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### **Development Of Aluminum Liquefaction Technology**

#### Turakhodjaev Nodir

Professor, Head Of Foundry Technologies Department, Tashkent State Technical University, Uzbekistan

#### Tashkhodjaeva Kamola

PhD Student, Department Of Foundry Technologies, Tashkent State Technical University, Uzbekistan

#### Abdullaev Kamol

Associate Professor, Department Of Machinery Technology Namangan Engineering Construction Institute, Uzbekistan

#### **Begmatov Dilmurod**

Senior Teacher, Department Of Machinery Technology Namangan Engineering Construction Institute, Uzbekistan

#### ABSTRACT

This paper examines the effect of hydrogen gas on the quality of the casting during the liquefaction of aluminum alloy. In addition, the technology for the separation of aluminum from  $Al_2O_3$  oxide, depending on the liquefaction temperature during the liquefaction of aluminum alloy.

#### **KEYWORDS**

Aluminum, chemical element, constitute, dissolved, hydrogen, molten, atmosphere.

#### **INTRODUCTION**

Aluminum (Al) chemical element, a lightweight silvery white metal of main Group 13 (III a, or boron group) of the periodic table. Aluminum is the most abundant metallic element in Earth's crust and the most widely used nonferrous metal. Because of its chemical activity, aluminum never occurs in the metallic form in nature, but its compounds are present to a greater or lesser extent in almost all rocks, vegetation, and animals. Aluminum is concentrated in the outer 16 km (10 miles) of Earth's crust, of which it constitutes about 8 percent by weight; it is exceeded in amount only by oxygen and silicon. The name aluminum is derived from the Latin word alumen, used to describe potash alum, or aluminum potassium sulfate,

K Al (SO<sub>4</sub>)<sub>2</sub>·12H<sub>2</sub>O.



A unique combination of properties puts aluminium and its alloys among our most versatile engineering and construction materials. All alloys are light in weight, yet some have strengths greater than that of structural steel. The majority of alloys are highly durable under the majority of service conditions and no coloured salts are formed to stain adjacent surfaces or discolour products with which they come in contact, such as fabrics in the textile industry and solutions in chemical equipment. They have no toxic reaction. Aluminium and most of its alloys have good electrical and thermal conductivities and high reflectivity to both heat and light. Aluminium and most of its

alloys can easily be worked into any form and readily accept a wide variety of surface finishes. Light weight is perhaps the best known characteristic of aluminium, with density of approximately 2.73 x 10<sup>3</sup> kilograms per cubic meter at 20° C as compared with 8.89 x 10<sup>3</sup> for copper and 7.86 x 10<sup>3</sup> for carbon steel.

# The source of dissolved hydrogen in molten aluminum

Only one element of dissolved gas component in aluminum is hydrogen. Hydrogen in molten aluminum (H) has an equilibrium relationship with hydrogen gas in ambient atmosphere. The American Journal of Engineering and Technology (ISSN – 2689-0984 Published: April 22, 2021 | Pages: 17-24 Doi: https://doi.org/10.37547/tajet/Volume03Issue04-03

(2)

$$H = \frac{1}{2} H_2(gas) \tag{1}$$

Equilibrium constant (KH) of eqn (1) is

$$K_{\rm H} = \frac{p_{\rm H^{\frac{1}{2}}}}{f_{\rm H}[\% \ H]}$$



Figure 1 Solubility of hydrogen at 1 atm. in 99.9985% pure aluminum.

where *f*H is the activity coefficient of hydrogen in aluminum, [%H] is the hydrogen concentration in aluminum and  $P_{H_2}$  is the partial pressure of hydrogen gas in the atmosphere. Figure 1 [1] shows the equilibrium hydrogen concentration in pure aluminum

with hydrogen gas of 1 bar. (It means the solubility of hydrogen into aluminum under the atmosphere of hydrogen gas partial pressure of 1 atm.) Aluminum reacts with water vapor at high temperature and generates hydrogen gas.

