Optimal conditions of vacuum distillation process for obtaining the high grade pure magnesium

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In this paper the procedure of obtaining of high purity magnesium by using the vacuum distillation process has been shown. Vacuum distillation process has been conducted under low and high vacuum at different operating temperatures and times. Also, the condenser structure in vacuum equipment is set up in such a way to get the optimal conditions for obtaining of high purity magnesium. Based on experimental results, optimal process parameters were determined which justify the economy and technology of the process.

Key words: *distillation, magnesium, vacuum, high purity*

1. INTRODUCTION

Demand for high purity magnesium (Mg) has been notable in many research fields such as metallurgy, solid state physics, atomic energy, organic chemistry, biology, nanomaterial's, etc. A variety of methods for preparing organomagnesium compounds utilize elemental magnesium, and for many of them the purity and form of the metal is important or even critical. Magnesium is readily available in a purity of 99.8%, and this grade is satisfactory for many routine purposes [1].

Magnesium alloys are currently of great interest, as their extraordinary low density, high specific strength, and easy-recycling ability are very attractive for further applications in aeronautics, electronics and other consumer goods [2–4]. In investigation of these alloys, for instance investigation on the microstructure and mechanical properties, high purity magnesium has been used for alloying with other elements [5, 6]. Also, high purity magnesium has been used as an anode material in seawater activated batteries because he offers several advantages such as a high electrode potential of −2.73 V vs. normal hydrogen electrode, a high faradic capacity, appropriate corrosion rate and low density [7, 8].

The reduction of uranium tetra-fluoride (UF_4) by magnesium with the magnesiothermic reduction is one of the main industrial methods for producing commercial pure uranium ingot. The magnesiothermic reaction is given by [9, 10]:

$$UF_4 + 2Mg = U + 2MgF_2$$
$$\Delta H = -208.71 \text{ kJ/mol (at 640°C)} \qquad (1)$$

Magnesium used for this purpose has to be exempted from elements with with a large neutron cross section such as Boron (B), Cadmium (Cd), Cobalt (Co) etc. These elements gave long-living high activity isotopes (eg. Co^{60}). The content of impurities in high pure magnesium used for this purpose must not exceed the following values [10]:

Fe	25 ppm
Mn	50 ppm
Al	30 ppm
Si	125 ppm
C	100 ppm
Zn	300 ppm
Cl	25 ppm
Co	10 ppm
Ni	10 ppm
Cd	0.5 ppm
B	0.3 ppm

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In this paper we will show a method for refining of technical magnesium (in different shapes and with different chemical compositions) in order to obtain high purity magnesium which satisfy the limitations mentioned below. The most common method for obtaining of high purity magnesium is vacuum distillation process [12, 13].

2. MATERIALS AND METHODS

Refining experiments were conducted in steel crucible which is schematically shown in figure 1. This crucible should satisfy the long term service on high temperatures (up to 800 ^{O}C) and high vacuum (up to $1 \cdot 10^{-3}$ Pa) in evaporator, with achieving of required drop temperature in condenser zone. Evaporator, length of 53 cm, is made of heat resistant Böhler steel, while condenser is made of common carbon steel [11]. Sealing of crucible on the closure was made with a rubber sealant. Crucible is heated in muffle furnace to a target temperature. In evaporator, vessel of heat resistant steel was loaded with crude magnesium. Condenser is cooled with water and placed outside of the furnace in order to obtain low temperatures, corresponding to the condensation of magnesium vapours.

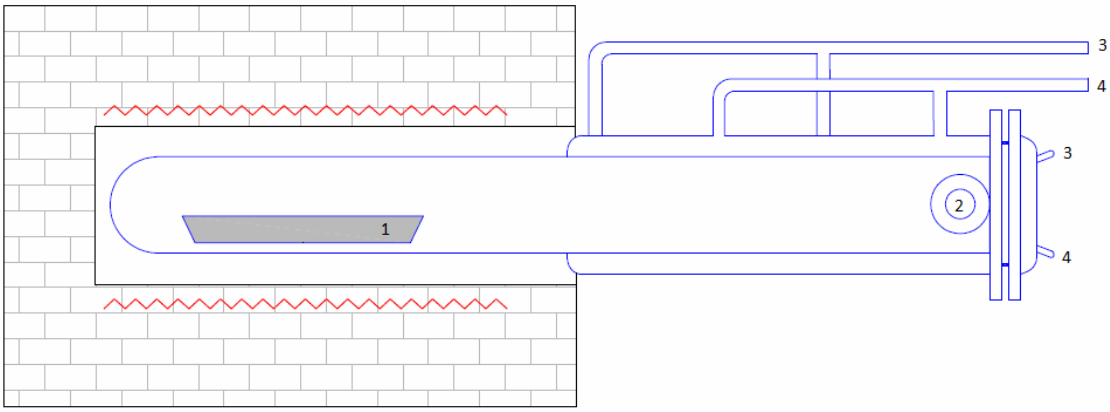

Figure 1 - Horizontal steel crucible for vacuum distillation process: 1-vessel for crude magnesium; 2-connection for vacuum system; 3-water inlet; 4-water outlet

