Grain refinement in Al-Si alloys induced by applying electric currents during solidification

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Abstract
The solidification of Al - 7 wt. % Si alloys under the influence of electric current pulses (ECP) through two parallel electrodes at the melt surface is investigated. An effective grain refinement was found if the ECP is applied during the initial solidification period (nucleation and recalescence). The grain size can be gradually reduced, which is likely due to the remelting process of high-order dendrite arms in the mushy zone driven by solute fluctuation and promoted by thermal fluctuation. This fragmentation process is mainly driven by electromagnetically forced convection. The grain refinement does not require the formation of nuclei from a solidified shell near the electrodes, which would result in a grain rain inside the sample.

Key words: Grain refinement, Al-Si alloys, electric current, electrovortex flows

Introduction
High-performance structural alloys play an important role in the daily life of human beings, particularly in the automobile and aircraft industries. The application of lightweight materials such as aluminium or magnesium alloys can reduce the weight of vehicles as well as the consumption of energy, which in turn has a positive effect on the energy efficiency and CO_2-emissions of vehicles. In addition, the safety and stability of vehicles hast to be guaranteed or even promoted. Hence, investigation and further development of high-performance structural alloys became an important research topic in academic and industry world, especially for the development of high-performance iron, aluminum, magnesium and titanium alloys [1-4].
Various techniques for enhancing the tensile strength can be applied to obtain high-performance structural alloys, such as the solid-solution strengthening, precipitation strengthening, secondary phase strengthening, strain hardening, grain refinement, etc. [5-7]. However, most of these treatments result in an undesired influence on the plasticity of alloys. This can be avoided by achieving grain refinement during the solidification process. Different approaches to achieve microstructures with finer grains have been developed, such as the technique of adding grain refiner [8], ultrasonic vibration [9], electromagnetic vibration [10], electromagnetic stirring (EMS) [11], and the so-called electric current pulse (ECP) technique [12-15].
The ECP technique considers a procedure where pulses of high electrical current are ducted through a solidifying metal alloy. However, the mechanism of grain refinement by ECP is not fully understood yet and remains a subject for discussion. Different reasons for grain refinement have been suggested, such as the fragmentation of dendrites induced due to Joule heating by the electric current, the reduction of the nucleation activity energy, or the break out and the transport of little grains from the boundary by the periodic Lorentz force. In particular, Liao et al. [12] conducted solidification experiments in liquid aluminum where current pulses were supplied through facing electrodes located at the bottom and the top of the sample. The authors assume that magnetic pressure pulses occur in the sample and separate a large number of dendrite fragments from the wall and the mushy zone, respectively, forming a so-called grain rain. From observations of significantly disturbed liquid metal surface in experiments with parallel electrodes installed through the free surface Li et al. [13] concluded that the periodic Lorentz force may create shock waves within the liquid. An essential effect of electric pulses on the nucleation rate is also suggested by Barnak et al. [14]. The authors make speculations about a reduction in the free energy difference between solid and liquid state and an increase in the liquid-solid interfacial energy, respectively. Another recent study revealed the significant role of melt convection for grain refinement [15]. It was demonstrated that the interaction of the applied current with its own induced magnetic field causes a significant electro-vortex flow in the solidifying liquid.
This paper presents an experimental study which should contribute to a better understanding of the grain refining effect by ECP. Systematic experiments are conducted to investigate the influence of several process parameters as the thermal boundary conditions and the duration of ECP application during the solidification process.

Experimental set-up and procedure

Fig. 1(a) shows the experimental setup which comprises a water cooling system, a double-walled cylindrical mould (diameter 50 mm), a temperature measurement system and an electric current power supply. The cooling system consists of a copper chill which was drained by water and kept at 20 °C.

