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Influence of impurities, strontium addition and cooling rate on microstructure evolution in Al-10Si-0.3Fe casting alloys

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ABSTRACT

The effect of impurities, Sr additions and cooling rate on the microstructural evolution of high-purity Al-10Si-0.3Fe and corresponding impure commercial alloys is examined by optical microscopy and in situ by X-ray synchrotron 3D tomography. At fast cooling rate (~470 K min^{-1}) the presence of impurities decreases the growth rate of primary Al dendrites and enables formation of the β phase. Besides a modification of the eutectic Si, the addition of Sr prevents the formation of the β phase and increases the growth rate of Al dendrites. A low cooling rate (~1 K min^{-1}) leads to the formation of α, γ, δ and β intermetallic phases. In all four alloys, the dominant phase is the δ phase, regardless of commercial impurities or the Sr level. Intermetallic phases formed during slow cooling rates are much coarser than those formed during fast cooling and they have different morphologies. Our results suggest that the velocity of growth and the final morphology and size of the intermetallic phases are mainly determined by diffusional processes which in turn are controlled by the cooling rate.

1. Introduction

Al-Si alloys are widely used to manufacture automotive or aerospace components due to their low density and good mechanical and processing properties [1,2]. Iron as natural impurity is always present in commercial alloys and tends to precipitate in combination with other elements during solidification and to form various types of intermetallic phases [3–5]. These intermetallic phases can markedly degrade the mechanical properties and also give rise to casting defects [3,6]. The most common intermetallic Al-Si-Fe-phases are β, α, γ and δ, which can be distinguished by their chemical composition and morphology. Their formation path differs and depends on the amount of Fe, the cooling rate during solidification, other impurities in the alloys and additions of further transition elements. The latter are partly responsible for the observed diversity of intermetallics always present in commercial Al-Si alloys.

The first solidified Fe-rich phase (after primary Al-dendrites) is the intermetallic β-phase of composition $Al_{4.5}FeSi$ or Al_5FeSi [7,8]. Its morphology has been described as needle-like or acicular in two-dimensions, as interconnected plates or blades in three-dimensions. The intermetallic phase is considered to be the most detrimental to the mechanical properties and has therefore been widely studied [9–12]. Many attempts have been made to prevent the formation of this coarse and brittle phase or to neutralize its negative effect on castability, casting porosity and mechanical properties [13,14]. Recently, the nucleation and grow mechanisms of Fe-rich β phases have been investigated by in-situ synchrotron tomography experiments by the authors' group [15]. Contrary to previous studies [16–19] this recent research revealed that the main reason for β phase formation is Fe supersaturation in the melt near the primary Al dendrites [15].

One common intermetallic phase is α-Al_8Fe_2Si, which is characterized by the so-called “Chinese script” morphology in two-dimensions. However, the three dimensional visualization of this phase [20–22] exhibits a very complex and highly curved surface reflecting imprints of the surrounding eutectic. The formation mechanism of the α phase has been described by Timpel et al. [20]. A further phase (γ) of similar morphology but different composition and structure has been reported recently by the authors' group

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[21].

The fourth intermetallic phase is the Fe-rich δ phase. Depending on its orientation with respect to a transmission electron microscopy (TEM) foil, such phases (Al_4FeSi_2, also termed Al_3FeSi_2 [20,22]) exhibit a lath or acicular shape in two-dimensions. In three-dimensions, the phase resembles plates or blades. Because of its morphology, the δ phase is often misleadingly interpreted as the β phase. Detailed investigations of the δ phase have elucidated the formation mechanism of this phase, which differs from that of the β phase [15].

Beside the influence of impurities on the formation of the Fe-rich intermetallic phases, the cooling rate during solidification plays an important role. An influence of the Si and Fe content on the formation of the Fe-rich intermetallic phases at different cooling rates has been reported [23]. Accordingly, β phase was obtained at a cooling rate of <1 K min^{-1} and an Fe content of 0.25 wt.% independent of the Si content. Increasing the cooling rate to >6 K min^{-1} leads to additional α phase formation. Finally, at cooling rates of >9 K min^{-1}, α is the only phase that forms.

To overcome the negative effect caused by the intermetallic β phases, some researchers proposed to add traces of transition elements such as Mn, Cr, Cu or Co which can neutralize embrittlement by β and promote the formation of the morphologically more compact α phases [3,24]. Furthermore, for commercial applications, elements such as Sr or Na are usually added to Al-Si alloys prior to casting to improve their mechanical properties because they modify the eutectic Si phase. It was found that the addition of Sr to Al-Si alloys decreases both the volume fraction and the size of the Fe-rich intermetallic phases [17–19,25]. The influence of Sr on the formation of the intermetallic phases is described in Refs. [21,22].

