RADIOISOTOPIC METHOD OF STUDYING THE EFFECT OF ELECTROMAGNETIC STIRRING ON THE DISSOLUTION OF SOLID METALS IN LIQUID METALS

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A study was made of the effect which the flow of liquid metal has on the diffusive dissolution in it of solid metals. The dissolution of alloying elements was examined radioisotopically, along with the kinetics of mass transfer under conditions of lead alloy and aluminum alloy formation.

Considerable interest has developed recently in the study of processes involved with the dissolution of solid metals in liquid metals. This growing interest is explained by the wide use of such processes in various branches of the domestic industry: in atomic magnetohydrodynamic power generation, in liquidmetal thermal power systems, in general metallurgy, and in foundries where diverse alloys are cast. The study made here was concerned with the effect of stirring the liquid metal with an electromagnetic pump on the diffusive dissolution of solid metals in a liquid one. The processes were studied with the aid of a special-purpose radioisotopic tracking system.

The purpose of the study was to establish the effects of various technological factors in the dissolution of alloying elements and on the kinetics of mass transfer in MHD type apparatus.

The MHD apparatus for this study (Fig. 1) operated on induction heating and melting of the metal, with controlled stirring by electromagnetic forces [1].

The metal in vat 1 and in channels 2, 3, 4 formed short-circuit loops comprising the secondary winding of the transformer 5. Upon application of a voltage to the primary transformer winding 6, therefore, a current was induced in the metal which heated the latter and, at the same time, generated one component of the electromagnetic force.

While the windings 8 of electromagnet 7 were additionally switched in, a magnetic flux was produced across the airgap of this electromagnet in the direction normal to the flow of current in the metal. Interaction between this magnetic flux and that current produced an electromagnetic force driving the metal through channel 3. The magnitude of this force could be regulated over a wide range, by adjustment of the voltage applied to the primary transformer winding, or to the electromagnet winding.

Thus, a special feature of this apparatus was that the metal flowing through the channels was subjected to a compound effect of several factors: temperature rise and perturbations due to scattered electromagnetic fields and alternating pulsations, both resulting from a variation in the magnitude and the direction of electromagnetic forces.

The first stage of this study was concerned with the dependence of the dissolution process on the temperature, the contact surface of injected alloying mass, and the discharge velocity of the metal jet from the channel.

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Fig. 1. Magnetohydrodynamic apparatus with a radioisotopic tracking system: 1) pumping vat; 2, 3, 4) pumping channels; 5) magnetic structure of the transformer; 6) primary winding of the transformer; 7) electromagnet; 8) electromagnet winding; 9) basket with antimony; 10, 11) source of γ radiation; 12) lead plate of the collimator; 13) detector of γ radiation.

TABLE	1.	Dissolutio	on Rate	of Antimony in Pb–Sb
Alloy, as	s a	Function	of the	Melt Temperature

	Dissolution rate of antimony, kg/sec				
ture, °C	antimony added without stirring	antimony added with stirring			
350	1,1×10-3	1,6×10-3			
400	1,5×10-3	2,1×10 ⁻³			
450	3,1×10 ⁻³	5,0×10-3			
500	4,1×10 ⁻³	6,4×10 ⁻³			
550	-8,3×10-3	8,3×10 ⁻³			

Into the vat with liquid metal was dropped a basket with constant-weight doses of an alloying element. Liquid lead was used as the solvent for the first test series. Antimony was used here as the alloying ingredient. The initial antimony content in the alloy was maintained at $6 \pm 0.1\%$ per weight of lead, and the transformer power was held constant. Prior to the addition of a specific dose of alloying mass into the liquid metal, a small batch of lead was also added so as to maintain the 6% concentration level of antimony. The temperature of the metal bath was measured with Chromel-Alumel thermocouples and a

model ÉPP-09 potentiometer, accurately within $\pm 3\%$. The velocity of the metal jet discharging into the vat during stirring was measured with Pitot tubes. The error of this method in measuring the velocity of liquid metal was $\pm 15\%$. The solvent for the second test series was aluminum, with manganese or chromium as the alloying element.

In the study of the effect of each said factor, the dissolution time was measured by the radioisotopic method with a specially installed device consisting of an γ -radiation source and detector [2]. As the source was used the radioactive Cs¹³⁷ isotope 50 mg equiv. of radium strong for the analysis of Pb-Sb alloys and 10 mg equiv. of radium strong for the analysis of aluminum alloys. A scintillation counter served as the detector, consisting of a NaI(Tl) single crystal and a model FÉU-13 photoelectron multiplier. The information coming from this photoelectron multiplier was read into a model PP-15 converter and then printed by a model ÉUM-23 digital readout device. The radiation source was placed in the top part of the basket for the analysis of lead alloys and outside the metal bath for the analysis of aluminum alloys.

The γ -radiation flux, attenuated during passage through the antimony basket and the molten metal, was recorded by the γ -radiation probe.

