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X-Ray Analysis in the Optimization of the Sintering Process of $SmCo_5$ Magnets

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Abstract: *The content of the $SmCo_5$ phase, as the carrier of magnetic properties, was controlled by X-ray diffraction analysis. The content of the $SmCo_5$ phase, as a function of sintering time and temperature, under constant heat treatment conditions, was observed through the intensity of its most significant diffraction peak, which corresponds to the (111) plane. By correlating these parameters with a mathematical treatment, a mutual dependence was established. A regression dependence was obtained that showed that the intensity of the diffraction peak of the (111) plane of the $SmCo_5$ phase depends upon the squares of both the sintering time and temperature, for given heat treatment conditions. It is possible to optimize the sintering conditions by calculating values of sintering time and temperature for which this dependence has its maximum.*

Keywords: *$SmCo_5$ magnet; X-ray diffraction; Sintering; Modelling*

Резюме: *При помощи метода рентгеноструктурного анализа наблюдали содержание $SmCo_5$-фазы в качестве носителя мегнитных свойств. При этом проведен анализ диффракционного максимума, соответствующего плоскости (111) в зависимости от температуры и времени спекания при постоянных условиях термической обработки. С помощью корреляционного анализа установлена взаимозависимость этих параметров. Полученное регрессионное соотношение показывает, что интенсивность диффракционного пика от плоскости (111) для $SmCo_5$-фазы зависит от квадратов как температуры, так и времени спекания для заданных условий термической обработки. Оптимальные условия спекания соответствуют расчетным значениям температуры и времени спекания, при которых указанная зависимость достигает максимума.*

Ключевые слова: $SmCo_5$ *магнит; моделирование; спекание; рентгеноструктурный анализ.*

Садржај: *У проведеним истраживањима садржај $SmCo_5$ фазе као носиоца магнетних својстава праћена је рендгенском дифрактометријском анализом и то преко њеног најизраженијег дифракционог пика који одговара (111) равни, а у зависности од температуре и времена синтеровања за константне услове термичке обраде.*

Корелацијом ових параметара и њиховом математичком обрадом утврђена је међусобна зависност. Добијена регресиона зависност показује да интензитет пика на (111) равни за $SmCo_5$ фазу зависи како од квадрата температуре, тако и од квадрата времена синтеровања за дефинисане услове термичке обраде. Израчунавањем вредности температуре и времена синтеровања за које добијена зависност има максимум, омогућена је оптимизација услова синтеровања.
Кључне речи: *$SmCo_5$ магнет; рендгенска дифракција; синтеровање; моделовање.*

Introduction

All rare earth permanent magnets contain more than two elements and are multiphase, metastable metallurgical systems with complex microstructures. Without making a serious mistake, [1] sintered $SmCo_5$ magnets may be considered as binary alloys. Non-metallic impurities, primarily oxygen, are inevitable during production, but when pressing and sintering are carried out in a proper manner, the sintered bodies are of high density without connecting open pores. This enables a drastic lowering of the oxygen content, except in the surface layer. [1-3]

The technological procedure for obtaining sintered $SmCo_5$ magnets, includes, as a usual production step, machining. There are two reasons for machining. The first is to obtain appropriate geometry and the second is to remove the surface layer, enabling full magnetic contact. During this operation the oxide layer is removed.

In the final sintered $SmCo_5$ magnets, beside the solid intermetallic $SmCo_5$ phase, various Sm-Co phases are present, as minor secondary phases. $SmCo_5$ is the principal flux producing phase in the magnets. The minor phases with different ratios of Sm (Sm:Co from 2:17 down to 1:3) influence the coercivity and its temperature dependence. [1]

The most frequently occurring secondary phase is Sm_2Co_7, the presence of which can be identified by metallographic investigation, with SEM and EPMA [4], while its detection by X-ray diffraction is complicated by the similarity of the interplanar spacing of the $SmCo_5$ and Sm_2Co_7 phase [5]. Only the diffraction peaks of $SmCo_5$ phase can be identified on the diffractograms of well sintered $SmCo_5$ magnets

When all these facts are take into consideration, the decision to observe the quality of sintered $SmCo_5$ magnets through the intensity of the diffraction peaks of the $SmCo_5$ phase, is an acceptable approximation. In this investigation, the most significant diffraction peak, which corresponds to the (111) plane of the $SmCo_5$ phase, was correlated to the sintering parameters, for heat treatment at 920°C.

In a previous paper, [6] it was established that for heat treatment at 900°C, the intensity of the most significant diffraction peak, corresponding to the (111) plane of $SmCo_5$ phase, [7] depends on the squares of both the sintering time and temperature.

The same principals of regression analysis are used in this work, but the conditions of heat treatment were different (920°C).

