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EVAPORATION OF ZINC IN AN ARGON – HYDROGEN MIXTURE

Many modern processes for the production and casting of metals and their alloys are carried out in protective gas atmospheres, which protect them, for example, from oxygen pollution. This applies, for example, to titanium, magnesium or aluminum alloys. Most liquid alloys are comprised of constituents that differ in vapor pressures, resulting in harmful phenomenon during melting due to evaporation of some of its components. This harmful process may be limited by the selection of a suitable gas atmosphere in which the liquid metal treatment process is carried out. In the paper, results of study on the impact of the type of gas atmosphere on the rate of evaporation of zinc in argon – hydrogen mixtures are presented. It should be noted that such mixtures are used, for example, in metal welding processes, in which it is also possible to evaporate a component of the so-called liquid metal pool. The research results showed that the rate of zinc evaporation increases with the increase of hydrogen content in the gas atmosphere.

Keywords: Evaporation; Gas atmosphere; Thermogravimetry; Argon-hydrogen mixture

1. Introduction

The process of metal evaporation accompanies virtually every pyrometallurgical operation related to the production of metals and their alloys as well as their refining. For example, during the process of smelting steel or processing it in various types of metallurgical aggregates it can occur the evaporation of its metallic admixtures, characterized by high values of equilibrium vapor pressure. The available literature data indicate that during these processing operations of the liquid steel, we can remove by evaporation such elements as chromium, manganese, copper, zinc or lead [1-4]. Some of them contaminate steel, while others are admixtures improving its properties. An unfavorable phenomenon of evaporation also occurs during the melting of titanium alloys, when the content of such alloying elements as aluminum and manganese in metal bath is decreasing [5-10]. Intensive evaporation also accompanies arc welding operations, where the presence of metal vapors have a significant impact on the arc properties as well as the size and shape of the weld pool being created. This mainly applies to welding operations on galvanized steel and brass [11-24]. The paper presents the results of tests on the evaporation rate of zinc in argon-hydrogen mixtures.

2. Research methodology

All experiments were carried out by thermogravimetry using the NETZSCH STA 449 F3 Jupiter analyzer. The method of measurement was identical for all experiments. The zinc sample, located in the alumina crucible, was putted on the TG thermoanalyzer carrier. Then, after the analyzer was calibrated, the appropriate gas was introduced into the analyzer chamber and its flow was determined at a given level. The sample was heated at a rate of 20 K/min. The sample was kept in isothermal conditions for 30 minutes. After this time, the heating system was turned off and the sample was cooled to ambient temperature. During the whole experiment, the weight loss of the sample was registered. Crucibles with an internal diameter of 6 mm were used in the tests. The experiments were carried out in argon-hydrogen mixtures containing from 5 to 35 % vol. hydrogen at temperature of 948 K. In addition, tests were carried out for zinc samples of varying mass. They were carried out at the same temperature in an Ar-15 % hydrogen mixture.

3. Results

Fig. 1 presents collectively examples of TG curves obtained for zinc evaporation processes carried out at 948 K in various

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argon-hydrogen mixtures. Fig. 2 presents TG curves obtained for zinc evaporation experiments in the Ar-15% H₂ mixture, carried out for samples of different mass. The determined average weight loss of zinc during measurements carried out for various gas mixtures is shown at Fig. 3.

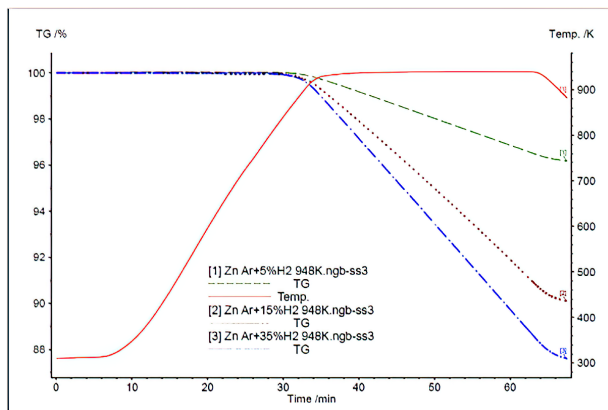


Fig. 1. TG curves obtained for zinc evaporation processes in various mixtures of Ar-H₂ (948 K)