Besides microscopic methods such as optical microscopy (OM), scanning and transmission electron microscopy (SEM, TEM), recent studies utilised tomography to investigate the microstructure of the eutectic Si as well as the Fe-rich intermetallic phases on the micrometer scale [15,20–22,26–28]. Ex-situ investigations of both the unmodified and the Sr-modified eutectic microstructures of as-cast Al-Si alloys were performed by Focused Ion Beam (FIB) tomography [20,21,26,27]. In these studies, the overall microstructure of the Al-Si alloys, the morphology of the eutectic Si phases and the true morphology of the Fe-rich intermetallic phases could be visualized. However, nucleation and growth of the Fe-rich intermetallic β phases was not revealed and could only be observed by in-situ X-ray tomography [29–32]. To clearly follow the nucleation and growth process of all phases during such an analysis, the melt has to be cooled very slowly during solidification. This results in a relatively long experiment time during which a large number of images (2D projections) have to be acquired to obtain the data for the 3D tomography reconstruction. The in-situ X-ray measurements of Refs. [15,29–32] only describe the formation of the intermetallic β phase. However, the cooling rate during solidification in in-situ experiments is not high enough to compare these microstructures with those obtained under industrial processing conditions. The cooling rate used in these in-situ experiments was either 1.4 K min^{-1} [32] or between 3 K min^{-1} and 20 K min^{-1} [30], while the cooling rate of industrial casting is usually ~470 K min^{-1}. As mentioned above the increase of the cooling rate resulted in the preferential formation of other Fe-rich intermetallic phases.

We investigated and compared the microstructural evolution during solidification at fast and slow cooling rates using OM and synchrotron X-ray 3D tomography. The influence of impurities and Sr addition was also studied. Four alloys of different composition were used, namely the well-defined high-purity alloys Al-10Si-0.3Fe and Al-10Si-0.3Fe-200 ppm Sr and the two corresponding commercial alloys containing typical impurities such as Cu, Mn, Ni, Zn, Ti, V, Ga and P.

2. Materials and methods

Al-10Si-0.3Fe (in wt.%) alloys with alloying elements of high purity (code 'P', >99.99%) and alloying elements of commercial purity (code 'C', >99.70%) were manufactured by Rheinfelden Alloys GmbH, Germany. For the modification of the eutectic Si, an Al-10Sr (wt.%) master alloy was added to the unmodified alloy in quantities leading to 200 ppm Sr content (code 'S'). All alloys were melted and cast into a cylindrical permanent mould of 30 mm in diameter and 200 mm in height leading to a measured average cooling rate of ~470 K min^{-1} within the first 40 s. The chemical compositions of all four alloys were measured by optical emission spectroscopy as listed in Table 1. The four alloy names are composed of the purity code ('P' or 'C') with an optional 'S' for strontium modification.

For optical microscopy, the cast rods were sectioned perpendicular to their axes, ground using standard metallographic procedures and finally polished with a colloidal silica suspension of 50 nm particle size (OP-U). All specimens were extracted from the centres of the castings 15 mm from the bottom of the cast rod and examined using a Zeiss Axiophot 2 with digital camera AxioCamHRc utilising differential interference contrast to visualise different phases.

For synchrotron X-ray tomography, small cylindrical samples of 3.2 mm in length and 1.5 mm in diameter were machined from the centre of a cast rod. Synchrotron X-ray tomography was carried out at the ID 19 beamline of the European Synchrotron Radiation Facility (ESRF). As-cast alloys were examined at 'room temperature' (ex-situ experiments). The temperature at which all phases in the alloys are completely melted was experimentally determined several times in the temperature range between 853 K and 939 K using synchrotron tomography. It was found that all phases are already completely dissolved at 873 K. During melting and solidification, the cylindrical samples remain close to their original shape due to the self-supporting nature of their oxide skin so that no container was necessary. For in-situ tomography, the samples were heated to a temperature of 939 K at 20 K min^{-1} and held for 5 min to ensure complete melting and homogenisation, after which they were cooled down at 1 K min^{-1} until complete solidification occurred at ~722 K. Samples were rotated over an angular range of 180° while acquiring 1000 radiographic X-ray projection images at ~18 keV photon energy. The total time for a full 180° scan is one second, while returning the rotation stage to its original position and data readout from the camera was ~60 s, resulting in a

Table 1
Chemical compositions of the four Al–10Si–0.1Fe alloys with and without Sr additions. The main elements Al, Si and Fe are given in wt. %, impurities in ppm.

alloy code	Al	Si	Fe	Cu	Mn	Mg	Ni	Zn	Ti	Cr	V	Ga	P	Sr
	wt. %			ppm										
P	89.7	10.0	0.3	7	7	4	17	33	11	4	21	80	<4	<1
PS	89.8	9.9	0.3	7	7	4	17	33	11	4	21	81	<4	197
C	89.8	9.9	0.3	16	16	4	49	96	21	9	65	92	7	<1
CS	89.6	10.1	0.3	16	15	4	47	96	20	8	63	90	8	221