

Current density optimization in the copper electrorefining process

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The way of estimating the costs of the electrorefining of copper and of selecting the optimum electrolysis current density has been presented. The analysis has been carried out for the flow electrolyser taking into account the capital costs, electrical energy costs, heating or cooling costs and the electrolyte pumping costs. Against a background of the present literature data the values of the above-mentioned cost coefficients have been estimated and the optimum current densities determined, and also the economic reasons discussed. The method presented does not depend on the system scale.

1. Introduction

At present the optimization of technical-chemical processes plays a very important role. Its purpose is to choose the process parameters so as to obtain the required amount of product of the necessary quality at minimum production costs.

Electrorefining of copper is now the most important purifying process of this metal. The rapidly increasing demand creates the necessity to look for new technological solutions of process intensification. The output of the electrolyser unit area is directly proportional to the current density used and therefore a current density increase seems to be the simplest way of intensification of the process discussed. Nowadays, most of the plants in the world work at current densities ranging from 200-400 A m⁻² [1].

It is the consensus of opinion, however, that increased current density results not only in the increase of electric energy consumption but also has a detrimental influence on the quality of the cathode deposits and the increase of losses of the noble metals. These last two phenomena could be avoided by the electrolyte circulation which results in, however, an increase of the process costs. On the other hand, the higher current density results in substantial savings, because of the greater efficiency of the unit of the cathode area, decrease of the cost of heating of the electrolyte by steam, decrease of the costs of the capital lock-up in the

equipment and in the processed metal and related interest on capital.

The problems relating to the optimization of the copper electrorefining process were the subject of interest for many researchers. Most of the papers are based, however, on the out-of-date data contained in the works of Hoffman-Schiffner [2]; several newer papers [3-5] do not cover this entire complicated problem. In this paper the formulas determining the optimum current density depending on different factors and cost coefficients have been given. The assumption of very broad limits of variation of these coefficients makes it possible to compare the data obtained with the industrial experiences obtained in different local conditions.

2. Economics of electrorefining

The analysis of the costs of the electrolytic refining of copper divides into the following components.

2.1. Capital costs

These costs consist of three parts:

(a) Investment costs determined by the production output and time, and in the first approximation independent of the magnitude of electrodes and current density used; to these costs belong also the costs of electricity and control-measurement

equipment installation, of electrolyte exchange system, wages, the general costs and repairs;

(b) Variable investment costs depending on the area of the electrodes and current density used; to these costs belong the costs of building the plants, plant amortization cost and interest cost;

(c) Cost of metal being used in the refining process and the cost of its interest; these costs decrease with the increase of the current density used.

2.2. Electrical energy cost

This cost is proportional to the current density used and depends on the local energy price. It also depends on the cost of rectifying and on the interest on the rectifying equipment.

2.3. Cost of the heating steam

Most of the electrical energy being used is being changed into heat, and therefore the steam consumption decreases with the current density increase. Wallden *et al.* [6] pointed out that the amount of heat emitted at the current density of about 560 A m^{-2} is large enough, and there is no need to heat the electrolyte additionally by steam.

2.4. Costs of pumping

These are related to the energy for pumping the electrolyte and to the interest and amortization of the capital invested in this equipment.

3. Proposed economic model of the electrolyser

Let us assume that electrodes of different signs are placed alternately in the electrolyte bath. Two neighbouring electrodes and the space between them make the rectangular channel. Let us assume that we are dealing with the channel electrolyser in which two electrodes of area A , and length l are placed at a distance h from each other. Let us denote the current density by j , and the difference of potentials by ϕ . Let Δp be the drop of the liquid pressure along the electrolyser. The hydrodynamics of a channel system of this type have been presented in [7]. In the case of the work of the electrolyser it is necessary to keep the temperature of the electrolyte at a constant value. Assum-

ing that in the given stabilized conditions Q_d is the total heat lost by the electrolyser in unit time and by balancing the energy the following equation is obtained

$$Q_d = \phi j A \pm M \sigma_1 \quad (1)$$

where M is the quantity of the heating or cooling medium flowing through the electrolyser heat exchanger, expressed in kg s^{-1} , and σ_1 is the amount of heat delivered by the heating medium (sign +) or received from the cooling medium (sign -) expressed in J kg^{-1} . The first term on the right-hand side of Equation 1 expresses the amount of heat emitted in the electrolyser as a result of the current flow. After transformation of Equation 1 the following expression for the heating (cooling) medium flow is obtained

$$M = \pm (Q_d - \phi j A) / \sigma_1. \quad (2)$$

The operation cost of the discussed system depends on the factors discussed in Section 2, to which the individual cost coefficients could be attributed

k_c capital cost coefficient ($\text{\$ m}^{-2} \text{s}^{-1}$),
 k_e electric energy cost coefficient ($\text{\$ m}^{-2} \text{s}^{-1}$),
 k'_s heating (cooling) medium cost coefficient ($\text{\$ kg}^{-1}$),
 k_p pumping cost coefficient ($\text{\$ J}^{-1}$).

