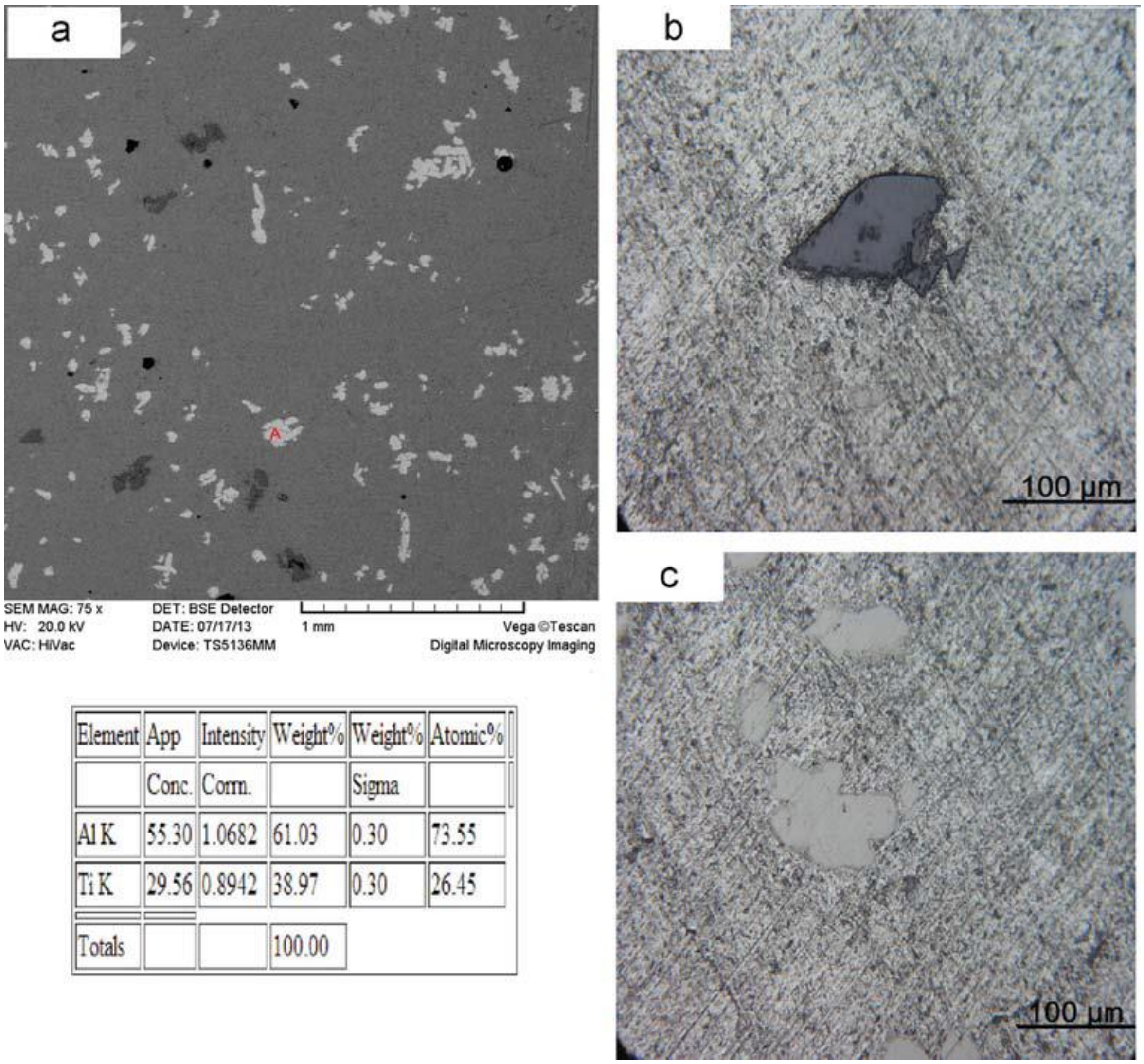


Fig. 6. The microstructure of a composite containing silicon, optical image (a), SEM image as well as EDAX analysis (b).

Although literature [21,23,24] reported that silicon could increase the wettability of ceramic particles in molten aluminum, no considerable wettability improvement was observed for a sample containing silicon (see Fig. 6). As SiC particles have a particle size larger than 60 mm, therefore, it seems that even a low value of solidification rate (as the mold temperature was pre-heated at about 450 ^{0}C) could not highly change the distribution of particles after solidification, although orange color rectangle (see Fig. 6a) shows some SiC particles, which seem having been redistributed during solidification, which might be pushed ahead of the solidification front. This figure also indicates that no reaction layer was formed around the ceramic particles and a clean interface could be observed between ceramic and metal. Fig. 6b is a SEM image of this sample shows that porosities are present in some parts of this sample. EDAX analysis for this sample indicates that a SiC particle is embedded in an Al–Si alloy.