$$3H_2O(g) + 2Al = Al_2O_3 + 3H_2(g)$$
 (3)

This hydrogen gas is the source of hydrogen in aluminum. In the cast house of aluminum industry they often in humid hot season experience more troubles on cast quality for the dissolved hydrogen in the melt. This is due to the chemical reaction between water vapor of higher partial pressure in ambient atmosphere of humid hot season and molten aluminum. Researchers proposed the numerical model of hydrogen pick-up from water vapor and he suggests the hydrogen concentration in molten aluminum which is kept for long time under the atmosphere of a constant water vapor pressure  $(p_{H_{2O}})$  should attain to the calculated value.

In this model (Fig. 2), at the interface between molten aluminum and atmosphere, researcher looks at various steps involved as follows. Water vapor diffuses through the boundary layer to be adsorbed at the metal surface, the adsorbed molecules reacts with aluminum, hydrogen molecules are desorbed from the surface,



Figure 2. The mechanisms for hydrogen dissolution into the molten aluminum from moisture in the atmosphere.

hydrogen molecules diffuse back out of the boundary layer, hydrogen molecules dissociate and form atomic hydrogen on the surface, hydrogen atoms diffuse through the metal boundary layer. By mathematical analysis of each steps using mass transfer coefficients k at gas-melt interface and the equilibrium constant for in the surface layer, the partial pressure of hydrogen gas ( $p_{H_2}$ ) at the interface is given by

$$p_{\rm H_2} = \frac{k_{\rm H_2O}}{k_{\rm H_2}} p_{\rm H_2O} \tag{4}$$

where  $k_{H_2O}$ ,  $k_{H_2}$  are mass transfer coefficients for  $H_2O$ ,  $H_2$  in gas and  $p_{H_2O}$  is the partial pressure of  $H_2O$  in the atmosphere. Therefore, the hydrogen concentration in molten aluminum ([%H] *I*) which is kept under the atmosphere of  $P_{H_2O}$  for long time is calculated from researchers.

$$[\% H]_l = \frac{1}{f_{\rm H} K_{\rm H}} \sqrt{\frac{k_{\rm H_2O} p_{\rm H_2O}}{k_{\rm H_2}}}$$
(5)

#### **Molten Metal Processing**

The mass transfer coefficients  $k_{H_{2O}}$  and  $k_{H_2}$  can be shown to be proportional to the square root of the diffusion coefficients in air,  $D_{H_{2O}}$  and  $D_{H_2}$ . Therefore,

$$[\%H]_{I} \approx \frac{1}{f_{\rm H}K_{\rm H}} \sqrt{p_{\rm H_2O} \sqrt{\frac{D_{\rm H_2O}}{D_{\rm H_2}}}} \tag{6}$$

The diffusion coefficients in air are  $D_{H_{2O}}=0.239$  cm<sup>2</sup>/sec at 8°C and  $D_{H_2}=0.634$  cm<sub>2</sub>/sec at 0°C [2]. As a first approximation it is assumed that their ratio do not change significantly with temperature. So, we can obtain researchers.

$$[\%H]_l \approx \frac{0.783}{K_{\rm H}} \sqrt{p_{\rm H_2O}}$$
(7)

There are few papers which deal with the experimental result of the hydrogen concentration dependence on P<sub>H2O</sub>. Fig.2. made an experiment to determine the hydrogen pick up of the molten pure aluminum from the water vapor of  $P_{H_{2O}}$  in the ambient atmosphere. Figure 3 shows the experimental apparatus which can keep the molten metal in the atmosphere of a constant partial pressure of the water vapor. The water vapor partial pressure  $P_{H_{2O}}$  was controlled by blowing the dry gas (air or inert gas) of which the dew point is below - 60°C through the molecular sieves (in the case of dry air) or the humidified gas through the pure water into the stainless steel box, and the value of  $P_{\text{H}_{2}\text{O}}$  above the melt surface was determined by the measurement of the dew point of the gas blew out of near the melt surface in the box. The temperature of the melts were controlled to 675±5°C, 700±5°C or 750±5°C. The molten aluminum of 99.99% pure in the high purity alumina crucible (inner diameter 80 mm, height 170 mm) was held in stationary state or stirred state by the rotating graphite impeller (dia. 45 mm, height 30 mm) at 530 rpm. The hydrogen concentrations of the molten aluminum before

