Feasibility of the Purification of Al_2Cu Intermetallic Compound via Zone Melting

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Abstract

Intermetallic compounds constitute a unique class of metallic materials that have potential to be new-generation materials for structural and functional applications. Due to the high interest and potential performance in eutectic composites and other Al-based material, Al_2Cu intermetallic compound is one of them. It is well known that trace impurities have a significant impact on the performance of crystalline substances. Higher purity is required to meet demand, particularly in the development of intermetallic compounds in the fields of magnetic, electrical, optical, semiconducting, and superconducting materials. The study of the impurities in Al_2Cu intermetallic compound is of great importance for the application of aluminum-based materials. However, there have been few reports on the purification of intermetallic compounds. According to highly promising efficiency of impurities removal via fractional crystallization technique, in this research, zone melting as one of the methodologies based on this technique, is applied. Here, the impurity behavior during the purification of Al_2Cu intermetallic compound is investigated. Experimental parameters such as zone movement speed, zone length and number of passes are optimized to achieve a reduction in impurity concentration. Analysis of impurity desorption/migration behavior revealed that impurities calcium (Ca), iron (Fe), magnesium (Mg), silicon (Si) and zinc (Zn) accumulated at the end of the ingot. This work systematically demonstrates the feasibility of direct purification of intermetallics and provides a basis for applying this process to other intermetallic compounds.

1 Introduction

Intermetallic compounds (IMCs) are materials composed of a combination of different metals that exhibit an ordered crystal structure, when the concentration of the alloy exceeds the solubility limit.

For example, the yield strength, a unique property derived from the long-range ordered crystal structure, increases with increasing temperature before attainment the melting temperature [1]. Unlike common metals, IMCs can contain metallic, covalent and ionic bonds. Due to the mixed bonding, they have different crystal structure and properties than the base metals [2]. Due to its excellent mechanical properties, IMCs are considered as an alternative to superalloys and are of particular interest for demanding applications in the aerospace and automotive industries [3]. In addition, intermetallic phases have attracted much attention for their potential in functional applications, such as superconducting materials, hydrogen storage materials, magnetic sensors, actuators and corrosion-resistant coatings [4].

Based on the effects of trace impurity elements on various properties of different materials [5-7], purity is important for further applications of IMCs in structural and functional materials. For example, high-purity NiAl has better dislocation mobility, which helps to hide potentially detrimental defects and cracks. A high dislocation density improves the tensile ductility of single crystals and doubles the fracture toughness of polycrystalline alloys [8]. The fracture toughness of commercial NiAl is significantly reduced upon cooling from 473 K to room temperature and annealing at that temperature. The same heat treatment dependence is not observed for high-purity NiAl [9]. Moreover, residual resistance (a rough index of sample purity and overall qual) of high purity TiAl samples is much lower than that of the commercial pure grades [10]. In addition, Al-Al_2Cu is one of the most intensively studied eutectic composites in the literature [11]. As Al_2Cu shows promising physical and mechanical properties, the understanding of Al_2Cu knowledge system is important for the development of Al-based materials. Therefore, in this study, the influence of zone melting methods on impurities in Al_2Cu is investigated.

2 Methods

2.1 Zone melting technique

The general term zone melting refers to a process for controlling the distribution of soluble impurities or solutes in crystalline materials. Zone meting can be applied to any crystalline material that can be safely melted and has a difference in impurity concentration between the liquid and the freezing regions [12]. A very useful parameter for discussing such issues is the distribution coefficient, K. It is defined as the concentration ratio of the solute in the solid to that in the liquid (C_S/C_L). Impurities with $K > 1$ tend to be incorporated into the base metal during crystallization. On the other hand, at $K < 1$, impurities are repelled from solidifying area, migrate with the molten zone and accumulate at the end of the bar. In rare cases, where the impurity has a $K = 1$, there is no segregation and the impurity uniformly distributed throughout the matrix. As shown in Figure 1, the molten zone passes through the charge in one direction. Impurities become concentrated in one or the other end of the bar, thereby purifying the remainder.

