Neutron and X-ray Diffraction Texture Analysis of Novel Al-Si-Mg Alloy

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Abstract. Crystallographic texture and microstructure of as-casted specimen of a novel aluminum-based alloy is analyzed. Pole figures recorded by means of neutron and X-ray diffraction methods are used as the primary data in texture analysis. The data collection is performed on the SmartLab diffractometer (Rigaku) diffractometer with Cu rotating anode X-ray tube, and the KSN-2 neutron diffractometer located at the research reactor LVR-15 in the Nuclear Research Institute, plc. Rez, Czech Republic. Software tools GSAS, X’Pert Texture, and MTEX are applied in the subsequent data processing. Additional characterization methods applied include elemental composition analysis by the instrumental neutron activation analysis, phase analysis by X-ray fluorescence and X-ray diffraction, and microstructure analysis by scanning electron microscopy and metallographic optical microscopy. Elemental composition of the alloy is dominated by Al (79.91 wt.%), Si (9.90 wt. %) and Mg (4.19 wt. %). Aluminum (cubic, Fm3m), silicon (cubic, Fd3m) and magnesium silicide (Mg2Si, cubic, Fm3m) phases are then identified as the main crystallographic phases. Crystallographic preferential orientation of the main Al phase is only analyzed and discussed; the other two phases do not provide well-resolved diffraction maxima necessary for a reliable texture analysis.

1. Introduction

Depending on the inherent single crystalline grain anisotropy, textured material as a whole can exhibit some anisotropy of mechanical, physical and even chemical properties. In principle, the preferred orientation of grains can be found even in simple powder samples. In this case, the texture is caused by an irregular habitus of the constituting grains, and the subsequent packing of these. Such shape texture can be of importance in, e.g., sintered materials or sediment rocks. However, vast majority of textured materials studied until recently has a form of compact polycrystalline aggregates, the observed crystallographic texture of which is invoked by their mechanical deformation, heat treatment, or combination of both [1].

Looking on the diffraction techniques used to characterize crystallographic textures of polycrystalline aggregates, the three radiation types are in use, including X-rays (provided by a conventional X-ray tube or a synchrotron source), electrons, and thermal neutrons (obtained from a stationary or a spallation source). Different interaction of the mentioned radiations with matter results then in different specifics of the related diffraction methods [2-4].

The Al-Mg-Si alloys show generally excellent casting properties needed for intended use of these e.g. in automotive and aerospace industries [5]. In order to fulfill the applications requirements, further research is recently in progress focused on further improvement of mechanical properties at elevated temperatures, and a higher corrosion resistance [6, 7]. There are two main ways of accomplishing the mentioned goals: modification/refinement of the alloying elements composition, and thermal treatment of the ‘as-cast’ raw alloy [7]. Composition of the alloy studied in this paper (Al80 Sι10 Mg4 Pb1 Fe0.7 Ca0.2) is selected with aim to improve the target properties. With exception of Pb, all the main constituting elements show a negative mixing enthalpy with the Al matrix [8]. Addition of a small amount of lead is intended to positively contribute to the corrosion resistance of the resulting alloy [9, 10]. The neutron (ND) and X-ray diffraction (XRD) methods are applied in order to investigate crystallographic texture of an as-casted Al-Mg-Si alloy sample; scanning electron microscopy (SEM), energy dispersive X-rays analysis (EDX), instrumental neutron activation analysis (INAA), metallographic optical microscopy and XRD methods are then used to elucidate microstructure and phase composition of the sample. Mechanical properties are tested by instrumental indentation method.
2. Experimental
Specimen of a circular cross-section 50 mm in diameter and thickness 5 mm was cut from the bottom part of the cylindrical ingot of the casted studied alloy sample and used in the ND and XRD measurements. No further processing of the as-casted specimen was performed.

Elemental composition of the sample was analyzed by INAA performed on the research reactor LVR-15 in the Nuclear Research Institute, plc. (NRI) Rez, Czech Republic. For quality control purposes, 75 – 100 mg aliquots of NIST standard reference material (SRM) 1633b Constituent Elements in Coal Fly Ash were prepared for irradiation. The amount of lead was determined by X-ray fluorescence analysis.

Microstructure of the samples was characterized by two main methods. On the larger scale, the sample structure was observed with metallographic microscope Neophot 32. The reflection micrographs were taken on the surface with the grain structure enhanced by electropolishing method. The microstructure details were then revealed by SEM (JEOL JSM840 A) providing the back-scattered electron mode and EDX analysis.

Instrumental indentation measurements were performed with NHT2 system (Anton Paar). The Vickers indenter pressed into the sample surface under the maximal load of 3 N was used. The obtained data were processed by the Oliver-Pharr method [11].

The theta/theta X’Pert PRO diffractometer with a Co X-ray tube was used for the XRD phase analysis. The X’Pert HighScore (PANalytical) software was used to process the obtained diffraction patterns.

The XRD pole figures (PFs) were collected on the SmartLab diffractometer (Rigaku) with a Cu rotating anode X-ray generator. Incomplete PFs {222}, {400} and {311} of the Al-phase were recorded with the maximum polar angle 75 degrees (measured from the specimen normal). The experimental PFs were then used to calculate the orientation distribution function (ODF) of grains which then served as a source for reconstruction of the complete PFs {200}, {220}, {111} and {311}. The calculations were performed with aid of the SW tool MTEX [12].