and after the treatments were measured by the nitrogen fusion method (I THAC-2002 manufactured by ADAMEL LHOMARGY was used.) for the carefully machined cylindrical samples from the ingots solidified into the Ransley's mould. Figure 4 shows the experimental result of the hydrogen concentration change in the molten pure aluminum which was held at 700°C in the dry air atmosphere with  $1.7 \times 10^{-4}$  atm. of  $p_{H_{20}}$  ( $p_{H_{20}}$  in the usual air in Japan is about 1.5x10<sup>-3</sup>-3.0x10<sup>-2</sup> atm.). The hydrogen concentration of the stationary melt slowly decreases, and after long time holding more than 300 min it looks like to attain a constant value which may be same as the equilibrium value of hydrogen concentration (0.07ml=100g =0.07p:p:m:) which had been attained after about 50 min holding while stirring the melt by rotating impeller. Figure 5 shows the time dependence of the hydrogen concentration in the stirred molten aluminum under the air atmospheres containing various amounts of water vapor. The hydrogen concentration of molten aluminum attains to the equilibrium value depending on  $P_{H_{2O}}$ irrespective of whether the initial hydrogen concentration is lower or higher than the equilibrium one.



Figure 3. The experimental apparatus for the investigation of hydrogen dissolution into the molten aluminum in the high purity alumina crucible (inner dia. 80 mm, inner height 170 mm) from moisture in the atmosphere.

At 675°C

$$[H_e] = 3.17 p_{\rm H_2O}^{0.470} \tag{8}$$

At 700°C

$$[H_e] = 3.49 p_{\rm H_2O}^{0.453} \tag{9}$$

At750°C

$$[H_e] = 3.16 p_{\rm H_2O} \,^{0.387} \tag{10}$$

The activation energy of hydrogen solution from water vapor was calculated to be 38806 cal/mol, 30977 cal/mol and 19467 cal/mol on each case of 1.0 x  $10^{-2}$ ,  $10^{-3}$  and  $10^{-4}$  atm. of  $P_{H_{2}O}$ .



## Figure 4. Hydrogen concentration change in molten 99.99% Al at 700°C under the dry air atmosphere of $P_{H_{2}O}$ =1.7 x 10<sup>-4</sup> atm.

They are in rough agreement with the activation energy of hydrogen solution from hydrogen gas, 28258 cal/mol [3]. These experimental result suggests the pick up of hydrogen from water vapor may occur by the model proposed by researcher and it is limited by slow mass transfer of hydrogen in molten aluminum to the surface, although these experimental values of the equilibrium hydrogen concentration is lager than the calculated value. It is supposed the oxide film of molten aluminum surface may affect the hydrogen pick up of molten aluminum, because the equilibrium hydrogen concentration with  $P_{H_{2}O}$ in inert gas atmosphere of  $N_2$  or Ar is lower than in air atmosphere and it is attained earlier than in air atmosphere.

#### CONCLUSION

In conclusion, the technology of production of quality castings by reducing hydrogen gas in the process of liquefaction of aluminum alloys has been developed. Hydrogen concentration change in molten 99.99% Al at 700°C under the dry air atmosphere of  $P_{H_{2O}} = 1.7 \times 10^{-4}$  atm. These experimental result suggests the pick up of hydrogen from water vapor may occur by the model proposed by researcher and it is limited by slow mass transfer of hydrogen in molten aluminum to the surface, although these experimental values of the equilibrium hydrogen concentration is lager than the calculated value.

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