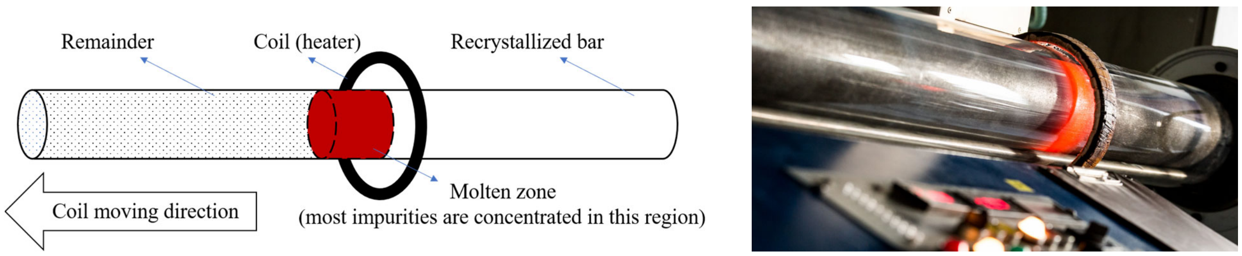


Figure 1: The sketch of the zone melting process with a single heater (left side) and the zone melting setup in IME/RWTH Aachen (right side)

2.2 Experimental procedures

To perform the melting process, high-purity aluminum and copper were melted to produce Al_2Cu ingots. Their stoichiometric proportions were of 45.92 wt.% and 54.08 wt.% for aluminum and copper, respectively. Two individual ingots were casted with different initial purities in an induction furnace by using the following raw materials:

- Ingot 1 – Al_2Cu: 99.998 % Al + 99.98 % Cu
- Ingot 2 – Al_2Cu: 99.98 % Al + 99.98 % Cu

Figure 1 (below side) shows the zone melting setup, established at IME/RWTH Aachen. The chamber pressure was maintained at 300 mbar by evacuating the quartz tube and rinsing with argon gas three times. The cast ingots were subjected to three zone melting passes. During zone melting, the pressure inside the chamber rose from 300 to 400 mbar maybe due to thermal expansion of the inert gas and volatilization of humidity and/or trace substances. The entire zone melting process was conducted automatically, with adjustable parameters for voltage, coil moving speed, number of passes and the start and end positions of the coil. The experimental parameters are listed in Table 1. The ingots after accomplishment of the zone melting were then sampled and analyzed.

Table 1: Experimental parameters for two different trails of zone melting

Parameter	**Ingot 1**			**Ingot 2**		
	Front	**Mid**	**End**	**Front**	**Mid**	**End**
Voltage [V]	232.5	240	232.5	232.5	240	232.5
Power (change with voltage, [kW]	7.7	7.9	7.7	7.7	7.9	7.7
Coil moving speed [mm/min]	0.5			0.5		
Number of passes	3			3		
Zone length [cm]	13			13		

3 Results and discussion

As characterization and analytical methodology, the X-ray fluorescence (XRF) analysis was performed to obtain the weight percentages of major elements in the cast ingots. Figure 2 shows the aluminum and copper content of the 1st ingot after 3rd pass of zone melting. It can be seen that aluminum and copper in the Al_2Cu intermetallics are stable after the process, confirming that the main components have not been affected during remelting and no compositional deviation regarding these two base elements could be detected. The result of the second ingot is consistent with the first one. It can be illustrated from Al-Cu binary phase diagram (see Figure 3) that the remelting stability of Al_2Cu is attributed to the near absence of additional phases between its solid and liquid regions. This is an important prerequisite for the subsequent analysis of the behavior of other impurity elements within the target IMC.