The recorded γ -radiation flux varied according to the following relation:

$$J = kJ_0 \exp\left(-\mu_1 \rho_1 l_1 - \mu_2 \rho_2 l_2\right). \tag{1}$$

In the course of dissolution, antimony was being replaced by a Pb-Sb alloy. The recorded γ -radiation flux was weakened in the process. As soon as the entire antimony in the basket had been dissolved, the γ -radiation flux reached a constant level at which it then remained. This instant of time was regarded as indicating the end of the dissolution process. From the size of the antimony doses and from the length TABLE 2. Dissolution Rate of Antimony in Pb-Sb Alloy, as a Function of the Contact Surface Area of Antimony Grains

	Dissolution rate of antimony, kg/sec				
face area of alloying	above th	alongside the jet			
mass, m ²	jet discharge velocity				
	0 m/sec	0.25 m/sec] 0.25 m/sec		
78.10-4	5,1.10-4	7,15.10-4	6,4.10-4		
140.10-4	10,4.10-4	11,9.10-4	11,4.10-4		
165-10-4	12,5-10-4	14,7.10-4	13,9.10-4		
2800-10-4	19,2.10-4	41,5.10-4	31.10-4		
•			1		

of the dissolution time, we also calculated the process rate. The error in measuring the process rate did not exceed 5%.

The effect of the temperature of the original metal on the length of the dissolution time of the alloying mass was determined with the metal temperature variation controlled within the 350-520°C range. The results of these tests are given in Table 1. An analysis of these data indicates that in each case the solubility curves are exponential. In the lower temperature range the dissolution rate can be raised appreciably by circulating the liquid metal and by placing the basket with the alloying mass under the jet. As the temperature rises to some definite level,

critical for the given alloy, the circulation of the liquid metal ceases to affect the dissolution rate of the alloying mass.

This can be explained by the fact that, at lower temperatures of the solvent metal, the dissolution rate is determined by both the entry of solute atoms into the boundary layer and the diffusion process occurring at the time. Causing the liquid metal to move relative to the solid alloying mass results in an appreciable acceleration of the diffusion process and, consequently, a shortening of the dissolution process. As the temperature of the solvent metal rises, the entry of solute atoms into the boundary layer becomes the predominant factor. At some temperature the diffusion rate reaches its maximum, even during only slight stirring of the metal as a result of circulation by electrodynamic forces which appear in the channels even when the electromagnet is turned off [3]. In this case, the flow of the liquid metal has a negligible effect on the dissolution rate of the solid alloying mass but rather tends, essentially, to equalize its concentration.

For studying the effect of the contact surface area of the alloying mass on the dissolution rate, we added doses of equal weights but different surface areas in contact with the liquid metal, i.e., with different degrees of comminution. These doses were placed in various zones of the bath, either above or along-side the jet discharging from the center channel, under static conditions or during stirring action by that jet discharging at a velocity of 0.25 m/sec. The results of this experiment are shown in Table 2. An analysis of these results indicates that the dissolution rate of the alloying mass increases with increasing contact surface and that it is highest with the basket above the stirring jet but lowest with the liquid metal unstirred. The dissolution rate does not increase proportionally to the area of contact, however. This can be explained by a deterioration of the wetting conditions in the melt when the contact surface becomes larger, especially with the solvent metal at standstill.

In Fig. 2 are shown test results pertaining to the effect of the stirring rate on the dissolution rate of the alloying mass, the stirring caused here by a jet of metal discharging from the channel into the vat. In this experiment the temperature of the metal was maintained at 360°C. The discharge velocity of the metal jet for stirring the bath was varied from 0 to 0.7 m/sec.

An analysis of these test data (Fig. 2) indicates that, with this method of setting the liquid metal into motion relative to the solute mass, the dissolution rate of the latter varies with the circulation velocity of the former according to the $|1-\exp(-\alpha v)|$ relation. The results indicate that, under optimized circulation modes, it is possible to attain dissolution rates attainable without stirring but with the metal temperature raised by 100-200°C.

Analogous experiments were performed with aluminum alloys containing 7.5% manganese, 20% manganese, or 10% chromium. The test conditions were identical. The trend of the dissolution rate in the metal bath was found to be the same here as in the case of Pb-Sb alloys (Fig. 2). However, the optimum circulation rate of the melt in the case of aluminum alloys was three times higher than in the case of Pb-Sb alloys. This could be explained by the higher kinematic viscosity of the Al-Mn and the Al-Cr systems: the kinematic viscosity of the Pb-Sb system is $\nu \approx 2 \cdot 10^{-7}$ m/sec², of both the Al-Mn and the Al-Cr systems: $\nu \approx 5.5 \cdot 10^{-7}$ m/sec². Furthermore, the diffusion rate is higher in the Pb-Sb system than in the Al-Mn and the Al-Cr systems. Consequently, in order to realize the most efficient flow pattern in a bath, simulated earlier in [4] and then confirmed in the Pb-Sb experiment, it appears necessary to ensure the same ratio of parameters defining the Reynolds number.