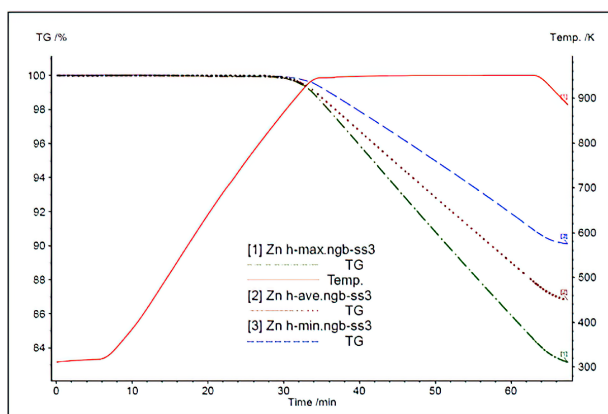


Fig. 2. TG curves obtained for zinc evaporation processes in the Ar-15% H₂ mixture (948 K – different sample heights)

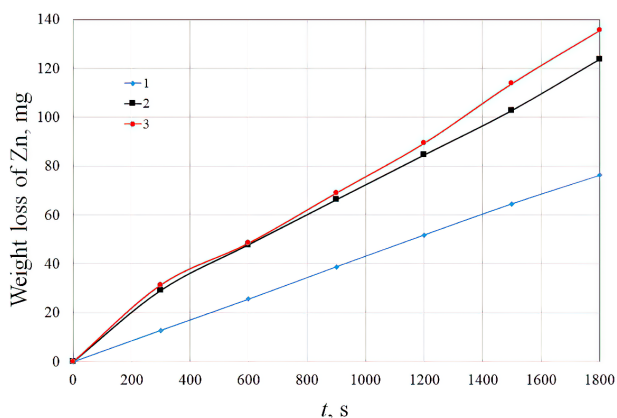


Fig. 3. Zinc weight loss for various gas mixtures (1-5% H₂; 2-15% H₂; 3-35% H₂)

TABLE 1 shows the average values of the zinc evaporation flux determined from experimental data.

TABLE 1

The average flux of zinc vaporization

Process temp., K	Applied gas atmosphere	Average sample weight, mg	Flux Zn, mg s ⁻¹
948	Ar + 5% vol. H ₂	1000	0.030
948	Ar + 15% vol. H ₂	1000	0.068
948	Ar + 35% vol. H ₂	1000	0.101
948	Ar + 15% vol. H ₂	500	0.042
948	Ar + 15% vol. H ₂	1500	0.075

4. Discussion

The analyzed process of evaporation of pure zinc is an example of a heterogeneous process taking place in a metallic liquid – gas phase system. From a kinetic point of view, it can divide it into two main stages, i.e.

- evaporation of zinc from the interface between liquid metal and gas phase,
- transport of zinc vapors from the interface between both phases into the gas phase.

If we assume that the analyzed evaporation process takes place by diffusion of its vapors through a stationary layer of inert gas, in this case argon-hydrogen mixture, then the distribution of zinc concentration in the gas phase is presented as in Fig. 4.

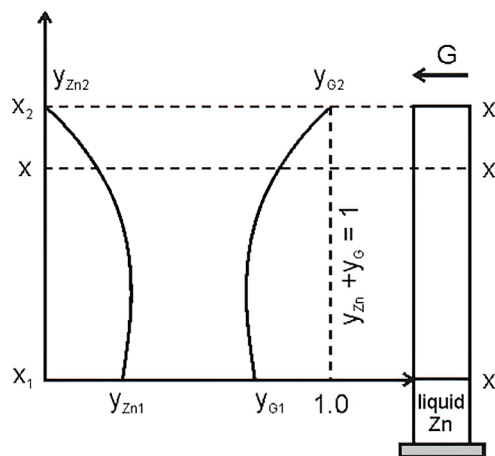


Fig. 4. Distribution of zinc concentration in a stationary gas phase over a liquid zinc bath

Under this assumption, the flux density zinc vapor may be determined from the equation:

$$N_{Zn} = \frac{PD_{Zn-G}}{RT(x_2 - x_1)} \ln(p - p_{Zn2}) / (p - p_{Zn1}) \quad (1)$$

where:

- D_{Zn-G} – diffusivity of zinc vapors in inert gas,
- p_{Zn1} and p_{Zn2} – respectively zinc vapor pressure at the surface of the liquid metal and at a height of x_2 above the liquid,
- $(x_2 - x_1)$ – height of the crucible free space above the liquid metal,

R – gas constant,
 T – temperature.

