Electron Beam Melting and Refining of Copper

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Abstract: *Experimental results for the quality of electron beam melting and refining of pyro-refined copper samples are analysed by an empirical model estimation approach. Investigation of the influence of the process parameters – melting power and time of refining on the removal efficiency of specific impurities, overall removal efficiency and material losses is performed.*

Keywords*: ELECTRON BEAM REFINING, COPPER, REMOVAL EFFICIENCY, MATERIAL LOSSES, ELECTRON BEAM MELTING*

1. Introduction

Copper (Cu) is one of the most important metals used in modern life and up to 50% of Cu production is used for making cables, heat exchangers, vacuum equipment, etc., and about 40% is used for production of different alloys with: Zn, Sn, Al, Ni and other valuable metals, which are needed for a number of industries.

The electron beam melting and refining (EBMR) of metals and alloys in vacuum is done using electron beam as a heating source. The aim of the metallurgical refining processes is the removal of metallic impurities and reduction of non-metallic components from the raw material. The increasing demand for purity of the produced materials has led to the use of water-cooled copper crucible in furnaces during EBMR, where the initial material is melted, refined and re-solidified without pollution at using a ceramic pot, and thus EBMR method is superior to the refining methods of the conventional metallurgy [1, 2].

In this paper, investigation of the influence of the process parameters – melting power and refining time on the refined quality characteristics – the removal efficiency of specific impurities, overall removal efficiency and the material losses, is performed based on empirical model estimation.

2. Response surface methodology

Response surface methodology is a group of mathematical and statistical procedures used in fitting an empirical model to the experimental data obtained in relation to an experimental design [3]. Regression models or the polynomial models of some order, giving an adequate functional relationship between a response of interest y (performance characteristic) and a number of associated control (or input) variables (process parameters) $x_1, x_2, \dots x_m$, are estimated:

$$\hat{y}(\vec{x}) = \sum_{i=1}^{k} \hat{\theta}_i f_i(\vec{x}), \tag{1}$$

where $\hat{\theta}_i$ are the estimates of the coefficients in the regression model.

The natural values of the factors (z_i) in the regression models are coded in the region [-1 ÷ 1] and the relation between the coded (x_i) and the natural values (z_i) is given by:

$$x_i = (2z_i - z_{i,max} - z_{i,min})/(z_{i,max} - z_{i,min}), \tag{2}$$

where $z_{i,min}$ and $z_{i,max}$ are the corresponding values of the minimum and the maximum of the process parameters during the experiment.

3. Experimental conditions

Electron beam melting and refining (EBMR) of Cu was performed using 60 kW equipment with one electron gun and a water-cooled copper crucible with diameter of 60 mm. The vacuum pressure in the melting chamber was in the range 5-8×10^{-3} Pa.

Two EBMR process parameters are varied in order to investigate the EBMR process of copper - z_1 is the electron beam power and z_2 – the refining time. The values of the electron beam powers were: 7.5 kW, 10 kW and 15 kW, while the refining times were set to different values in the range between 3 and 30 minutes [4, 5]. Fifteen experiments were performed for different combinations of these process parameters. The measured quality characteristics are the following: the removal efficiency of Sn (y_1, %), removal efficiency of Ag (y_2, %), removal efficiency of Zn (y_3, %), overall removal efficiency (y_4, %) and the material losses (y_5, g).

4. Estimation of regression models

The dependencies of the quality characteristics of the refined Cu: y_1 – the removal efficiency of Sn [%], y_2 – the removal efficiency of Ag [%], y_3 – the removal efficiency of Zn [%], y_4 – the overall removal efficiency [%] and y_5 – the material losses [g] on the variation of the process parameters: z_1 – the melting power [kW] and z_2 – the refining time [min] are estimated.