Experimental

Commercial $SmCo_5$ powder, prepared by the Reduction-Diffusion process was used as the starting powder in this work. A schematic presentation of the investigated technological procedure, together with the applied technological parameters, is given in Fig.1.

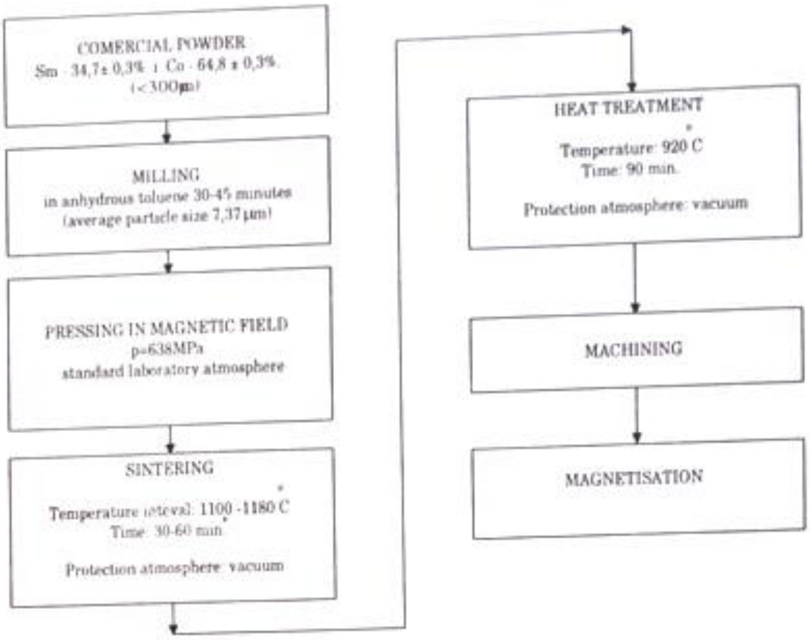

Fig.1 Schematic presentation of the investigated technological process of obtaining sintered $SmCo_5$ magnets

Sintering at temperatures of 1100, 1120, 1140, 1160 and 1180 °C, for sintering times of 30, 45 and 60 minutes, was investigated. Sintering and heat treatment was performed in an electroresistant Lenton furnace. The furnace had a maximal working temperature of 1500°C, electronic programming and temperature control with a precision of ±1°C. At the temperature of 1100°C, sintering was only investigated for 60 minutes, and at 1180°C only for 30 minutes. Heat treatment, for all the investigated sintering times and temperatures, was always at 920°C for 90 minutes. The specimens were rapidly cooled to room temperature after heat treatment.

The presence of the $SmCo_5$ phase, after the investigated sintering conditions followed by constant heat treatment conditions, was observed using powder X-ray diffraction analysis of the polycrystalline specimens. The X-ray analysis was performed using a PHILIPS PW 1710 diffractometer and a copper anticathode (λ=0.154178nm).

Results and Discussion

The intensity of the most significant diffraction peak, which corresponds to the (111) plane of the $SmCo_5$ phase [7], after all the investigated sintering temperatures and time intervals, are given in Tab. I.

A comparative presentation of the diffractograms of the sample after sintering at a temperature of 1140°C, for different times is given in Fig. 2. The heat treatment for all the sintered samples was the same- 90 minutes at 920°C. The first four most important diffraction peaks for the $SmCo_5$ phase are marked on the diffractograms.

The trend of the change in the intensity of the (111) plane peak of $SmCo_5$ with time, is graphically indicated.

Tab. I. Dependence of the intensity of the diffraction peak of the (111) plane of the $SmCo_5$ phase, on the sintering parameters

Sintering temperature, °C	1100	1120	1120	1120	1140	1140	1140	1160	1160	1160	1180
Sintering time, min.	60	30	45	60	30	45	60	30	45	60	30
Peak intensity of the (111) plane, Cts	22	26	31	38	35	48	31	44	36	28	21