In order to obtain fractional distillation of magnesium and its impurities on different temperatures, multi zone system was installed into condenser by setting of the partition plates of stainless steel. First zone was marked as one immediate to evaporator from the first partition plate. Size and number of zones are variable and can be simply moved and placed in condenser as desired.

Temperature was measured by pyrometer placed inside of stainless steel pipe in such a way to manage temperatures measurement along the entire length of crucible. Vacuum system was connected with crucible via inlet on the condenser. Distillation experiments were performed on the pressures of the low and high vacuum and temperatures from 550-800 ^{O}C at different times.

Crude magnesium used as starting material for testing is divided in three different groups by shapes: chippings, strips and scraps. In experiment, ten samples of each variation of parameter were taken for distillation process, and then mean value was calculated and entered in the table. The mean chemical compositions of these groups are noticeably different and shown in Table 1.

Table 1. Analyses of impurities in crude magnesium, mean values

Group	Chemical composition of impurities, in ppm								
	Fe	Mn	Ni	B	Si	Zn	Cd	Al	C
1 (chippings)	57	400	5	0.86	107	300	0.4	10	5
2 (strips)	86	850	5	0.86	500	1000	0.4	100	5
3 (scraps)	250	162	5	0.2	500	2000	2.01	-	-

Variable parameters in this experimental work are: temperature, vacuum pressure and time of distillation process. The aim of this work is to determine optimal condition of process in aspect of pureness of final product in economic and environmental friendly manner.

3. RESULTS AND DISCUSSION

Experiments for distillation process, with two zone system, were performed on 60 samples (10 for each variation of parameters) of approximately the same amount (from 50 to 110 grams) and temperatures of

550-800 ^{O}C under vacuum pressure of $1.3\cdot10^{-3}$ to 11 Pa (high and low vacuum). The mean values were calculated and represented in Table 2 and 3.

According to the results, represented in Table 2 and 3, lower temperatures of distillation, approximately 600 – 650 ^{O}C, and lower vacuum pressures gives advanced purification of impurities. Also, we can note that the boron content decrease to 0.2 ppm and concentration of Cd remains on 0.4 ppm. Higher concentration of Zn is expected due to the higher vapour pressure of Zn compared to the Mg, but it is possible to depart Zn from Mg by placing of two zone system. Time of complete distillation of magnesium is 40 min on lower vacuum pressure and temperature of 600 ^{O}C, so there is no need for longer times of distillation process.

Processes of distillation with multy-zone system were conducted on high vacuum (with argon working atmosphere) and different temperatures and times along with fraction condensation (Groups: 7-10, Table 4-5). Also, multiple distillation process was conducted under three stages, on temperature of 700 ^{O}C, and vacuum of $1.3\text{-}2.7\cdot10^{-3}$ Pa (Groups: 11', 11'' and 11'' Table 4-5).

Table 2. Parameters of distillation process of crude magnesium, two zone system, mean values

No. of group	Crude Mg, gr	$T_{dis,}$ ^{O}C	$T_{con,}$ per zones, ^{O}C		Time, min	Vacuum, Pa	% Mg per zones	
			I	II			I	II
1	50	610	485	-	90	11	84.5	-
2	60	745	570	465	105	9	35.8	64.2
3	90	800	-	588	45	11	-	100
4	110	550			60	$1.3\text{-}2.7\cdot10^{-3}$	42	58
5	100	600			60	$4\text{-}6.7\cdot10^{-3}$	59	41
6	100	600			40	$4\text{-}6.7\cdot10^{-3}$	100	

Table 3. Chemical composition of impurities in magnesium obtained by distillation process, two zone system, in ppm

No. of group, zone		Fe	Mn	Al	Si	Ni	Co	Cd	B	Zn	Cl	C
1, I		5	9	10	55	5	-	0.4	0.20	50	-	-
2	I	38	200	10	25	5	-	0.4	0.20	50	-	-
	II	5	65	-	25	5	-	0.4	0.20	435	-	-
3, II		9.2	9.2	10	25	5	5	0.4	0.20	14	-	-
4	I	10	20	10	500	5	5	0.4	0.30	-	-	-
	II	10	14	10	210	5	5	0.4	0.20	2000	-	-
5	I	10	20	12	50	5	5	0.4	0.84	-	-	-
	II	250	154	20	77	5	5	0.4	0.20	2000	-	-
6, I and II		10	20	10	500	-	0.5	0.4	0.2	-	-	-