They are presented for coded in the region [-1 ÷ 1] values of the process parameters in Table 1, together with the square of the multiple correlation coefficient R^2 and the square of the adjusted multiple correlation coefficient R^2_{adj}. The values of both coefficients are high (maximum 100 %) and the estimated models can be considered as good enough for prediction and investigation.

Table 1: *Regression models.*

	Regression models	R^2 [%]	R^2_{adj} [%]
$\hat{y}_1(\vec{x})$	$99.388388 + 0.96370857x_1 + 0.08831581x_2 - 0.3525x_1^2 - 0.09437149x_1x_2$	99.89	99.84
$\hat{y}_2(\vec{x})$	$97.442395 + 0.05581727x_1 - 0.03479241x_2 - 0.15916955x_1x_2$	95.01	93.64
$\hat{y}_3(\vec{x})$	$94.047439 - 7.8928562x_2 - 7.120348x_1x_2 - 6.641097x_1x_2^2$	73.97	66.87
$\hat{y}_4(\vec{x})$	$98.759141 - 4.3757779x_2^2 - 3.1445406x_1x_2 + 4.3840616x_1^2x_2 + 4.2965256x_1x_2^2$	80.73	73.02
$\hat{y}_5(\vec{x})$	$9.638259 + 8.4167181x_1 + 7.4642417x_2 + 6.5539856x_1x_2$	99.99	99.99

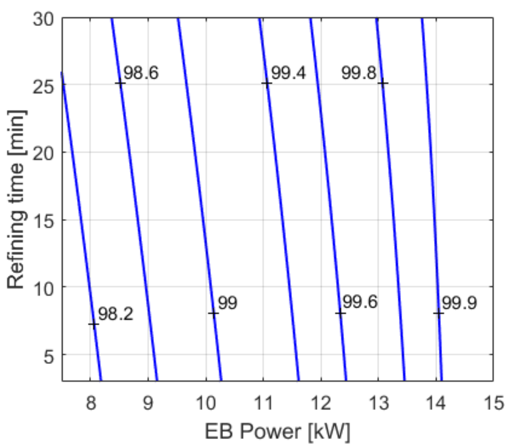

Fig. 1 *Contour plot of the removal efficiency of Sn – y_1, depending on the variation of the melting power (z_1) and the refining time (z_2).*

The obtained by QstatLab [6] regression models for the investigated quality characteristics are used for the investigation of the process of electron beam refining of Cu.

In Fig. 1 – Fig. 3 contour plots of the removal efficiency of Sn – y_1, removal efficiency of Ag – y_2 and removal efficiency of Zn – y_3 depending on the variation of melting power (z_1) and the refining time (z_2) are presented.

From Fig. 1 it can be seen that the removal efficiency of Sn is 99.9%, if the EB power is higher than 14 kW regardless of the chosen refining time in the investigated range.

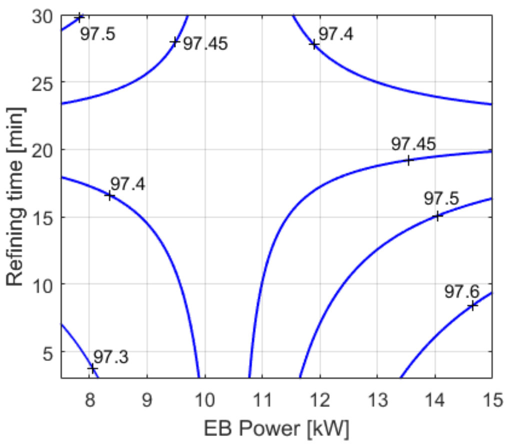

Fig. 2 *Contour plot of the removal efficiency of Ag – y_2, depending on the variation of the melting power (z_1) and the refining time (z_2).*

Fig. 2 shows the contour plot of the removal efficiency of Ag. It can be seen that the removal efficiency is 97.4% or more, but the maximum value (97.6%) is obtained in the range of working regimes where the refining time is less than 6 [min] and the EB power is higher than 14 kW.