If we assume that:

$$p_{Zn2} = 0 \quad (2)$$

and also that the p_{Zn1} pressure is equal to the equilibrium pressure of zinc, then the dependence on the density of the zinc flux takes the form:

$$N_{Zn} = \frac{PD_{Zn-G}}{RT(x_2 - x_1)} \ln(p/p - p_{Zn1}) \quad (3)$$

To determine the value of zinc vapor density from Eq. (3), it is necessary to know the zinc equilibrium pressure over the bath (p_{Zn1}) and the zinc vapor diffusivity in the argon – hydrogen mixture. To estimate the equilibrium pressure of zinc vapors, the thermodynamic data from the HSC Chemistry ver. 6.1 program were used [25]. The change in this pressure in the temperature range 948-1048 K is shown in Fig. 5. On the basis of the determined curve, one can notice a dynamic increase in the zinc vapor pressure over the metal bath with increasing temperature.

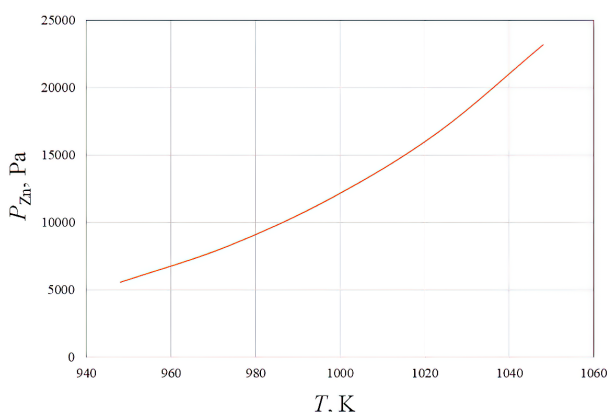


Fig. 5. Zinc equilibrium pressure in the temperature range of 949-1048 K

The Hirschfelder equation based on the kinetic theory of gases was used to determine zinc diffusivity:

$$D_{AB} = 5,954 \cdot 10^{-24} \cdot \frac{T^{\frac{3}{2}} \left(\frac{1}{M_A} + \frac{1}{M_B} \right)^{\frac{1}{2}}}{p \cdot \sigma_{AB}^2 \cdot \Omega_{D_{AB}}} \quad (4)$$

where:

D_{AB} – mutual diffusion coefficient, m^2s^{-1} ,
 p – general pressure, Pa,
 M_A, M_B – molar masses of A and B gases, $kg\ mol^{-1}$,
 σ_{AB} – constant force of gas particles interaction, m,
 $\Omega_{D_{AB}}$ – function describing the properties of A and B gases.
 The force constant σ_{AB} was determined from the equation:

$$\sigma_{AB} = \frac{\sigma_A + \sigma_B}{2} \quad (5)$$

where: σ_A, σ_B – constants force of components A and B .

The value of the function $\Omega_{D_{AB}}$ was estimated from the equation:

$$\Omega_{D_{AB}} = 1,075 \left(\frac{k \cdot T}{\varepsilon_{AB}} \right)^{-0,1615} + 2 \left(\frac{10k \cdot T}{\varepsilon_{AB}} \right)^{-0,75 \log(k \cdot T \cdot \varepsilon_{AB}^{-1})} \quad (6)$$

where: ε_{AB} – constant force of interaction of gas components A and B .

This constant was determined from the equation:

$$\varepsilon_{AB} = \sqrt{\varepsilon_A \cdot \varepsilon_B} \quad (7)$$

where: $\varepsilon_A, \varepsilon_B$ – constants force of components A and B .

TABLE 2 summarizes the characteristic parameters of gases used for calculating the vapor diffusivity values of D_{Zn-G} zinc. TABLE 3 sets out the determined D_{Zn-G} values in the gas mixtures in which the experiments were conducted. A graphic interpretation of the effect of temperature on the analyzed zinc diffusivity is shown in Fig. 6.