The ND measurements took place at KSN-2 neutron diffractometer (equipped with a Huber Eulerian Cradle 511.5 goniometer) located at the horizontal channel of the research reactor LVR-15 in NRI Rez. The KSN-2 diffraction device offers a good sample irradiation intensity with the best inter-planar distance (d) resolution value of Δd/d = 0.007 in the range of d ~ 1.0÷0.1 nm. A beam of monochromatic neutrons with the mean wavelength λ = 0.1362 nm obtained by reflection from the single-crystal Cu (200) monochromator was used to record the complete experimental PFs of the plane sets {111}, {200}, {220} and {311} of the Al-phase. The same procedure as for XRD PFs was then used to obtain the re-calculated PFs {200}, {220}, {111} and {311}.

3. Results and Discussion
3.1 Sample composition and morphology
The elemental composition obtained by INAA is in Tab. 1. The coarse-grained Al phase dominates the metallographic micrographs, accompanied by precipitates of other phases (Fig. 1). The estimated mean size of Al grains is ca 40 μm.

Table 1. Elemental composition of the studied as-casted sample. Only the main constituting elements with the concentration exceeding 500 ppm are included.

|       | Al    | Si    | Mg   | Pb    | Fe     | Ca    | Cr      | Mn     |
|-------|-------|-------|------|-------|--------|-------|---------|--------|
| [wt%] | [wt%] | [wt%] | [wt%] | [wt%] | [wt%]  | [mg/kg]| [mg/kg] |
|       | 79.91 | 9.90  | 4.19 | 1.10  | 0.69   | 0.18  | 644     | 1644   |

The Vickers hardness (HV = 95.5 ± 5.5 MPa) and Young modulus (E = 81.8 ± 2.9 GPa) values determined by instrumental indentation method exceeds in ca 20 per-cent the values reported for pure aluminum. Such enhancement is well known and occurs due to the precipitation hardening effect.
Morphology and phase composition obtained from SEM is shown in Fig. 2. In agreement with the optical micrograph (Fig. 1), the matrix consists of Al grains and contains further segregated phases, including mono-elemental particles (Cu, Pb) and grains of intermetallic phases (Mg$_2$Si, Al$_8$Si$_6$FeMg$_3$).

**Figure 1**: Typical micrograph obtained by metallographic microscopy. Electrolytic polishing is used to enhance the grain boundaries contrast. The pink background corresponds to aluminum grains; precipitates of other phases appear at the Al grains boundaries.

**Figure 2**: SEM micrograph recorded in the backscattered electrons mode. The matrix is formed by aluminum (black grains). Other phases identified by SEM/EDX are marked.

The XRD patterns used in phase composition analysis (Fig. 3) revealed presence of several main phases including aluminum (cubic, Fm$\bar{3}$m), silicon (cubic, Fd$\bar{3}$m), aluminum iron magnesium silicide Al$_8$Si$_6$FeMg$_3$ (hexagonal, P-62m), lead (cubic, Fm$\bar{3}$m) and magnesium silicide Mg$_2$Si (cubic, Fm$\bar{3}$m) (Tab. 2).

**Table 2.** Concentration of the main phases as identified by the quantitative X-ray phase analysis.

|        | Al    | Si    | Mg$_2$Si | Pb    | Al$_8$Si$_6$FeMg$_3$ |
|--------|-------|-------|----------|-------|----------------------|
| [% wt.]| 79.91 | 9.90  | 4.19     | 1.10  | 0.69                 |
3.2 Crystallographic texture

The PFs re-calculated for the Al phase from the primary XRD and ND data are given in Fig. 4 and Fig. 5, respectively. The directions marked “X” and “Y” are arbitrarily selected and “physically indistinguishable” from the point of the cylindrical symmetry of the casting cuvette. The recalculated PFs agree well with the primary experimental PFs (not shown here). The observed isolated maxima reflect the coarse-grained microstructure of the Al phase. Taking into account the large size of grains, it is worth to note that the PFs obtained by ND should be considered statistically more significant than the data obtained by XRD.

The PF {200} contains, for both applied methods (although more distinctly for XRD), some extremes located in the central part of the projection, surrounding the normal direction identical with the axis of the casting cuvette. In an ideal and sharp form, such alignment of the {100} poles parallel to the normal direction is well known to appear in cast aluminum alloys [1]. However, contrary to the XRD PFs, the ND PFs show maxima near the outer rim of the PFs, indicating some preference in the radial direction. The apparent difference of the XRD PFs from the ND results is likely due to additional surface deformation introduced during the specimen cutting and thus, the ND results are more reflective of the texture in the bulk. The XRDs PFs have maxima just below 5 m. r. d (mean random distribution); whereas, the maxima observed in the ND PFs are below 3 m. r. d. reflecting a relatively random texture in the cast material.

**Figure 3:** XRD patterns used in phase analysis. The phases corresponding to the main diffraction maxima are indicated.

**Figure 4.** PFs {111}, {200}, {220} and {311} of the Al phase recalculated from the XRD data.

The scale given in m. r. d. units.
4. Conclusions
The performed research showed that the microstructure of the investigated as-cast sample of Al-Si-Mg alloy is composed of a coarse-grained aluminum matrix filled with precipitates of other phases localized at the Al grain boundaries. The main minor phases include silicon, aluminum iron magnesium silicide, lead, and magnesium silicide. Presence of the phases was confirmed by EDX and quantitative XRD phase analysis. The Vickers hardness and Young modulus show enhancement following from the precipitation hardening process amounting to 20 percent increase of the values observed for pure aluminum.

The aluminum matrix shows crystallographic texture of low-to-medium sharpness, with isolated maxima that are in line with the coarse-grain character of the aluminum phase. The pole orientations are declined from the positions known to appear in casted Al and Al-Si alloys characterized by the texture component with prevailing orientation of {100} poles along the casting cuvette axis. Randomization of the aluminum matrix texture and reduction of its sharpness can be likely attributed to the coarse-grained microstructure of the Al matrix interpenetrated by other segregated phases, and influence of the alloying elements.

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