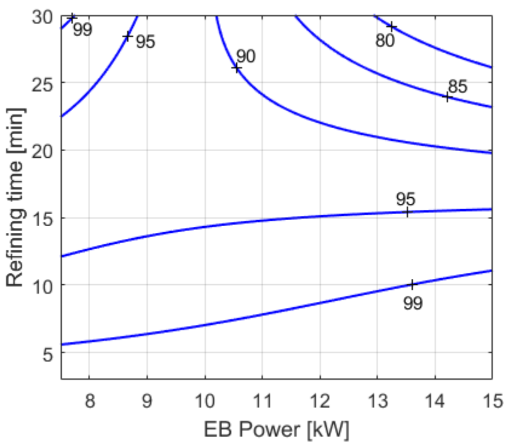

Fig. 3 *Contour plot of the removal efficiency of Zn – y_3, depending on the variation of the melting power (z_1) and the refining time (z_2).*

From Fig. 3 it can be seen that the removal efficiency of Zn can be 99%, if the refining time is equal to 5 min regardless of the chosen EB power or for EB powers higher than 13 kW and refining time less than 8 min.

The contour plot shown in Fig. 4 represents the overall removal efficiency (y_4) depending on the variation of melting power (z_1) and the refining time (z_2). It can be seen that the highest overall removal efficiency can be obtained, if the refining time is in the region from 17 min to 23 min and the EB power is set on its minimal value (less than 8 kW) or the EB power is higher than 14.7 kW and the refining time is more than 20 min.

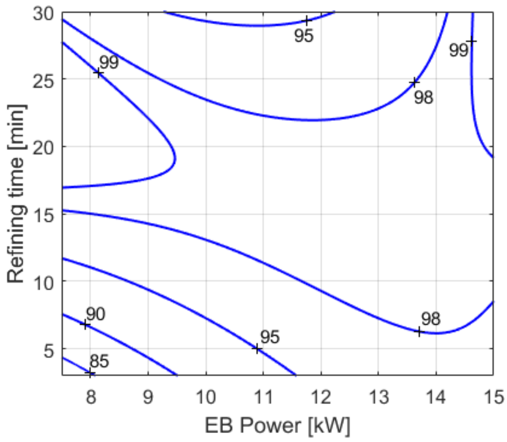

Fig. 4 *Contour plot of the overall removal efficiency – y_4, depending on the variation of the melting power (z_1) and the refining time (z_2).*

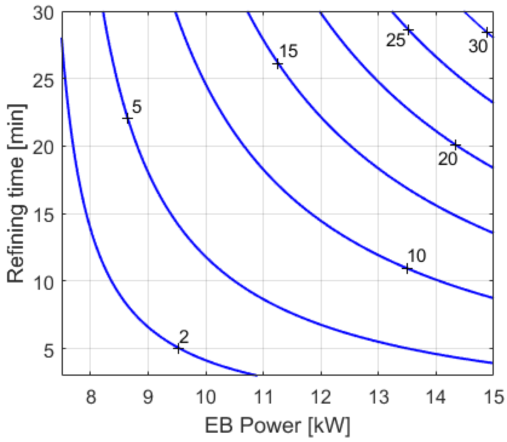

Fig. 5 *Contour plot of the material losses – y_5, depending on the variation of the melting power (z_1) and the refining time (z_2).*

Fig. 5 represents the investigation of the material losses during the process electron beam melting and refining of pyro-refined copper. From the contour plot it can be seen that the losses under 2 g can be obtained, if the EB power is less than 8.5 [kW] and the refining time is less than 7 min.

5. Correlation analysis

Another way to analyze the removal efficiencies of different inclusions is to analyze the coincidence of the refining process parameters in direction of simultaneously removing the inclusions or if a certain set of process parameters favors the refining of one of the inclusions at account of another inclusion. Correlation analysis can answer the question for presence of such correlation between different pairs of inclusions.