NiCr - Ni thermocouples were located at the different heights along the vertical axis (figure 1(a)) of the samples. Each thermocouple was embedded in an INCONEL (trademark of INCO Alloys International, Huntington, WV) sheath with outer diameter of 1.5 mm and covered by Al_2O_3 tubes. Temperature data were acquired at a frequency of 5 Hz with a resolution of 0.01°C and a measuring error of ±1 °C. Two cylindrical stainless steel bars having a diameter 8 mm were used as electrodes. The parallel electrodes were dipped into the melt through the free surface with a distance of 36 mm and a penetration depth of 10 mm. The lateral surface of the electrodes is coated by an electrically insulating material (boron-nitride, BN) so that the electric current enters the liquid metal only through the front surface of the electrodes. Fig. 1(b) shows the waveform of the rectangular electric current pulses applied. In this paper we present results obtained at a current intensity of 480 A (I_0), a frequency of 200 Hz (f) and a pulse length of 0.5 ms (t_p). An Al - 7 wt. % Si hypoeutectic alloy was prepared using pure Al (99.99 wt. %) and Si (99.999 wt. %). The alloy was modified by Sr of 200 ppm in a clay-graphite crucible.

For sample preparation the molten alloy was poured into a stainless mould of 50mm diameter and solidified. Each sample was cut at the same weight of 310 g. The preparatory sample was inserted into the double walled mould and heated up to 750 °C in a resistance furnace. After a holding time of 45mins the temperature was reduced from 750 °C to 720 °C which has been kept constant for another 30mins. Then, the mould with the melt was taken from the furnace and covered with a stainless steel lid. Both thermocouples and electrodes are fixed at the lid. The electrodes were preheated to 700 °C in order to avoid an immediate solidification at the electrodes. The electric current was applied during different stages of the cooling process.

The sample was cut along the longitudinal section for metallographic analysis. One section was ground on SiC paper, etched in a solution of 60 mL HCl, 30 mL HNO_3, 5 mL HF, and 5 mL H_2O. A digital camera (Konica Minolta) was employed to obtain a photograph of the macrostructure of the longitudinal section. The other section was ground and polished from 6μm to 1μm, electro-etched of 50 mm × 30 mm in Barker etching reagent at 25 V, 15 Hz for 180 s, then examined by an optical microscope MeF4 (Leica Microsystems, Wetzlar). Polarized light was used for the quantitative analysis of the grain size. Image processing was carried out using the software Leica Application Suite V3.6 (Leica Microsystems). The grain size was measured by the linear intercept method in the software package aquinto a4i (Fa. Aquinto AG).

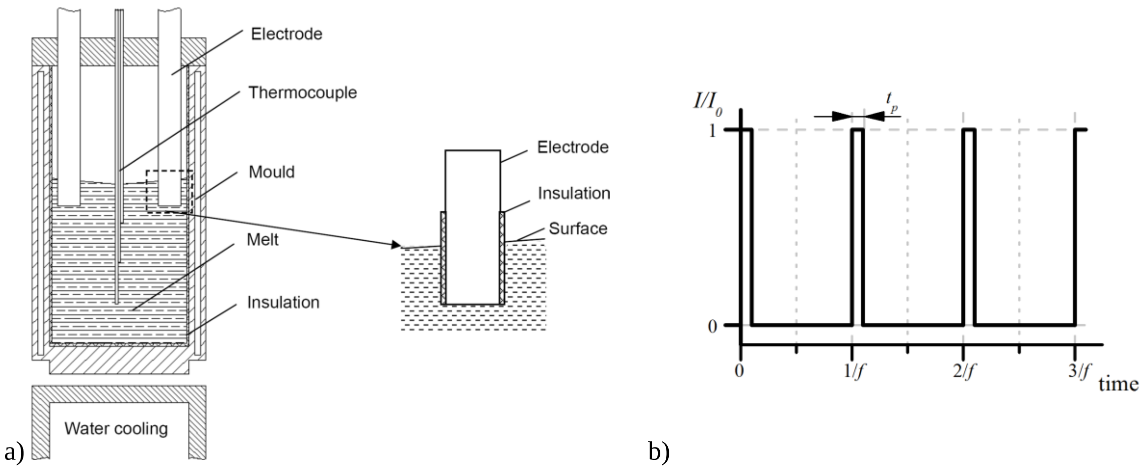

Fig.1: Schematic view of (a) the experimental setup, (b) typical waveform of an electric current pulse (ECP).