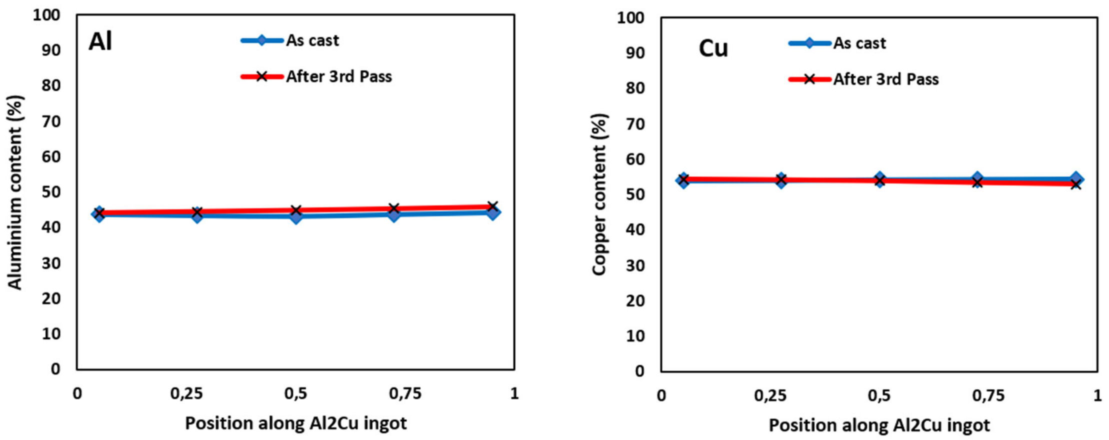


Figure 2: Al (left) and Cu (right) content of the ingot after casting and