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MEASUREMENT OF OXIDE-LAYER THICKNESS OF INTERNAL GRANULES IN HIGH-PURITY ALUMINIUM*

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High-purity aluminium samples and mixtures containing different amounts of alumina were irradiated by 13 MeV ^{3}He particles. The aim of the investigation was to determine the oxide-layer thickness on the surface of internal aluminium granules. Measurement was carried out by determining the bulk oxygen concentration in the samples, and calculating the oxide-layer thickness, by using model conditions about the microstructure of the aluminium samples.

Introduction

The extended use of aluminium in its high-purity form and in different types of alloys made in the conventional way and/or by powder metallurgy technology makes it necessary to investigate the purity and the microstructure of these materials. Oxygen is a very important element in connection with metals both on the surface and in the volume, too. A limited number of methods is known for the determination of bulk oxygen concentration in aluminium. Now we only deal with nuclear methods, especially Charged Particle Activation Analysis (CPAA).

This method is very useful when the half-life of the radio isotope produced from the element in question is different from the half-lives of the other confusing isotopes. The detection limit and accuracy of CPAA is quite good, compared with other methods [1]. The ^{3}He and triton particles are the best choices for bombardment [2, 3] because of the good irradiation and detection parameters. Our cyclotron can accelerate protons, deuterons, ^{3}He and ^{4}He particles, so we applied ^{3}He for irradiation.

For oxygen determination the $^{16}O(^3He,p)^{18}F$ and $^{16}O(^3He,n)^{18}Ne \rightarrow ^{18}F$ reactions were used [4]. Their cross-sections and yields data are published in [5]. The half-life of the produced ^{18}F isotope (110 min) is long enough for comfortable measurement.

The purpose of our recent investigation was to determine the surface layer thickness of an aluminium-oxide layer on the surface of internal granules of an aluminium sample pressed from high-purity (4N) aluminium powder. The main oxygen content in this type of aluminium metal is supposed to be on the surface of the internal granules.

*Dedicated to Academician D. Berényi on his 60th birthday

Experimental

The aluminium samples were positioned in an evacuated analytical chamber and were irradiated by ^{3}He particles. The beam energy was chosen to be 13 MeV and the current was 1–4 microampers. The irradiation time was 5–20 min depending on the sample type. The gamma-spectra of the irradiated targets were measured by Ge(Li) detector and the data were collected by a Canberra multichannel analyser type S35-plus in order to investigate the gamma-lines of all activated elements. For the determination of the oxygen concentration the $E_\gamma = 511$ keV the annihilation peak of ^{18}F was used after separating it from other radiation by half-life measurement. The time between the end of irradiation and the beginning of the measurement (cooling time) was approximately an hour.

Two types of samples were irradiated, both of them were disk-shaped and pressed from powder. The first type of samples was pressed from high-purity (4N) aluminium powder, the other was made from a mixture of high-purity aluminium and aluminium-oxide powder about 50 μm in particle diameter.

Determination of the oxide-layer thickness on the surface of granules

To avoid the inaccuracy in beam-current determination we used standard samples with known oxygen content. These samples were made from a homogeneous mixture of aluminium and alumina powder. By means of the standard samples we could use the relative method for the determination of oxygen concentration.
Model conditions and the initial data and formulae were:

r: radius of the Al granules supposed to be spheres

d: thickness of oxide layer on the surface of the granule

$V_1 = (4/3)\pi r^3$: aluminium volume in a single granule measured in cm^3

$V_2 = 4\pi d r^2$: oxide volume in a single granule measured in cm^3

$m = m_1 + m_2 = V_1\varrho_1 + V_2\varrho_2$: mass of a granule measured in g

n=1/m : number of granules in 1 g material

O=m$_2$N : number of oxygen atoms in a granule disregarding the bulk oxygen content in the granule

C=On : number of oxygen atoms in 1 g aluminium powder

N=1.7705 10^{22} : number of oxygen atoms in 1 g of alumina

$M = M_1 + M_2$: mass of the mixture of M_1 g aluminium powder and M_2 g alumina powder

N(O) : number of oxygen atoms in 1 g mixture

ϱ_1 ϱ_2 : density of aluminium and alumina respectively

$a = \varrho_2/\varrho_1$: ratio of densities

Using the above definitions we get for N(O)

$$N(O) = \frac{M_1 C + M_2 N}{M} = \frac{N}{M}\left[\frac{M_1 da}{da + r/3} + M_2\right] . \qquad (1)$$

After substituting of $M_2 = 0$ in (1) the number of oxygen atoms in 1 g aluminium powder is given as

$$N'(O) = N\frac{da}{da + r/3} . \tag{2}$$

The ratio of (1) and (2) gives a simple formula which contains among other parameters the thickness of alumina on the surface of a single granule.

$$R = \frac{N(O)}{N'(O)} = 1 + \frac{M_2 r}{3Mda} . \tag{3}$$

From (3) we get

$$d = fr , \tag{4}$$

where

$$f = \frac{1}{3a(R-1)\left[\frac{M_1}{M_2} + 1\right]} . \tag{5}$$

The f involves only such parameters which can be measured easily by using standards and the relative method.

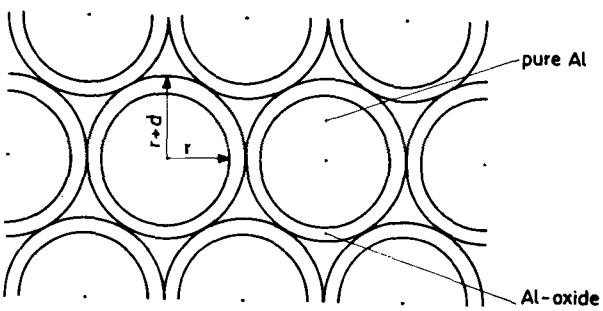

Fig. 1. Model conditions and parameters for the microstructure of aluminium samples pressed from powder

Results and discussion

The average granule size of the investigated aluminium powder was about 50 μ in particle diameter. Knowing the average granule diameter we could determine the thickness of the alumina layer on the surface of a single granule, which in this case was: $d = (8.5 + 3)$ nm.

The inaccuracy in thickness determination is due to the approximated value of granule diameter, and to the interfering radiation of other isotopes. To avoid these errors longer irradiation time, and chemical separation of the ^{18}F isotope should be used.

Applying the above described method, the quality of the products of powder metallurgy can be improved. It is also possible to determine the surface-oxide layer thickness not only in the case of aluminium but of other metals, too.

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