TECHNOLOGY OF ALUMINUM DEEP PURIFICATION BY ZONE MELTING METHOD USING ULTRASONIC WAVES

A.I. NADJAFOV, R.R. BABAYEVA
Institute of Physics of NAS of Azerbaijan
AZ 1143, H.Javid ave.,131, Baku
a.najafov@mail.ru

The method of three-stage zone melting for deep purification of A85 (99,85%Al) aluminum mark is used. The ultrasonic waves for intensification of liquid mixing and diffusion processes during zone melting are used. The optimal parameters of ultrasonic waves are defined by experimental methods. It is shown that the use of ultrasonic waves with frequency 20 kHz and intensity 0,4Vt/cm^2 allows us to increase the aluminum purity level up to 99,999% in comparison with usual process of zone melting.

Keywords: purification, aluminum, zone melting
PACS: 81.05 Rm

INTRODUCTION

The modern industry can't be imagined without aluminum and its melts. The aluminum and its melts are used in many industry fields: aircraft building, shipbuilding, transport, technique, building, cable production, oil and chemical industries and etc.

The purer aluminum is mainly widely applied in electronics from electrolytic capacitors up to microprocessors, in cryoelectronics, cryomagnetics, in critical parts of chemical equipment working in corrosion-active mediums or at extremely low temperatures, modern radar engineering and especially critical products of atomic power engineering.

The purity of industrial aluminum (99,5 – 99,8%) in series of cases isn't enough, as the higher degree of industrial aluminum purification obtained by electrolysis for these goals, is required. Note that last time many investigations concerning to different methods of aluminum purification (electrolytic, crystallized, distillation) are carried out [1-3]. The zone purification method of pure aluminum obtaining is the more practical, industrial and effective one [2-7].

The zone melting principle is in multiple passing of molten zone along aluminum ingot. These impurities can be divided into three groups by values of distribution coefficients $K=C\text{sol}/C\text{liq}$ (where Csol is impurity concentration in solid phase and Cliq is impurity concentration in liquid one) which essentially define the purification efficiency from impurities. The impurities decreasing the aluminum melting point are to first group. They have $K<1$, locate in molten zone and are transferred by it to ingot end part at zone melting. Ga, Sn, Be, Sb, Ca, Th, Fe, Co, Ni, Ce, Te, Ba, Pt, Au, Bi, Pb, Cd, In, Na, Mg, Cu, Si, Ge, Zn belong to these impurities.

The impurities increasing the aluminum melting point are to second group. They have $K>1$ and locate in solid part (heading) of ingot. Nb, Ta, Cr, Ti, Mo, V are to these impurities.

The impurities with distribution coefficient which is very close to 1 (Mn, Sc) belong to the third group. These impurities aren't practically scavenged at aluminum zone melting.

According to [8-9] the purification efficiency from impurities at zone melting depends on redistribution coefficient $K=C_{sol}/C_{liq}$. However, the crystallization front travel rate and liquid stirring rate before crystallization front essentially influence on impurity distribution between liquid and sold phases at crystallization. It may be shown that for impurity having K<1 the moving crystallization front displaces the impurities with bigger velocity than their diffusions from liquid in growing crystal.

By this reason the region with increased impurity concentration forms before the crystallization front. The thickness of this layer, composition and impurity concentration determine the parameters of diffusion impurity from liquid in growing crystal. The thickness of this layer also depends on hydrodynamics of liquid metal and usually has the size about 10^{-3}-10^{-5} m. The diffusion coefficient has the value in limits of 10^{-9} - 10^{-8} m^2/sec, although their values aren't determined for all impurity types. The component excess (K<1) will increase in case of diffusion difficulty in liquid phase on crystallization front during zone travel. The forming solid phase is deplated by this component. This leads to solid phase crystallization not from the main phase but from the layer neighbouring to crystallization front in which impurity concentration increases that leads to decrease of impurity separation effect. Thus, the difficulty of the component diffusion in liquid state prevents to the separation or impurity purification at zone melting. The ultrasound is applied in works [5, 8-10] with the aim of impurity concentration decrease on crystal-melt boundary and intensity increase of impurity diffusion into melt depth. It is established that influence of ultrasonic waves of determined frequency increases the intensity of impurity diffusion into liquid depth and increases the purification efficiency [8-11].The ultrasonic waves are used by us for the intensification of mixing of liquid and diffusion processes during aluminum zone melting. We give below the results of carried out experiments.

EXPERIMENTAL PART

Taking into consideration the aluminum chemical activity, the aluminum zone melting is carried out in

131 H.Javid ave, AZ-1143, Baku
ANAS, G.M.Abdullayev Institute of Physics
E-mail: jophphysics@gmail.com

crucibles, which are inertial ones for the process. The high melting point and inertia to cleanable material are the main requirements to the crucible material. The carbon-base glass is the more stable material to aluminum melt. The carbon-base glass has the high thermostability in inertial gas and vacuum up to 3000° C. The high-quality polished surface, maximal chemical stability and low acceptance by metal melts and other substances are the main advantages of carbon-base glass crucibles and boats. The boats from carbon-base glass of SIGRADUR mark (Germany) are used in our experiments.