The cost of operating the process per unit time, can then be expressed as

$$K = k_c A + k_e j A \phi + k_p \frac{\Delta p}{l} V h A + k'_s |M|. \quad (3)$$

The first term represents the capital costs per unit time, the second one the costs per unit time associated with the electrical resistive losses, the third one the pumping cost per unit time, and the fourth one the heating (or cooling) cost. It is convenient to employ the costs for making the product calculated per coulomb of the admitted charge, in the electrochemical systems. This is easily achieved by dividing both sides of Equation 3 by the total electrolysis current jA . Substituting Equation 2 into Equation 3 the following equation is obtained

$$k = \frac{k_c}{j} + k_e \phi + k_p \frac{\Delta p}{j l} V h + k'_s \left| \frac{Q_d}{j A \sigma_1} - \frac{\phi}{\sigma_1} \right|. \quad (4)$$

After writing out the modulus in Equation 4 and denoting the ratio $Q_d/A = q$ and $k'_s/\sigma_1 = k_s$ and after transformation the following expressions are obtained

$$\text{for } \frac{q}{j} < \phi$$

$$k = (k_c + k_s q)/j + (k_e - k_s)\phi + k_p(\Delta p/jl) Vh \quad (5)$$

$$\text{for } \frac{q}{j} > \phi$$

$$k = (k_c - k_s q)/j + (k_e + k_s)\phi + k_p(\Delta p/jl) Vh \quad (6)$$

$$\text{for } \frac{q}{j} = \phi$$

$$k = k_c/j + k_e\phi + k_p(\Delta p/jl) Vh. \quad (7)$$

The equation of the form analogous to Equation 7 was discussed in [7]. Let us introduce after [7] the dimensionless parameters $(Sh)^*$ and f^* , defined by the equations

$$(Sh)^* = \frac{1}{\alpha F D c} \left(\frac{k_c^* \sigma h}{k_e^*} \right)^{1/2} \quad (8)$$

$$f^* = \left(\frac{2\rho^2 k_c^*}{k_p} \right)^{1/3} \frac{h}{\mu} \quad (9)$$

where α is the ratio of the electrolysis current to the limiting current, D the coefficient of diffusion of the Cu^{2+} ion, F Faraday's constant, c electrolyte concentration, μ viscosity index, ρ density, σ the true electric conductivity, and k_c^* and k_e^* take the following values

$$k_c^* = \begin{cases} k_c - k_s q & \text{for } q/j > \phi \\ k_c + k_s q & \text{for } q/j < \phi \\ k_c & \text{for } q/j = \phi \end{cases} \quad (10)$$

$$k_e^* = \begin{cases} k_e + k_s & \text{for } q/j > \phi \\ k_e - k_s & \text{for } q/j < \phi \\ k_e & \text{for } q/j = \phi \end{cases} \quad (11)$$

It is easy to show that in the case where there is no need to mix the electrolyte by causing flow, and if the polarization of the electrodes is negligible in comparison to the voltage drop in the solution, the optimum current density is expressed by the

formula

$$j_{\text{opt, ideal}} = (k_c \sigma / k_e h)^{1/2}. \quad (12)$$

If it is necessary to mix the electrolyte, using the definition of the Sherwood number

$$(Sh) = \frac{j_{\text{lim}} h}{F D c} \quad (13)$$

and Equations 7 and 11, and assuming $j = \sigma \phi / h$ the following expression for the cost coefficient is obtained

$$k/k_{\text{opt, ideal}} = \frac{1}{2} \left[\frac{(Sh)}{(Sh)^*} + \frac{(Sh)^*}{(Sh)} + \frac{f^3}{f^{*3}} \frac{(Sh)^*}{(Sh)} \right] \quad (14)$$

where

$$k_{\text{opt, ideal}} = 2 \left(\frac{k_e k_c h}{\sigma} \right)^{1/2} \quad (15)$$

$$f = \lambda^{1/3} (Re) \quad (16)$$

$$\lambda = \frac{\Delta p}{l \rho V^2 / 2h} \quad (17)$$

and V and (Re) represent the mean flow rate and the Reynolds number. $k_{\text{opt, ideal}}$ represents the minimal cost of obtaining a copper mass unit in the case where costs of pumping and heating of the electrolyte as well as affects resulting from electrode overvoltage are of no importance. The coefficient k corresponds to the electrolysis under a current density minimizing unit costs of copper production taking into account the necessity of pumping and heating the electrolyte. The $k/k_{\text{opt, ideal}}$ ratio is therefore a measure of the deviation of the system from the conditions of electrolysis, among other things, without needing to mix and heat the electrolyte.