Results

The macrostructures of the solidified samples are presented in figure 2. The reference sample without applied ECP shows the vertical columnar structures aligned parallel with the temperature gradient. A columnar to equiaxed transition (CET) occurs at the middle of the sample and several coarse equiaxed grains become visible (figure 2(a)). A significant grain refinement of macrostructure is generated by the application of ECP, as shown in figure 2(b) and (c). The columnar growth is almost completely suppressed and finer equiaxed grains appear. While ECP was active during the entire solidification period in figure 2(b), the ECP treatment was switched off after the initial solidification period in figure 2(c), when a mushy zone with a significant solid fraction occurs. There are no remarkable differences between both samples with respect to the grain morphology.

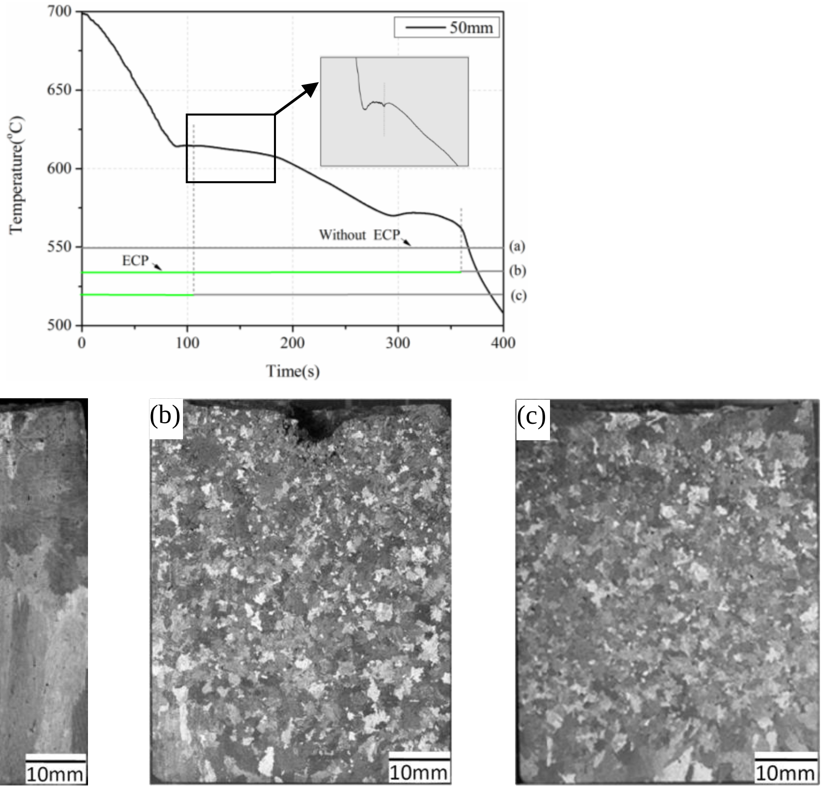

Fig. 2: Macrostructure in the longitudinal section of the solidified Al – 7 wt. % Si alloy: (a) without ECP, (b) with ECP treatment, (c) ECP treatment during the initial solidified period.

Li et. al [13] suggested a grain rain as the main mechanism for grain refinement by ECP, in particular, the electromagnetic force causes the detachment of a large number of nuclei from a thin solidified shell formed at the free surface. We conducted a special experiment to verify this assumption. The temperature of the free surface was controlled by a stainless steel block which was installed just above the melt surface at a distance of 7 mm. At first, the solidification was carried with a hot, preheated block in order to keep a high surface temperature and to avoid the formation of a solidified shell. Figure 3(a) shows cooling curves and the solidified macrostructure obtained from this experiment. The temperature of the sample with hot block measured at a height of 60mm (sample height is 65 mm) is about 618 °C at a point of time, when the greater part of the sample is already solidified. This temperature is higher as the liquidus temperature of about 615 °C. Thus, it is clear that the alloy is still liquid above 60 mm at the surface. Nevertheless, the macrostructure reveals a distinct grain refinement. The same experiment was repeated with a cold block at room temperature. The cooling curves in figure 3(b) shows a temperature of about 604 °C at a position of 60 mm. The temperature at higher positions is significantly lower which indicates the existence of a solidified layer at the surface. However, the comparison of the macrostructures dominated by equiaxed grains in both cases does not reveal a significant difference. The measurements of the grain size yield a mean value of 0.8 mm in both cases. Obviously, the nuclei for equiaxed solidification do not necessarily originate from a solidified shell at the surface.