Titanium is an element, which acts as a grain refiner in aluminum alloys [28] and it was reported [13] that the presence of this element is also helpful with regard to wettability improvement. Fig. 7 shows the microstructure of a sample containing titanium.



Element	App	Intensity	Weight%	Weight%	Atomic%
	Conc.	Corrn.		Sigma	
Al K	55.30	1.0682	61.03	0.30	73.55
Ti K	29.56	0.8942	38.97	0.30	26.45
Totals			100.00		

Fig. 7. The microstructure of a composite containing titanium; SEM image as well as point EDAX analysis of point A (a), high magnification optical image around a SiC particle (b), and a high magnification optical image around an Al3Ti particle (c).

Fig. 7a indicates that few ceramic particles (mostly in agglomerated form) were incorporated in the Al–1 wt. % Ti alloy, meaning that this element is not effective in this regard.

This figure shows the formation of some other phases (white in color) beside the SiC particles. An EDAX analysis was used to detect the chemical composition of these phases. As it can be seen from the EDAX analysis (see point A in Fig. 7a), Al and Ti were detected, while the atomic percent of Al was almost three times higher than that of Ti, indicating that these phases might be Al_3Ti intermetallic compound, making a hybrid composite with two kinds of reinforcements.

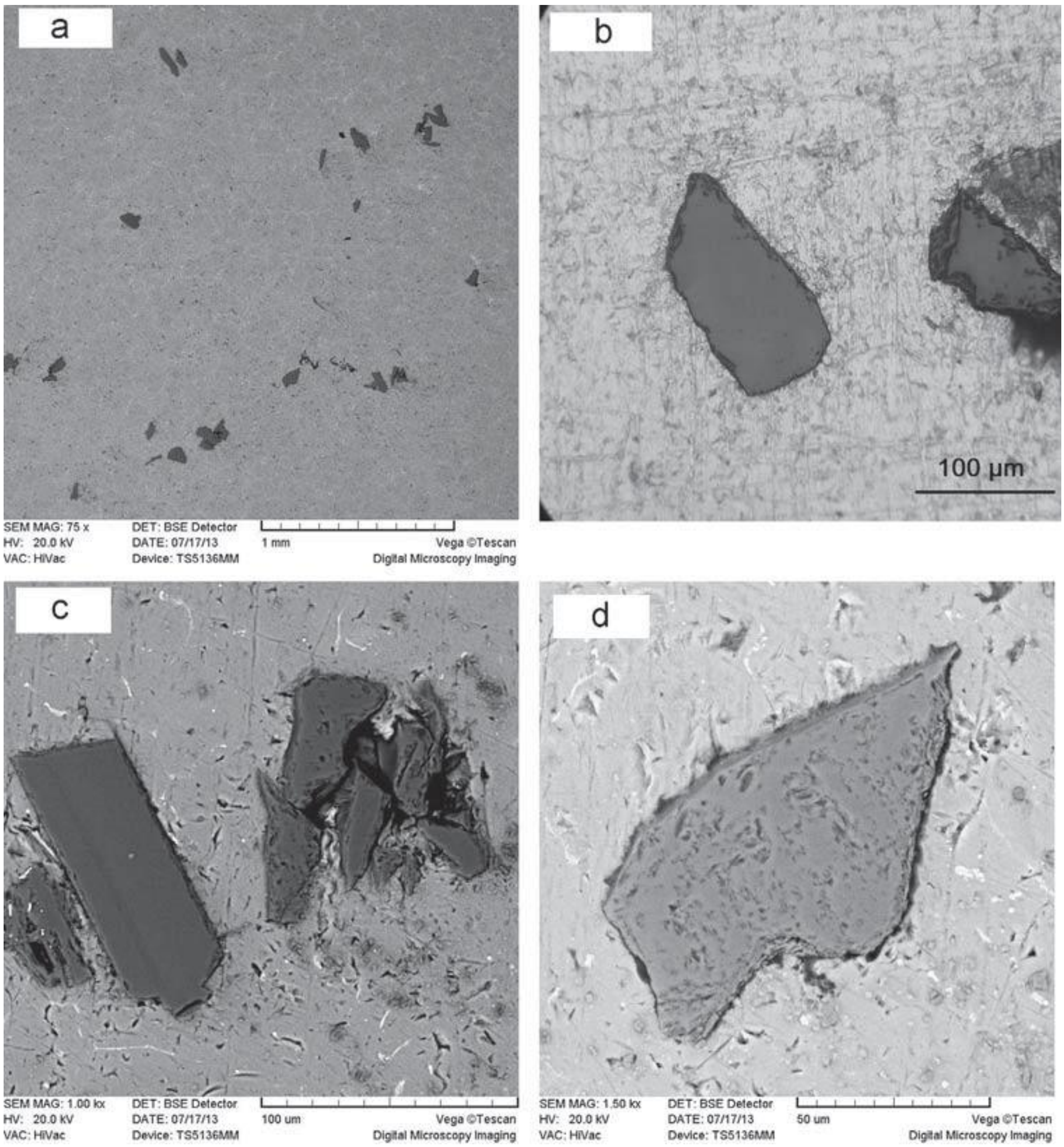
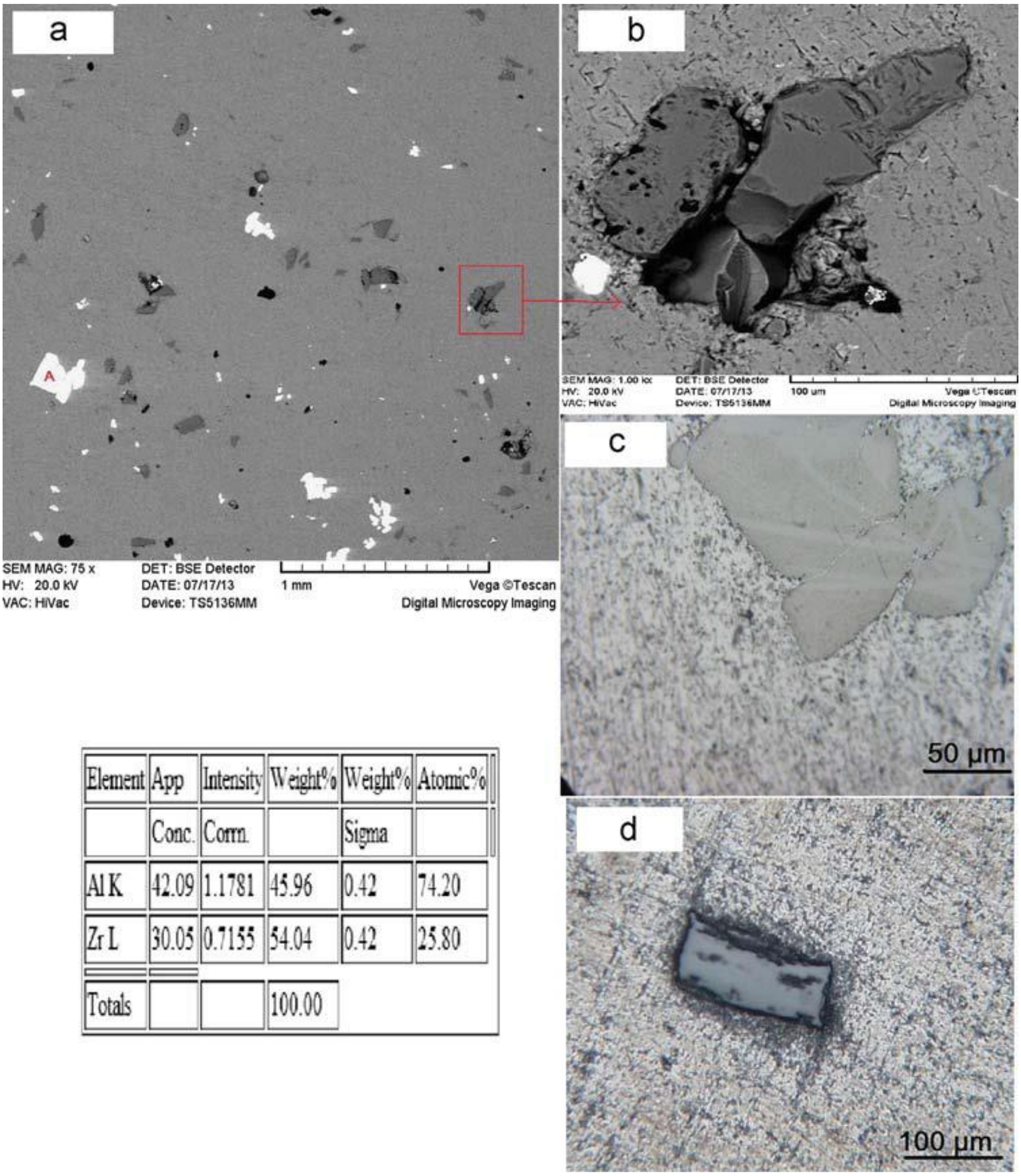


Fig. 8. The microstructure of a sample containing zinc, low magnification SEM image (a), OM image (b), and high magnification images (c and d).