Assuming that the Sherwood number (Sh) may be expressed by the formula

$$(Sh) = \beta f^n \quad (18)$$

where β and n are the dimensionless parameters, whose values depend on the system's geometry only, the following expression for the optimum value of the Sherwood number $(Sh)_{\text{opt}}$ is obtained

$$\left[\frac{(Sh)_{\text{opt}}}{(Sh)^*} \right]^2 = 1 - \left(\frac{3}{n} - 1 \right) \left(\frac{1}{\theta} \frac{(Sh)_{\text{opt}}}{(Sh)^*} \right)^{3/n} \quad (19)$$

where θ is a dimensionless hydrodynamic parameter

$$\theta \equiv \frac{\beta f^{*n}}{(Sh)^*} = \frac{(Sh)(f^*)}{(Sh)^*}. \quad (20)$$

The optimum electrolysis conditions determined by $(Sh)_{opt}$ correspond to the cost coefficient k_{opt} expressed by the following formula

$$\frac{k_{opt}}{k_{opt, ideal}} = \frac{1}{2} \left\{ \frac{3 - 2n}{3 - n} \left[\frac{(Sh)_{opt}}{(Sh)^*} \right] + \frac{3}{3 - n} \left[\frac{(Sh)^*}{(Sh)_{opt}} \right] \right\}. \quad (21)$$

In [7] the features of the model described with Equation 7 have been discussed. The need of heating or cooling the electrolyte results in the break on the curve k versus j , at the place corresponding to $j = j_{LQ} = (q\sigma/h)^{1/2}$. Fig. 1 presents typical k versus j curves. At the current density j_{LQ} there is no need either to heat or to cool the electrolyte. The minimum of the curve represents the economic optimum of current density.

4. Numerical example

Assuming, according to Ibl and Adam [3], the cost of building one bath with 35 electrodes each one having an area of 1 square meter is \$8150 and

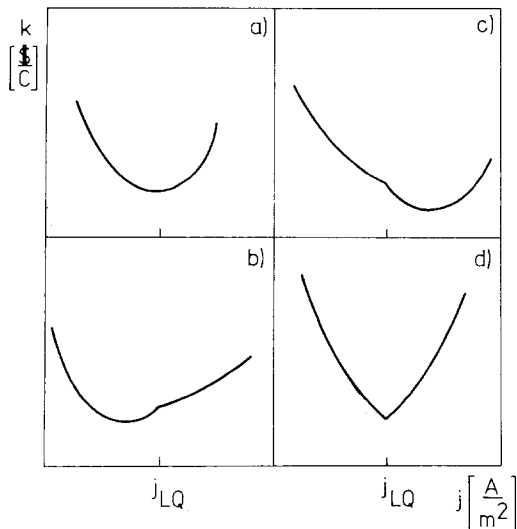


Fig. 1. Typical curves of process costs coefficient k versus current density j : (a) hypothetical case of zero costs of cooling and heating media, (b) case of optimum current density j_{opt} below j_{LQ} (electrolyte to be heated), (c) case of optimum current density j_{opt} above j_{LQ} (electrolyte to be cooled), (d) optimum current density j_{opt} equal to j_{LQ} .

assuming a linear interest for a period of 15 years and 10% interest rate, the following value of the investment cost coefficient given in square meters of the active electrode area per hour is obtained

$$8150 \times \frac{100/15 + 10}{100} : 70 \text{ m}^2 : 8760 \text{ h} \\ = 0.00222 \text{ \$ m}^{-2} \text{ h}^{-1}.$$

B. Ruehl [8] indicates that the investment costs make up an average of 28%, cost of the wages 18% and the repair and auxiliary means costs 14% of total costs. Taking this into account the coefficient of wage costs is equal to $0.00147 \text{ \$ m}^{-2} \text{ h}^{-1}$ and the coefficient of the repair costs is equal to $0.00111 \text{ \$ m}^{-2} \text{ h}^{-1}$.

Let us assume further that the amount of metal in the processing is equal to 160 kg Cu m^{-2} of the active electrode area and that the gold and silver contents are equal to 30g and 3 kg per ton of copper. The interest on this value equals 10%. However, this value is not subject to amortization, because it does not decrease (neglecting the present relatively high changes of prices). At prices of \$1350 per ton of copper and \$139 and \$3700 per kilogram of silver and gold the interest rate on the metal under processing is equal to $0.00343 \text{ \$ m}^{-2} \text{ h}^{-1}$. Therefore the capital costs coefficient is

$$k_c = 0.00222 + 0.00111 + 0.00147 \\ + 0.00343 = 0.00823 \text{ \$ m}^{-2} \text{ h}^{-1}.$$

As has already been mentioned, the value of the coefficient k_c estimated in this way may be treated as independent of the current density used and the area of electrodes only at the first approximation.