TABLE 2

Selected characteristic parameters of gases and metal vapors

Gas	Characteristic parameters of gases		
	$\sigma \cdot 10^{10}$	$\varepsilon/k, K$	ε_{AB}
H ₂	2.96E-10	33.3	4.59E-22
Ar	3.41E-10	124	1.71E-21
Zn _g	2.59E-10	1348	1.86E-20

TABLE 3

Zinc vapor diffusion coefficients in argon, hydrogen and Ar-H₂ mixtures

Temp., K	D_{Zn-G}, m^2s^{-1}			
	D_{Zn-Ar}	D_{Zn-H_2}	$D_{Zn-Ar-5\%H_2}$	$D_{Zn-Ar+35\%H_2}$
948	2.44E-04	4.30E-04	2.53E-04	3.04E-04
973	2.55E-04	4.57E-04	2.65E-04	3.25E-04
998	2.69E-04	4.82E-04	2.79E-04	3.44E-04
1023	2.84E-04	5.07E-04	2.95E-04	3.62E-04
1048	3.00E-04	5.33E-04	3.12E-04	3.82E-04

The obtained test results show that the rate of evaporation of zinc increases with increasing hydrogen content in the Ar-H₂ gas mixture. This is justified because according to the data presented in TABLE 3, the values of zinc diffusivity in hydrogen are higher than in argon. For example, for a mixture

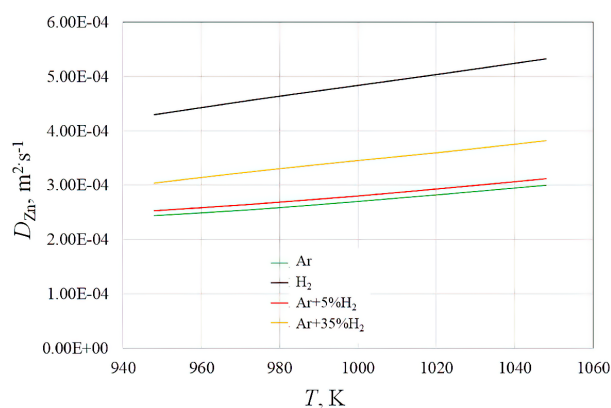


Fig. 6. Zinc vapor diffusivity in argon, hydrogen and their mixtures

TABLE 4

Zinc flux density values

Applied gas atmosphere	Estimated height of free space above the liquid metal, mm	Zinc flux density, $\text{mg}\times\text{cm}^{-2}\times\text{s}^{-1}$	Zinc flux density, $\text{mg}\times\text{cm}^{-2}\times\text{s}^{-1}$
Ar + 5% vol. H ₂	5.5	0.106	0.210
Ar + 15% vol. H ₂	5.5	0.240	0.236
Ar + 35% vol. H ₂	5.5	0.351	0.260
Ar + 15% vol. H ₂	2.5	0.265	0.390
Ar + 15% vol. H ₂	8.1	0.221	0.149

containing 5% hydrogen, the density of the zinc flux was on average $0.106 \text{ g}\times\text{cm}^{-2}\times\text{s}^{-1}$ and for a mixture containing 35% vol. it was $0.351 \text{ g}\times\text{cm}^{-2}\times\text{s}^{-1}$ (TABLE 4). The flux density values determined from the Eq. (3) were of the same level as the values determined from the experimental data. The noticeable differences may result from an error in the estimation of the actual height of the crucible free space above the liquid zinc.

It has been also shown that the evaporation rate decreases with the increase of the free crucible space above the liquid metal (TABLE 4). Fig. 7 presents changes in the density of the zinc flux during selected experiments. They show that as the duration of the experiment increases, the density decreases. This is justified because the height of the crucible free space above the liquid metal increases.

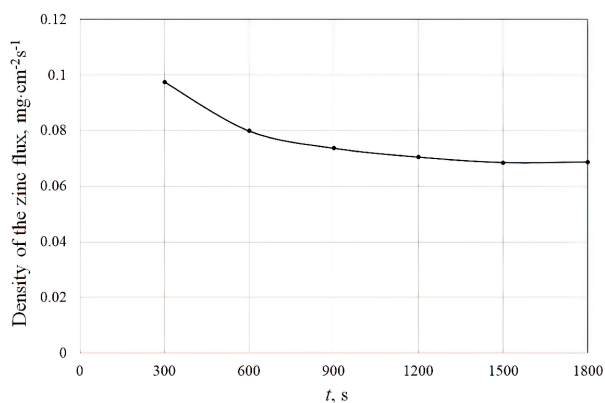


Fig. 7. Changing the density of the zinc flux during the experiments

5. Conclusions

Based on the results of evaporation rate of zinc in gas mixtures with argon – hydrogen, carried out at a temperature of 948 K, it was shown that:

- as the hydrogen content of the mixture increases, the rate of zinc evaporation increases. This is justified because the rate of diffusion of zinc in argon is lower than in hydrogen,
- increase in hydrogen content from 5 to 35% vol. resulted in an increase in evaporation rate from $0.106 \text{ g}\times\text{cm}^{-2}\times\text{s}^{-1}$ to $0.351 \text{ g}\times\text{cm}^{-2}\times\text{s}^{-1}$,
- if the free height of the gas space above the liquid metal increases, the rate of evaporation also increases.

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REFERENCES

- [1] R.G. Ward, Journal of the Iron and Steel Institute **201** (1), 11-15 (1963).
- [2] X. Chen, N. Ito, K. Nakashima, K. Mori, Tetsu-to-Hagane **81**, 959-964 (1995).
- [3] L. Savov, D. Janke, ISIJ International **40**, 95-104 (2000).
- [4] H. Ono-Nakazato, K. Taguchi, T. Usui, ISIJ International **43**, 1105-1107 (2003).
- [5] L. Blacha, J. Mizera, P. Folega, Metalurgija **53**, 51-54 (2013).
- [6] A. Powell, J. Van Den Avyle, B. Damkroger, J. Szekeley, U. Pal, Metallurgical and Materials Transactions **28**, 1227-1239 (1997).
- [7] L. Blacha, S. Golak, A. Jakovics, A. Tucs, Archives of Metallurgy and Materials **59**, 1, 275-279 (2014).
- [8] L. Blacha, R. Burdzik, A. Smalcerz, T. Matuła, Archives of Metallurgy and Materials **58**, 1, 197-201 (2013).
- [9] H. Nakamura, A. Mitchell, ISIJ International **32**, 583-592 (1992).
- [10] E. Fromm, Metallurgical Transactions A. **9A**, 1835-1838 (1978).
- [11] J. Łabaj, M. Jodkowski, Rudy i Metale **63**, 3-8 (2018).
- [12] E.T. Turkdogan, P. Grieveson, L.S. Darken, The Journal of Physical Chemistry **67**, 1647-1654 (1963).
- [13] B. Murphy, Journal of Physics D Applied Physics **43**, 1-31 (2010).
- [14] I. Semenov, I. Krivtsov, V. Demchenko, A. Semenov, et al., Modelling and Simulation in Materials Science and Engineering **20**, 1-12 (2012).
- [15] S. Deng, Q. Guo, H. Wang, et al., The International Journal of Advanced Manufacturing Technology **106**, 4423-4436 (2020).
- [16] Y. Luo, Applied Mathematical Modelling **37**, 6177-6182 (2013).
- [17] R. Fabbro, F. Coste, D. Goebels, M. Kielwasser, Journal of Physics D: Applied Physics **39**, 401-409 (2006).
- [18] T. Zacharia, S.A. David, J.M. Witek, Metallurgical Transactions B **22**, 233-241 (1991).
- [19] R.R.G.M. Pieters, C. Goos, B. Rietman, M. Richardson, Welding in the World **52**, 33-41 (2008).
- [20] R.R.G.M. Pieters, J.G. Bakels, M.J.M. Hermans, G. Den Ouden, Journal of Laser Applications **18**, 199-204 (2006).
- [21] C. Meran, M. Yuksel, A. Gulsoz, T. Sekercioglu, Science and Technology of Welding and Joining **9**, 131-137 (2004).
- [22] F. Huggera, K. Hofmann, S. Steina, M. Schmidta, Physics Procedia **56**, 576-581 (2014).
- [23] F. Gharavi, I. Ebrahimzadeh, K. Amini, B. Sadeghi, Materials Research **4**, 1-12 (2019).
- [24] S. Emami, T. Saeid, Acta Metallurgica Sinica (English Letters) **28**, 766-771 (2015).
- [25] The HSC Chemistry ver. 6.1 database.