The amount of this intermetallic phase is appreciable, although in some parts they are present in agglomerated form. Fig. 7b and c show the interfaces of a SiC and an intermetallic phase with the Al matrix for this sample, revealing that both of them, especially the intermetallic phase (see Fig. 7c) have a relative clean interface with the matrix alloy. It was reported in literature that silicon addition could avoid the reaction between Al and SiC [9].

Zn is a well-known alloying element in aluminum alloys. The presence of this element and its effect on the wettability of ceramic particles in molten aluminum would be worthful to study.



Element	App	Intensity	Weight%	Weight%	Atomic%
	Conc.	Corrn.		Sigma	
Al K	42.09	1.1781	45.96	0.42	74.20
Zr L	30.05	0.7155	54.04	0.42	25.80
Totals			100.00		

Fig. 9. The microstructure of a sample containing zirconium, low magnification SEM image as well as EDAX analysis of point A (a), high magnification SEM image (b), OM images of interface between Al3Zr with matrix (c) and SiC with matrix (d).

Fig. 8 shows the microstructure of a composite with a matrix of Al–1 wt. % Zn. Fig. 8a indicates that zinc is not powerful like Mg or Ca to ease the injection of ceramic particles. In fact, this element might not highly reduce the contact angle θ between matrix and SiC particles. No reaction layer around the SiC particles was revealed for this sample (see Fig. 8b), while a more precise SEM study (see Figs. 8c and d) indicates that a narrow gap is present around the SiC particles, which such gaps might lead to debonding and fracturing during mechanical tests.

As the ceramic particles were not pre-heated in this study, the formation of an air gap around the particles in this study might be due to the gas layers, which are always present on the ceramic particles [1,6,7,10,14].

Fig. 9 shows the microstructure of a sample containing Zr. Fig. 9a demonstrates that Zr has a positive effect like all the previous elements (not as much as Mg and Ca) with regard to the wettability improvement of SiC particles by molten aluminum. However, the presence of porosities and the Al_3Zr intermetallic phase is also evident for this sample. The presence of micro-pores in the SiC particles as Fig. 9b shows is strange and the authors could not submit any reason in this regard. However, a very clean and matched interface was obtained between Al_3Zr and matrix (see Fig. 9c) and a reaction layer was found around the ceramic particles for this sample (see Fig. 9d). It is very important to note that the casting process has many parameters and some of them might affect the pores formation and metal/ceramic interface in addition to the effects from the presence of alloying elements.

Fig. 10 shows the effect of interfacial-active elements on the incorporation and wettability of SiC particles by molten pure Al. As this figure shows, the most effective elements on the incorporation of ceramic particles are Mg, Ca, Si, Zn, Zr, and Ti, in decreasing order. The full details of this method, which could quantitatively make a connection between the calculated areas of SiC particles to the wettability was published by Hashim et al. [11].

In accordance with the Ellingham diagram [29], MgO and CaO are more stable than Al_2O_3, leading to dissolution of the alumina layer, which is the main factor of reduced wettability. Therefore, it is expected that Mg and Ca could be more effective than the other elements to improve the wettability of the SiC particles by the matrix alloy.