The aluminum purification process is begun from metal surface purification on oxide film. Note that aluminum oxide forms the additional crystallization centers at solidification of a metal that leads to disturbance of impurity redistribution effect between ingot solid part and its molten zone. Besides, the aluminum oxide is the impurity active adsorbent that also badly influences on metal purification.

The metal ingots are cut by crucible sizes and the surface is etched by 10% hydrochloric acid at 60°C. Further, the ingots are washed in distillated water, swilled out by deionized water and dried in vacuum. Later, the metal degassing is carried out. The metal degassing before its zone melting is carried out by the way of melting and its endurance in vacuum 10^{-3} mm of mercury.

Further, the preliminary aluminum vacuum remelting is carried out. The melting and endurance of aluminum in vacuum (~10^{-3} mm of mercury) in melted state at 720-740°C essentially decrease (3-5times) the content of hydrogen and magnesium and zinc impurities [12-13]. The quantity of zinc impurity after the endurance during 2 hours is decreased in 4 times. Besides, the average gas content of initial aluminum 0,21cm3/100gr after endurance during one hour is decreased in more than 2 times and after 8 hours the gas content in metal isn't practically revealed. However, as analyses show the quantity of impurities of cuprum, iron and silicon aren't practically changed after melting and long endurance of aluminum.

The scheme of installation used for aluminum purification is shown in fig.1. It is carried out in horizontal variant. The device consists of two independent purification reactors. Each channel has three successive cascades of heaters. The two independent material purification processes in quantity 50-250gr in temperature interval 50-1050ºC are carried out on the device.

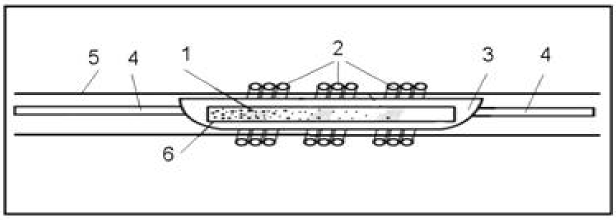

Fig.1. The scheme of installation of cascade zone melting. 1 is crucible with substance; 2 are haters; 3 is vacuum-degassed ampoule; 4 are holders; 5 is guide tube.

The two similarly prepared and evacuated quartz ampoules with aluminum put into carbon-base glass crucibles are loaded on the device. It is shown in fig. 2.

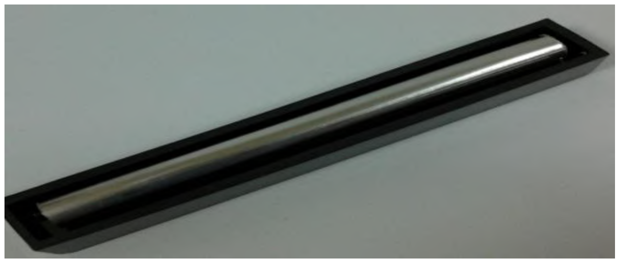

Fig.2. The boat from carbon-base glass for aluminum zone melting.

The prepared ampoules with substances are put into guide quartz tubes of the device. The heating resistors forming the narrow ingot molten zone, moving towards with heater, shift outside along ampoule with aluminum ingot. The zone temperature is 750° C. The device heaters are defended with oval screen allowing concentrating the thermal radiation inside the heating ring. Note that furnace electric resistances are comfortable ones in work and don't require the complex equipment for electric power supply and process regulation. The aluminum ingot length is near 250mm, the molten zone width is 30-35mm, the velocity of zone displacement is chosen on the base of experiment results and it is 20mm/h. The cascade passing number along ingot is 10.

The aluminum zone purification without external influences is carried out in first reactor. The same process of zone recrystallization is carried in second channel by ultrasonic oscillation influence. For this two soldered adaptations parallel each other are in ampoule ends and the ultrasonic vibrator sensor is attached to one of these ampoule ends. The ultrasonic generator of G3-109series with piezoelectric transformer is used in our case. The other end of this ampoule is put into device holder clutch filled by smooth silicon material of «Elstolux-M» mark for damping of ultrasonic oscillations.

The ultrasonic oscillation frequency is chosen in interval 15-20kHz with the fact that the cavitation threshold increase in melt takes place at frequency increasing more than 20kHz, i.e. minimal value of sound pressure enough for cavitation appearance at given conditions increases. The intensive mixing of melt components doesn't take place at frequency less 15kHz. The intensity of radiation is chosen in interval 0,3 - 0,5 Vt/cm^2. Our results allow us to recommend the frequency given intervals and ultrasonic oscillation intensities.

The analyses of obtained ingot purity are carried out after 10 cycles of zone melting. The initial and end parts (approximately 20% of ingot length) are cut off the each ingot. The samples which are pressed up to necessary dimensions are cut off midsection of ingots (60% of length) and analyses on roentgen-fluorescent spectrometer "S8 TIGER" are carried out on them. The analysis results of purified aluminum ingots are given in Table1.