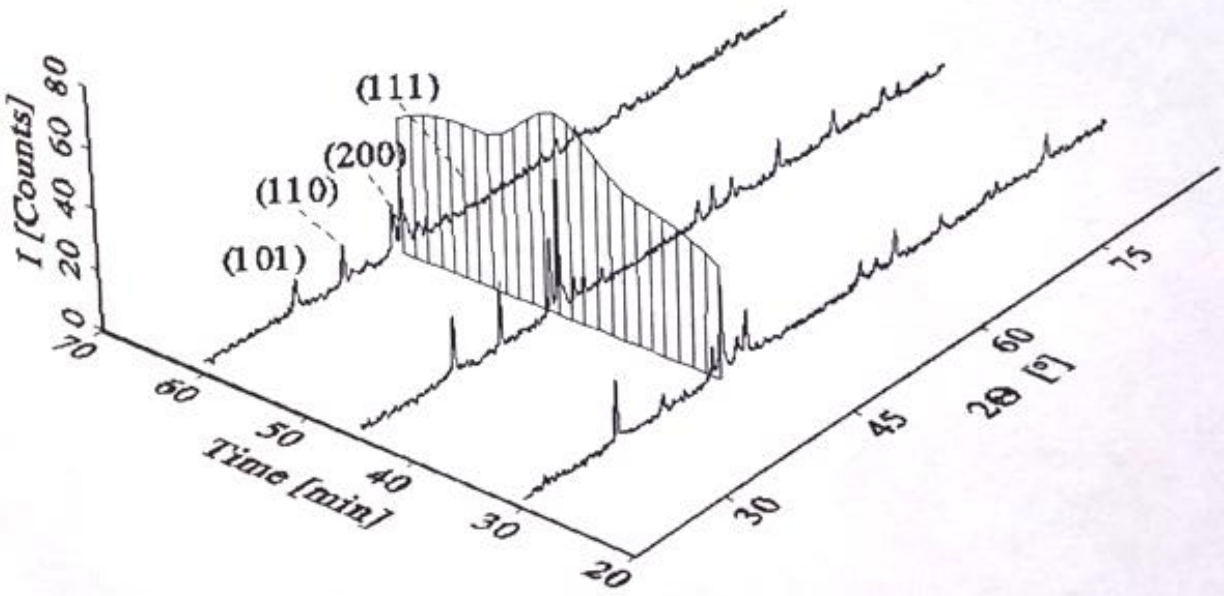

Fig.2 Trend of the change in the intensity of the (111) plane peak of $SmCo_5$ with time, for samples sintered at 1140°C

The experimental data obtained by X-ray analysis was treated by mathematical method [8-10]. The independent variables were time and temperature of sintering, while the dependent variable was the intensity of the most expressed peak, which corresponds to the (111) plane of the $SmCo_5$ phase. The simplest non-linear dependences were investigated to establish the most appropriate one.

The investigated regressional equations of the dependence of peak intensity upon sintering time and temperature are given in Table II, together with calculated average square errors . The calculated values of temperature and time of sintering for which these functions have maximums are also given in Tab. II.

Tab. II. The investigated regressional equations for the dependence of the peak intensity of the (111) plane of the $SmCo_5$ phase on the sintering time and temperature

No.	Regressional equations	Average square error	t_{max}, °C	τ_{max}, min.
1.	$k_1 t^2+k_2\tau^2+k_3 t+k_4\tau+k_5$	5,50572	1141,7	44,1
2.	$k_1 t^2+k_2\tau^2+k_3 t \tau+ k_4$	7,26607	-	-
3.	$k_1 t^2+k_2\tau^2+k_3 t \tau+k_4t+ k_5\tau+ k_6$	3,78799	1141,9	43,26
4.	$k_1 t^2+k_2\tau^2+ k_3$	8,06691	-	-

In Table 2., k_1, k_2, k_3, k_4, k_5 and k_6 are parameters; t - the sintering temperature in °C ; τ- the sintering time in minutes ; t_{max} - the sintering temperature for which the diffraction peak of the (111) plane has a maximum; τ_{max} -sintering time temperature for which the diffraction peak of the (111) plane has a maximum.

Regressional equations No. 2. and 4. do not have a maximum in the investigated interval, and for this reason, as well as the greater average square errors, these equations are not considered to be adequate ones. For dependencies of No.1 and 3, the temperatures and times of sintering after which the diffraction peak of the (111) plane has a maximum, are similar.

It is obvious that the smallest average square error is obtained when equation No. 3 is used. This function was chosen as the most appropriate, and is represented by the regression equation (1):

$$I= -0,0175\, t^2 -0,0220\, \tau^2 - 0,0205\, t\, \tau +40,8538\, t + 25,3125\, \tau -23830 \qquad (1)$$