Table 4. Parameters of distillation process of crude magnesium, multy-zone system, mean values

No.	In. Mg, gr	$T_{dis,}$ ^{O}C	$T_{con,}$ per zones, ^{O}C					Time min	Vac., Pa $\cdot10^{-3}$	% Mg per zones, Residue				
			I	II	III	IV	Res.			I	II	III	IV	Res.
7	270	590	515	485	-	-	590	30	3.7	20.2	10	-	-	69.8
8	160	660	525	430	-	-	660	40	4.3	19	66	-	-	18
9	300	700	625	588	470	400	700	50	7.3	26.1	34.3	31.9	2.7	5
10	70	800	-	610	550	510	-	60	2.7	-	42	40	18	-
11'	110	700	620	590	480	400	700	60	1.3	30	29.5	25	9.5	6
11''	75	700	620	590	480	400	700	60	1.3	35.3	30.2	24.3	4.2	6
11'''	54	700	630	600	480	405	700	60	2.7	38	33.2	21.1	3.5	4.2

According to the results, represented in tables 4 and 5, it is observed that with increasing of temperature, during the fractional distillation process, the number of zones rises.

Impurities in the condensate do not vary greatly, both in multy zone system whit one stage and with two and three stages.

Table 5. Chemical composition of impurities in magnesium obtained by distillation process, multy-zone system, in ppm

No. of group, zone		Fe	Mn	Al	Si	Ni	Co	Cd	B	Zn	Cl	C
7	I	5	10	10	25	5	5	0.4	0.2	76	-	-
	II	10	10	10	25	5	5	0.5	0.2	125		
8	I	9	20	10	96	5	3	0.4	0.2	50	-	-
	II	5	10	10	25	5	5	0.4	0.2	125		
9	I	5	8	10	25	5	5	0.4	0.2	30	-	-
	II	5	10	10	25	5	5	0.4	0.2	59	-	-
	II	5	10	10	10	5	5	0.4	0.2	50	-	-
	IV	5	10	10	20	5	5	0.5	0.2	96	-	-
10	II	115	30	50	100	5	0.5	0.4	0.2	34	-	-
	III	39	25	50	80	5	0.5	0.4	0.3	81		
	IV	20	25	50	60	5	0.5	0.5	0.3	200	-	-
11’	mean	10	20	10	50	-	5	0.5	0.3	150	-	-
11’’	mean	10	20	10	50	-	5	0.4	0.3	90	-	-
11’’’	mean	10	20	10	50	-	5	0.4	0.3	50	-	-

4. CONCLUSION

Based on the presented experiments of vacuum distillation process for producing of high purity magnesium, we can conclude the following:

- The optimum conditions for distillation of crude magnesium are: temperatures of 600-660 ^{O}C, vacuum pressure of 1.3-4.3·10^{-3} Pa and time of 40 min, with two zone system.
- For removing of Zn and Cd, higher temperatures of distillation are required, from 700-750 ^{O}C in order to capture high concentration of Zn and Cd in last, fourth zone, which can be removed.
- Multiple distillation process is reasonable only for removal of volatile impurities such as Zn, Cd and alkaline metals.

This method is simple and efficient in technological, environmental and economic point of view. The main disadvantage of this method is high initial investment in the equipment, which can be offset with price of high grade pure magnesium.

5. ACKNOWLEDGEMENT

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REZIME

OPTIMALNI USLOVI PORCESA VAKUUMSKE DESTILACIJE ZA DOBIJANJE MAGNEZIJUMA VISOKE ČISTOĆE

U ovom radu opisana je procedura dobijanja metalnog magnezijuma visoke čistoće, korišćenjem tehnologije vakumske destilacije. Ispitivan je uticaj različitih parametra temperature i vremena vakuumske destilacije, kao i uticaj niskog i visokog vakuma na proces dobijanja magnezijuma visoke čistoće. Takođe, struktura kondezatora, u opremi za vakuumsku destilaciju, je podešavana na takav način da se dobijaju optimalni uslovi procesa vakuumske destilacije. Na osnovu eksperimentalnih ispitivanja, određeni su optimalni parametri procesa vakuumske destilacije, koji opravdavaju ekonomiju i tehnlogiju procesa.

Ključne reči: *destilacija, magnezijum visoke čistoće, vakuum*