According to results given in table 1 the main substance purity increases almost on one order after 10 passes of aluminum purification of mark A85 on installation of cascade zone melting. Moreover, the content of Fe and Si decreases almost in 30 times, Zn decreases almost on one order and cuprum quantity decreases in three times. The titanium impurity quantity becomes less roentgen-fluorescent sensitivity value (10^{-4}%).

The change of impurity content in aluminum after zone melting and after zone melting with application of ultrasonic waves is compared in table 1. These data show the significant decrease of impurity content in aluminum after application of ultrasonic waves during zone melting. According to carried out analyses the quantity of content of Fe and Si impurities in aluminum ingot decreases up to $3\cdot10^{-4}$% level and content of Cu and Zn impurities decreases up to $2\cdot10^{-4}$% that exceeds the aluminum purity level obtained using the zone melting after 10 cycles of three-cascade zone purification and application of ultrasonic waves with frequency 20kHz and intensity 0,4Vt/cm^2.

The obtained positive result of aluminum purification level by zone melting method with application of ultrasonic waves is the result of intensification of liquid mixing and diffusion processes during zone melting processes.

Table 1.

The analysis results of aluminum ingots purified by zone melting method.

Purification method	Chemical composition and impurity quantity in %					Main substance in %
	Fe	Si	Cu	Zn	Ti	
Initial aluminum of mark A85*	0,08	0,06	0,01	0,02	0,008	99,83%
Three-cascade zone melting (10 passes)	0,003	0,002	0,003	0,002	-	99,99%
Three-cascade zone melting (10 passes) with ultrasonic wave application	0,0003	0,0003	0,0002	0,0002	-	99,999%

* Chemical composition of initial aluminum is given according to data State Standard RF 1069-2001.

CONCLUSION

1. The deep purification of aluminum of A85 (99,85%Al) mark is carried out by three-cascade zone melting method. It is shown that the application of ultrasonic waves with frequency 20kHz and intensity 0,4 Vt/cm^2 allows us to increase on one order the purity level of purified aluminum up to level 99,999% of main substance.

2. The method can be used in industrial production for obtaining of essentially pure aluminum.

ACKNOWLEDGEMENTS

The work is supported by fund of ”Scientific-investigation programs” of Azerbaijan National Academy of Sciences.

[1] *Khimicheskaya ektsiklopediya*. Gl. red. I.L. Knunyants. 1988, Izd. Sov. Entsikl. Moskva, (in Russian)

[2] *A.I. Belyayev, G.E. Volfson, G.I. Lazarev*. Poluchenie chistogo alyumunuya. M., Metallurgiya, 1967, 268s. (in Russian)

[3] *A.I. Belyayev.* Metallurgiya lyogkikh metallov. 1970, M, Metallurgiya, 368 s. (in Russian)

[4] Allyuminievie splavi. Metallovedenie allyuminiya i ego splavov. Алюминиевые сплавы. Spravochnik. Otvet. red. *I.N. Fridlyander.* M, Metallurgiya, 1983, 277 s. (in Russian)

[5] *V.N. Vigdorovich*. Ochistka metallov i poluprovodnikov kristallizatsiey. M, 1969. 296 s. (in Russian)

[6] *S.G. Marayev i dr*. Trudi VAMI, 1970, vip. 680, s. 110. (in Russian)

[7] *N. Parr*. Zonnaya ochistka i yeyo tekhnika. M., Metallurgizdat, 1963, 211 s. (in Russian)

[8] Povishenie effektivnosti tekhnologichweskikh protsesov v pole akusticheskikh kolebaniy. Sb. Pod redaktsiyey *N.N. Khavskogo*. M, 1981. (in Russian)

[9] *O.V. Abramov.* Vliyanie ultrazvuka na protses zonnoy plavki naftalina. Akusticheskiy jurnal, 1967, t.13, v.2, s.161-168. (in Russian)

[10] *B.A. Agranat, V.I. Bashkirov, Yu.I. Kitaygorodskiy*. Kavitatsionnoe razrushenie poverkhnostnikh plyonok v akusticheskom pole pri povishennom staticheskom davlenii. Akusticheskiy jurnal, 1967, t. 13, s. 283-285. (in Russian)

[11] *B.A. Agranat, M.N. Dubrovkin i dr.* Osnovi fiziki i tekhniki ultrazvuka. M., Visshaya shkola, 1987, 352 s. (in Russian)

[12] Allyuminievie splavi. Primenenie allyuminievikh splavov. Spravochnoe rukovodstvo. Redaktsionnaya kollegiya *I.V. Gorinin i dr.*, Moskva, "Metallurgiya", 1978. (in Russian)

[13] Allyuminiy. Svoystva i fizicheskoe metallovedenie. Spravochnik. *J.E. Khetch*. "Metallurgiya", 1989. (in Russian)

Received: 20.11.2015