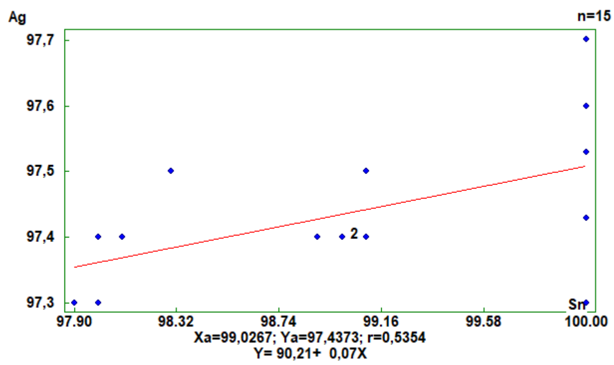

Fig. 6 *Scatter plot of the removal efficiency of Sn – y_1 and Ag – y_2.*

In Fig. 6 – Fig. 8 the scatter plots for the different combinations of inclusion removal efficiencies obtained by QstatLab [6] are presented.

From Fig. 6 it can be seen that there is moderate positive linear relationship between the removal efficiencies of Sn and Ag. The correlation coefficient is r_1 = 0.54 and performed test for significance defines it as a significant correlation. This means, that the choice of EBMR process parameters that favor the refining of Sn will favor the refining of Ag as well.

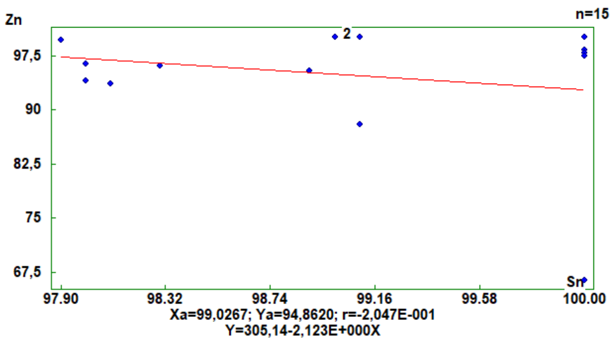

Fig. 7 *Scatter plot of the removal efficiency of Sn – y_1 and Zn – y_3.*

Fig. 7 shows that there is no significant correlation between the removal efficiencies of Sn and Zn. The Pearson linear correlation coefficient is small and insignificant.

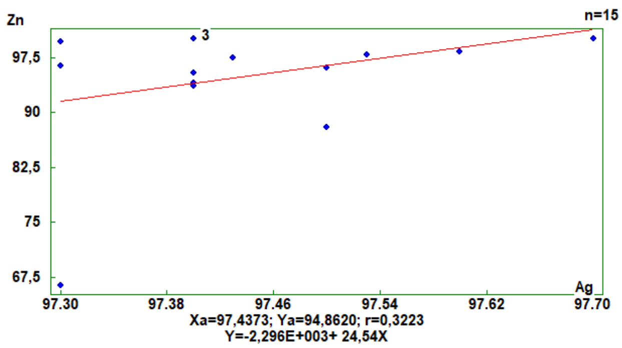

Fig. 8 *Scatter plot of the removal efficiency of Ag – y_2 and Zn – y_3.*

Fig. 8 shows week positive correlation between the removal efficiencies of Ag and Zn – the Pearson linear correlation coefficient is 0.32. After making hypothesis testing for its significance it is proven as insignificant correlation coefficient.

Consequently, the only significant correlation found is between the removal efficiencies of Sn and Ag. The choice of optimal EBMR refining process parameters should be made according the technological requirements for all quality characteristics and compromise solutions should be found that favors the refining of one or another of the inclusions to a certain extend.

5. Conclusions

Regression models for the removal efficiency of specific impurities, overall removal efficiency and the material losses depending on the variations of the electron beam melting and refining process parameters at Cu melting were estimated.

The obtained regression models can be used for future investigation and optimization of the process electron beam melting and refining of pyro-refined copper.

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