Fig. 2. Dissolution rate of antimony (v_d , kg/sec) as a function of the stirring rate in the metal bath (v, m/sec), with a metal volume: 1) 3.2 dm³; 2) 4.8 dm³; 3) 6.4 dm³; 4) 8.0 dm³.

The second part of the study was concerned with the kinetics of mass transfer during the dissolution of alloying antimony in lead melt.

The processes occurring in a liquid metal bath in our apparatus were simulated with translucent liquids at values of the Reynolds number corresponding to a velocity of metal circulation from 0 to 1 m/sec and, apparently, under conditions of developed turbulence. In this case, in addition to the metal flow through the channel-vat-channel system, there occurred also large transverse oscillations contributing to the mass transfer between adjacent layers. The analysis of mass transfer kinetics was also based on radioisotopic measurements. Here the melt was located between the source and the detector of γ radiation.

In order to single out the test zone in the liquid metal, a leaden collimator with nine collimating ducts 50 mm long and 30 mm in diam-

eter was provided here. The dissolution process in the melt was tracked by changes in the alloy concentration within these nine typical bath zones, and this concentration was determined as a linear function of the melt density. The measure of that density was the recorded γ -radiation flux. The latter varied exponentially with the increase of alloying content [2]. Eventually, the flux J reached a constant level. This served as an indication of a constant alloy concentration within the test zone. The measurements were made at velocities of the metal jet which produced stirring action in the bath and which were varied from 0 to 0.65 m/sec. According to the test data, conditions more favorable to equalizing the alloy distribution prevailed either in the region adjoining the jet or near the lateral channels. Velocities within the 0.4-0.5 m/sec range appeared most effective in terms of equalizing the chemical composition of the entire alloy mass.

The optimum stirring mode, which had been established earlier to ensure both an accelerated dissolution of alloying elements and a homogeneous alloy composition, was also duplicated under industrial conditions for casting binary alloys of lead with 6.5% Sb and ternary alloys of lead with 5% Sb + 0.2% As to be used in storage batteries. These alloys were produced at a temperature of 400°C, to ensure a stable composition throughout the ingot, while a high dissolution rate was attained by means of electromagnetic stirring.

An examination of specimens revealed that, in terms of their mechanical properties, they were not inferior to the better batch of production samples. Their microstructure was characterized by a more uniform distribution of the eutectic phase along grain boundaries than in conventionally produced alloys (Fig. 3). A batch of storage-battery grids cast from our alloy was successfully tested for strength, without exhibiting a tendency of the material to crack formation.

Analogous experiments performed with aluminum alloys (Al-Mn and Al-Cr) have demonstrated the feasibility of successfully duplicating these alloying processes with the original metal (aluminum) at temperatures of 840 and 900°C, respectively, rather than at the recommended 1100-1200°C temperatures, which makes it possible to reduce the gas inclusions in the alloys and to extend the usability of mold coatings.



Fig. 3. Microstructure of lead-antimony alloy (6% Sb): a) produced by the conventional process; b) produced in the magnetohydrodynamic apparatus.

A chemical analysis of the solid product, sampled from various zones (top, middle, and bottom) in a standard mold containing an Al-Mn alloy processed by the method shown here, indicated a variation in the manganese content within not more than a few hundredths of a percent, namely 7.46, 7.45, and 7.51% respectively.

Samples of the same alloy from the bottom and the middle zone were also examined by x radiography on a model URS-55 instrument (with copper as the source of radiation), and the lines on the principal interference pattern were those of aluminum. In addition, there appeared also strong lines of the Al_6Mn phase.

Another x radiogram obtained on a model URS-504 instrument (with iron as the source of radiation) established the existence of the Al_6Mn phase in both the middle and the bottom zone of an ingot, while the amplitudes corresponding to the intensity lines of this phase in each zone did not differ much from one another, indicating a uniform distribution of this phase over the ingot volume.

The described apparatus, together with the radioactive system for specimen analysis, has thus been useful for studying both the dissolution of solid metals in liquid metals and the attendant processes of mass transfer during combined thermal and hydrodynamic conditioning.

Such a type of apparatus is very suitable for producing alloys of hard-to-dissolve and easy-to-melt elements. Specifically, the optimum stirring mode would be a jet discharge from the center channel into the vat at velocities within 0.3-0.5 m/sec for producing lead alloys and within 0.8-1.0 m/sec for producing aluminum alloys.

NOTATION

J is the recorded γ -radiation flux;

- J_0 is the primary flux of γ radiation;
- k is a constant coefficient determined by the apparatus design;
- μ_1 is the mass coefficient of attenuation in the Pb–Sb alloy;
- μ_2 is the mass coefficient of attenuation in antimony;
- ρ_1 is the alloy density;
- ρ_2 is the density of antimony;
- l_1 is the thickness of the liquid Pb-Sb layer between radiation source and radiation detector;
- l_2 is the total thickness of antimony grains in the path of γ rays;
- α is the solubility of a solid metal in a liquid one;
- v is the velocity of the liquid metal relative to the solid alloying mass;
- ν is the kinematic viscosity.

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