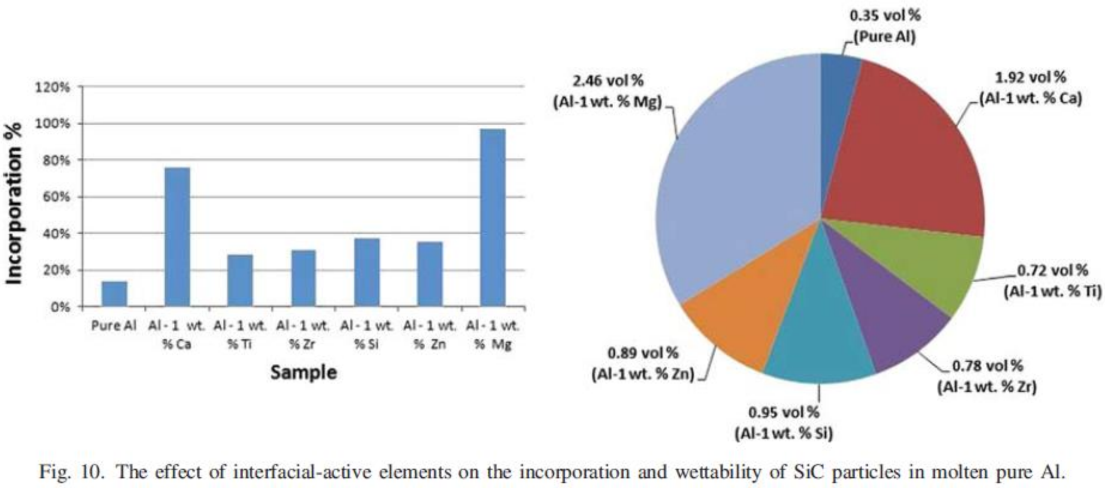


Fig. 10. The effect of interfacial-active elements on the incorporation and wettability of SiC particles in molten pure Al.

3.2. Microhardness measurements

During the thermal expansion of MMCs, internal stresses are developed around the particles due to a difference in the CTE of the matrix and the particle, and they are relieved by the formation and movement of dislocations [18]. However, the formation of porosities would lead to crack initiation during local plastic deformation, leading to a reduction in hardness value. In this regard, ten random points were selected for each sample for microhardness measurements (Vickers). Five points were selected in the matrix of the sample (some places that do not contain SiC particles) and five random points were selected around the SiC particles.

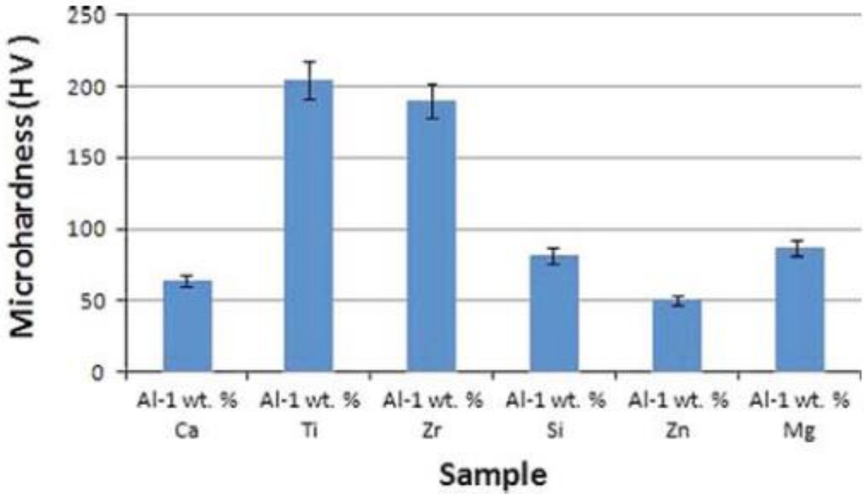


Fig. 11. Values of Vickers microhardness test for casting samples.

Fig. 11 shows the microhardness values of all the samples. As it can be seen, the presence of SiC particles, which entered into the matrix and also the formation of intermetallic phases are found to be more effective than porosity effect during microhardness test. The samples containing Ti and Zr have high microhardness values due to the intermetallic formation. Fig. 7 justifies that a higher value of intermetallic phases was present for Al–1 wt. % Ti rather than a sample containing Zr (see Fig. 9a), and therefore, a higher hardness value was obtained for this sample. Also, it seems that eutectic like morphology formation of a sample containing Si caused this sample to have a higher microhardness value compared to a sample containing Ca. This figure indicates that even 1 wt. % of alloying elements has a very sharp effect on the mechanical properties of metal matrix composites.

4. Conclusions

1. Ca and especially Mg are the most effective alloying elements, which could ease the dispersion of ceramic particles into the molten aluminum.
2. Porosities were observed for all the samples, showing that their formation is inevitable after casting, especially when no external pressure was used during solidi fication.

3. Intermetallic phases of Al_3Ti and Al_3Zr were formed during stir casting when Ti and Zr were added to the matrix, leading to a considerable increase in hardness of the composites.
4. In-situ intermetallic phases which are formed during casting produce a very clean and defectless interface with matrix.
5. A reaction layer at the Al/SiC interface was observed not for all of the samples, meaning that even 1 wt. % of interfacial-active element could affect the quality of matrixreinforcement interface.

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