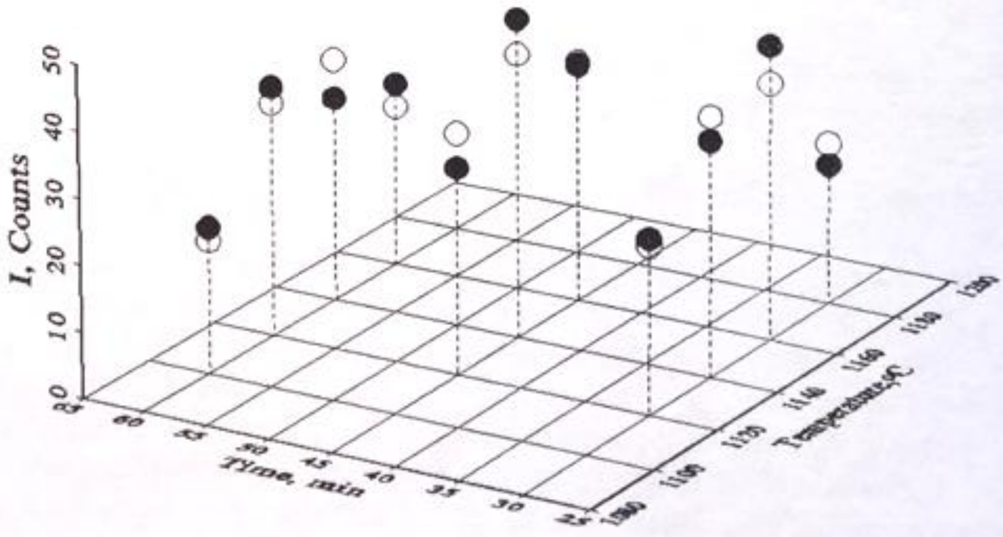

Fig.3. The intensity of the (111) plane diffraction peak of the $SmCo_5$ phase: experimental (●), according to the model(○)

A graphical representation of the dependence of the experimental results (black circles), obtained by measuring the intensity of the (111) plane peak of the $SmCo_5$ phase, on sintering temperature and time, as well as the values obtained using the regression equation (1) (white circles) is shown in Fig. 3. Heat treatment, for all the investigated sintering times and temperatures, was always at 920°C for 90 minutes.

A graphical representation of the regression dependence (1) is given in the Fig. 4. It is obvious that the intensity the intensity of the diffraction peak of the (111) plane of the $SmCo_5$ phase has a maximum in the interval 1141-1142 °C, and the sintering time in the interval 43-44 minutes.

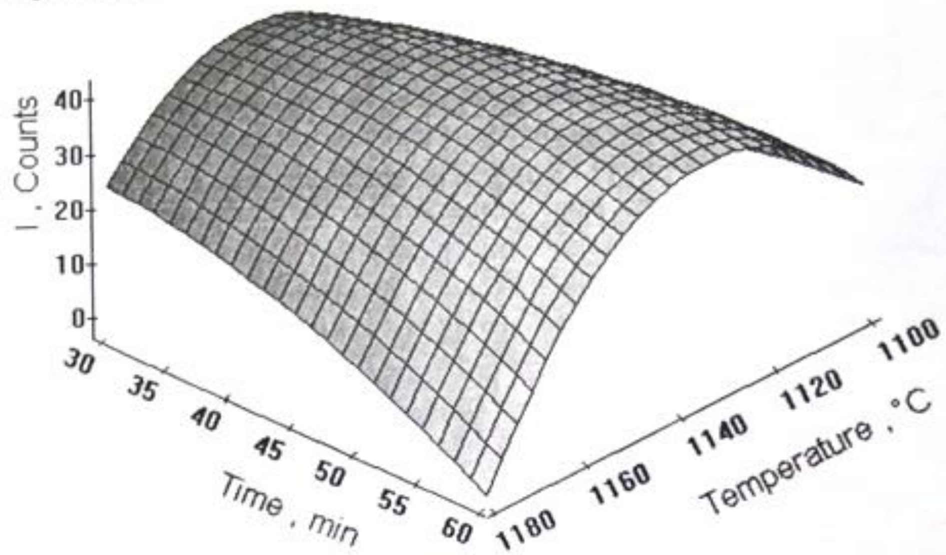

Fig. 4. Graphical presentation of the regressional model

Experimentally it was confirmed that the best results are obtained when sintering is performed under conditions resulting in the maximum intensity of the diffraction peak for the (111) plane.

The following conclusion may be drawn:

A. As the intensities of the diffraction peaks are small, it is more appropriate to say that there is a trend instead of a dependence. However, it is obvious (Fig.4.) that the intensity of the diffraction peak of the (111) plane of the $SmCo_5$ phase has a parabolic dependence on the time and temperature of sintering. The obtained regression model shows that the intensity of diffraction peak depends on the squares of both the sintering time and temperature for a given heat treatment.

B. Using this model, the highest intensity of the diffraction peak of the (111) plane of the $SmCo_5$ phase, is obtained using a sintering temperature of 1141-1142°C and a sintering time 41-42 minutes. It was confirmed experimentally that the best magnets are obtained using such sintering conditions.

C. It was also confirmed experimentally that it is possible to optimize the sintering parameters for the production of the investigated $SmCo_5$ magnet with only few experimental data using the established regressional equation.

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