Conclusions

This paper presents some contributions to a better understanding of the grain refinement effect in Al – 7 wt. % Si alloys with applied electric current pulse (ECP) technique.

(1) The application of ECP during the initial solidification period (nucleation and recalescence) is very efficient to achieve grain refinement in Al – 7 wt. % Si alloys.

(2) The grain refinement does not require the formation of nuclei from a solidified shell near the electrodes, which would result in a grain rain inside the sample.

Our results support the findings of Räbiger et al. [15] who suggested that the grain refinement can be ascribed to the forced melt flow driven by the Lorentz force. This convection covers the entire liquid zone and provokes increased fragmentation rates in the mushy zone growing from the bottom of the mould.

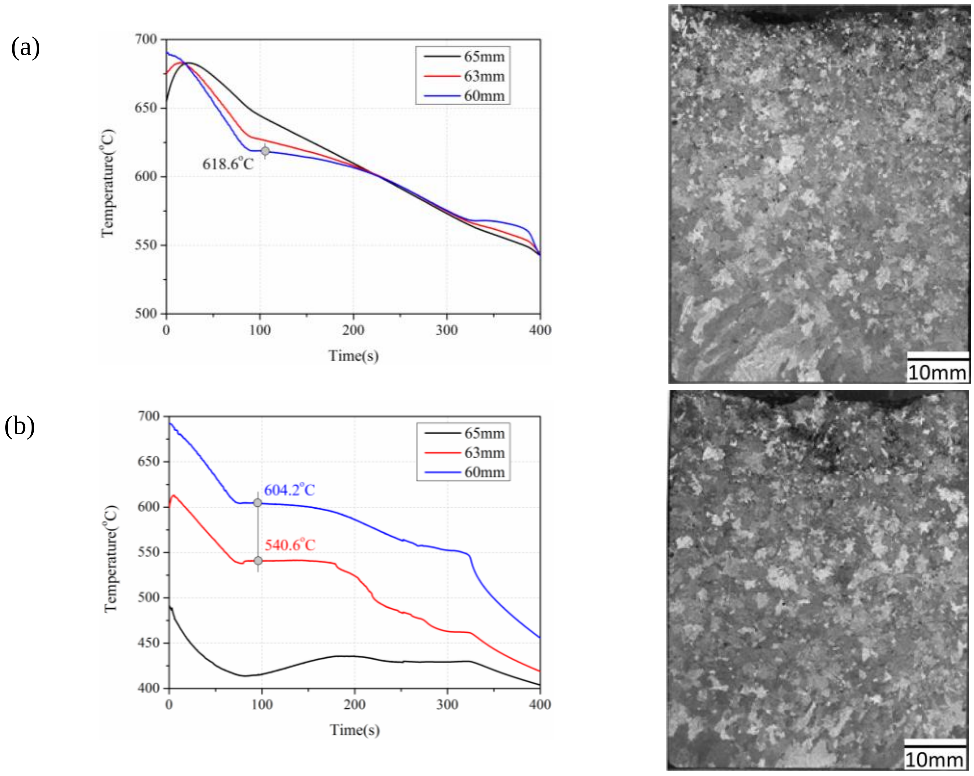

Fig. 3: Cooling curves and macrostructures of Al – 7 wt. % Si samples with ECP of I_0 = 480 A, f = 200 Hz, t_P = 0.5 ms under different conditions: (a) hot block, (b) cold block (room temperature).

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