the 3rd pass of zone melting

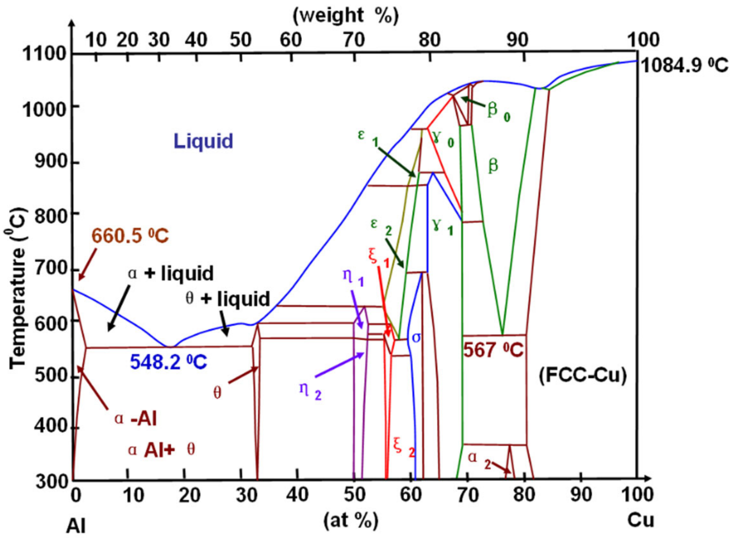
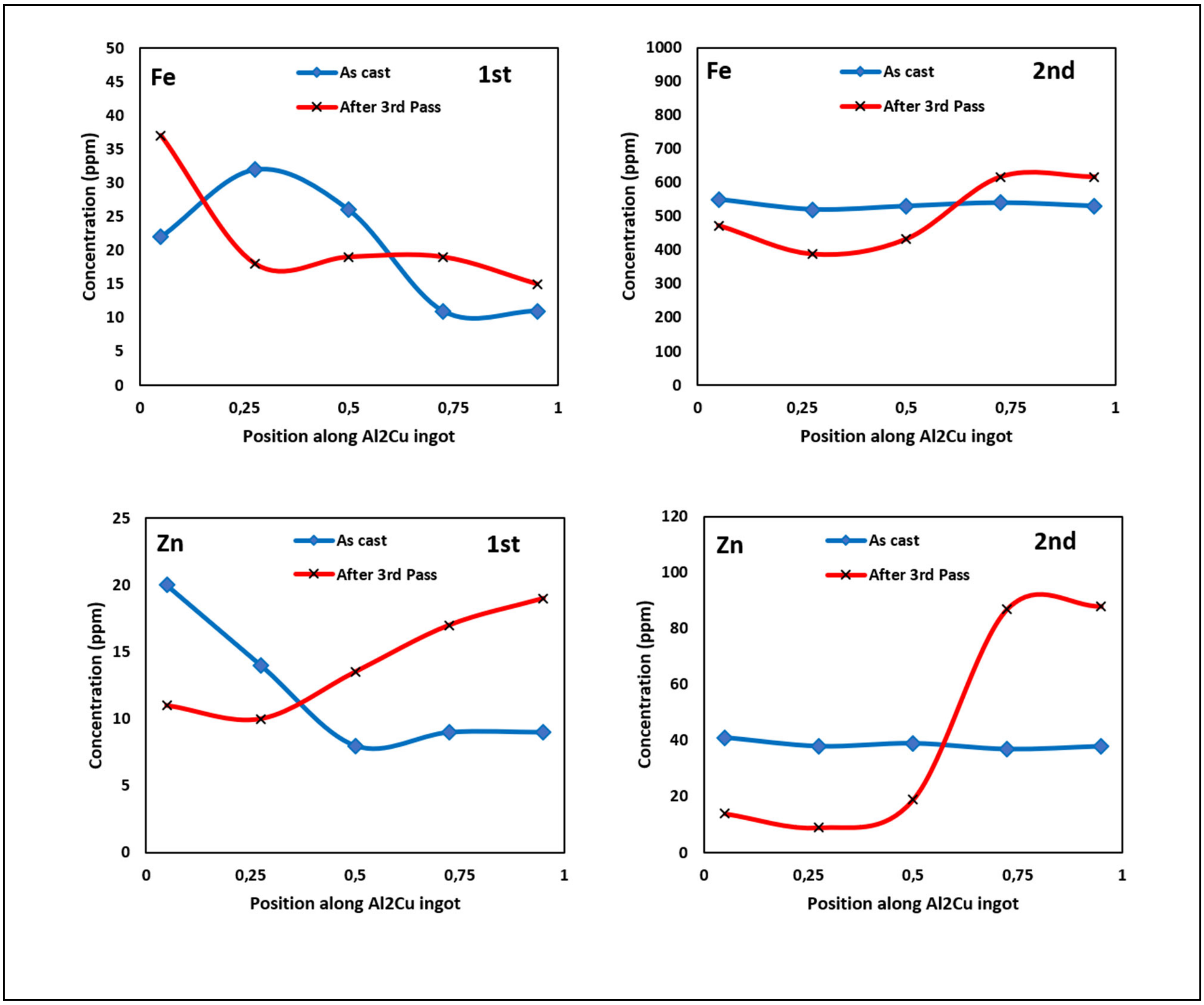


Figure 3: The Al-Cu equilibrium phase diagram [13]

Another analytical method, the inductively coupled plasma optical emission spectrometry (ICP-OES) was performed to determine the impurity distribution along the ingot. Figure 4 illustrates the ICP-OES results of different impurities before and after the zone melting process. The only difference between the first and the second ingot is on the purity of the Al in the initial raw material. It can be observed that after the 3rd pass impurities iron, zinc, magnesium, silicon and calcium tend to decrease in the front part of the ingot and accumulate at the end. This tendency can be explained by the distribution coefficient of these elements being less than unity.



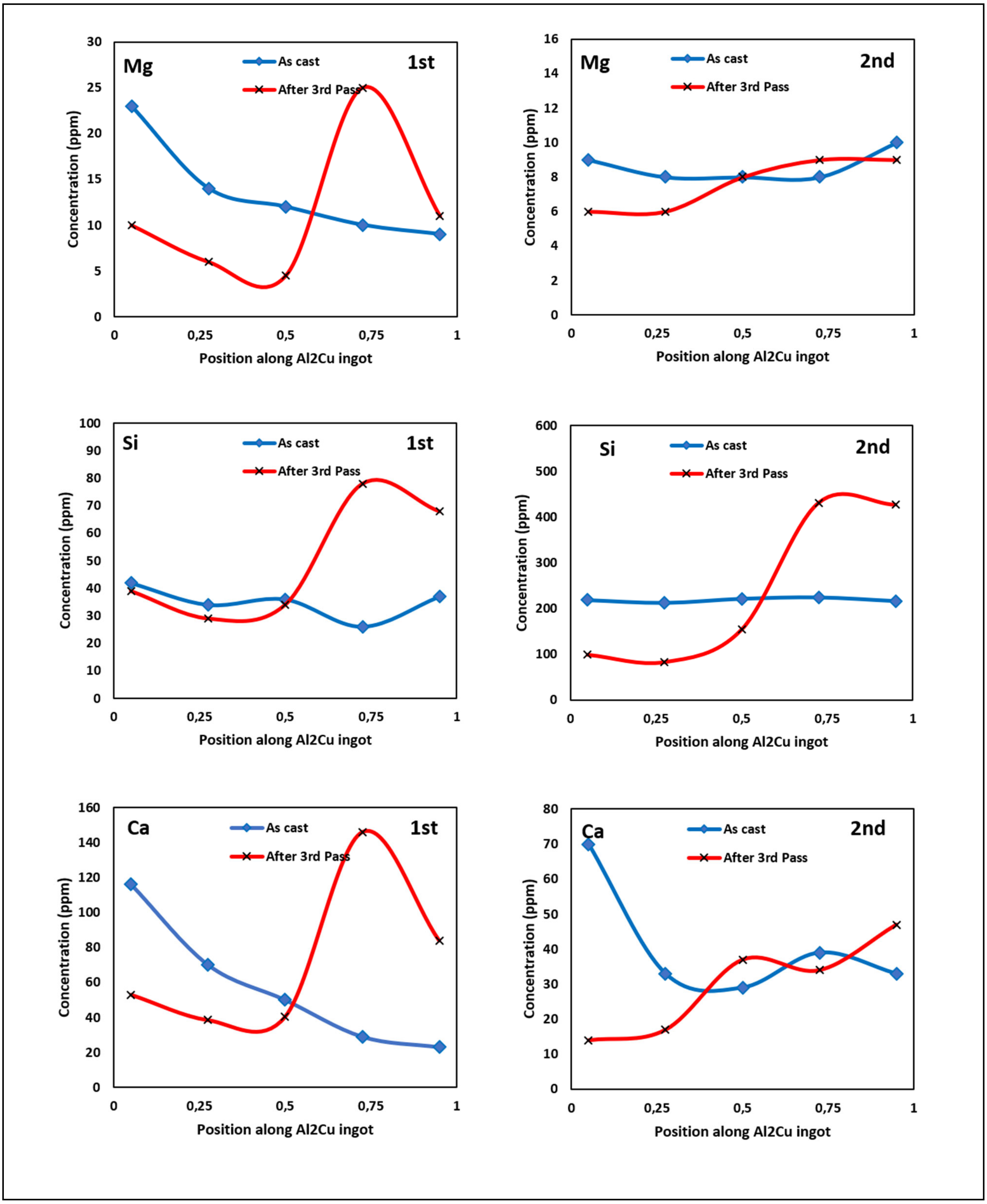


Figure 4: Concentrations of impurities Fe, Zn, Mg, Si and Ca along the 1st (left side) and 2nd (right side) ingot after casting and the 3rd pass of zone melting

It can be seen that the impurity content in the front end of the 1st ingot in the as-cast state is higher. This means that homogenization is more difficult for the synthesis of the ingot with higher purity. It is also difficult to evenly distribute very low levels of impurities to every position of the ingot during the casting process. The raw materials of the synthesis were different purity aluminum while keeping the purity of copper unchanged. The total purity of the 1st ingot is higher than the second one. However, not all individual impurities are lower in the 1st ingot as can be seen from the ICP results. The concentrations of magnesium and calcium are slightly higher. In the case of iron, the situation is a bit more complicated. The behavior of iron in the 2nd ingot is reasonable, but its increase in the front of the 1st ingot after three passes is contrary to expectations. The unexpected results could be due to analytical errors and contamination during sample preparation. In addition, from the available results, there is no noticeable difference in the purification efficiency of the target IMCs with different initial purities. This study shows the feasibility of impurities being separated. From the point of view of zone melting, the results can be further optimized by adjusting experimental parameters such as zone length, number of passes, power, coil moving speed, etc. Further studies will chemically analyze and compare the results of other passes to strengthen the credibility of the conclusions on the impurity separation behavior. More details will be shown in slides on the EMC conference and published in another journal.

4 Summary

The general feasibility of the purification of Al_2Cu via zone melting crystallization technique has been verified in this study. The main observation confirmed the stability of the main constituent elements of the target IMCs, i.e. Al and Cu. The impurities, which were focused in this work, were Fe, Zn, Mg, Si and Ca. Determination of the impurity migration after three passes of zone melting for two different bars showed that Fe, Zn, Mg, Si and Ca were clustered at the end of the ingot with a tendency to migrate that could be explained by the distribution coefficient higher than unity. Considering the possible errors in the detection and sample preparation process, further studies with higher initial impurity content, increasing the number of samples taken from the intermediate melting passes, and parameter optimization shall be conducted in IME/RWTH Aachen. This study provides the basis for the fractional crystallization principle to be used for the purification of other IMCs.

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