* In fact, two factors may be distinguished in wage costs:

(a) wage costs of personnel employed in the exchange of cathodes and anodes, dependent on the amount of copper being produced, and

(b) wage costs of inspectors supervising the process, proportional to the number of the cells (areas of electrodes) under supervision.

With the fixed output of the refinery, an increase in current density corresponds to a decrease in the area of electrodes and thus to the decrease of element (b). In conversion to unit area of electrode, the cost coefficient (a) grows while the cost coefficient (b) remains unchanged. Since it is known from industrial practice that wage costs make up usually 20% of total costs, it is more advisable, in our opinion, to consider them even in an approximate way rather than to ignore them as done by the majority of authors. The above applies similarly in regard to repair costs.

It should be noted that a considerable increase in current density must be associated with substantial changes in the construction of the electrolyser and in the technology of the process, which will cause an increase in investment costs borne, but only once, and a rise in repair costs. On the other hand, they will significantly reduce labour consumption [9], the related expenditure and the interest accumulated by the 'frozen' metal converted to unit area and unit time. These opposing factors justify the adoption of the assumption of independence of capital costs from the current density used.

Let us assume further that the electrical energy cost including the cost of rectifying and associated losses equals $0.01 \text{ \$ (kWh)}^{-1}$, or $k_e = 2.78 \times 10^{-9} \text{ \$ J}^{-1}$. One ton of water steam delivers about 0.6 Gcal , which at the price of $2.50 \text{ \$ ton}^{-1}$ makes the unit cost $1 \times 10^{-9} \text{ \$ J}^{-1}$. If current density exceeds the value of about 600 A m^{-2} further heating becomes unnecessary and a need arises to remove the surplus heat. Assuming that a ton of cooling water removes approximately 25 Mcal at a price of $2.50 \text{ \$ (100 m}^3)^{-1}$ [10], the cost coefficient of removing from the system 1 J of energy is $2.4 \times 10^{-10} \text{ \$ J}^{-1}$.

The specific costs of pumping k_p per 1 kW of power consist of the share of r_E related to the energy consumption and of r_I related to the investment. Assuming a time unit of 1 hour , then $r_E = k_e$ (cost of 1 kWh). The cost of the pumping equipment for the electrolysis plant is generally only estimated as it depends on the construction. Assuming, according to Ibl and Adams [3], that in the first approximation the cost is independent of the pumping power at the given cell cross-section, and that it is proportional to the cross-section (as well as to the area of electrodes) and equal to $40 \text{ \$ m}^{-2}$ of the electrode and assuming 10% of amortization, the following value of the capital cost coefficient k_c is obtained

$$\begin{aligned} k_c &= 0.00823 + \frac{40}{8760 \times 10} \\ &= 0.00869 \text{ (\$ m}^{-2} \text{ h}^{-1}). \end{aligned}$$

On the other hand in the investment costs, only the expenses associated with the power of motors used is equal to $20 \text{ \$ kW}^{-1}$ and 10% of amortization rate, are taken into account. Then

$$r_E = k_e = 2.78 \times 10^{-9} \text{ \$ J}^{-1} \approx 2.8 \times 10^{-9} \text{ \$ J}^{-1}$$

$$r_I = 6.3 \times 10^{-11} \text{ \$ J}^{-1}$$

$$k_p = r_E + r_I \approx 2.8 \times 10^{-9} \text{ \$ J}^{-1}.$$

In order to estimate the thermal losses of the electrolyser the thermal balance for stabilized conditions has been calculated assuming the electrolyte temperature to be 60° C and the typical technical parameters and thermal conductivities of the materials. The value of the parameter Q_d/A estimated in this way is equal to $200 \text{ J m}^{-2} \text{ s}^{-1}$ and is in accordance with the value calculated for the data contained in [6]. By way of an example, the value of the parameter Q_d/A calculated from the technological data of the Legnica copper works (Poland) amounts to $110 \text{ J m}^{-2} \text{ s}^{-1}$.

For the calculations the concentration of the copper ions in 1 M solution, the ratio of the electrolysis current to the limiting current of copper liberation $\alpha = 0.37$ and diffusion coefficient $D = 1 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$ have been assumed.

5. Analysis of the numerical example

The nonlinear equation (Equation 19) has been solved with the numerical calculations for $(Sh)_{\text{opt}}$, assuming different values of electrolysis parameters. The optimum value of the electrolysis cost coefficient k_{opt} has also been calculated against this background. The calculations were made assuming the Sherwood number (Sh) to be a function of the coefficient f described by Equation 18, and the values of coefficients n and β have been taken from [7]. Two cases have been considered.

(a) the turbulent flow through the empty channel, when

$$\beta = 0.040(Sc)^{1/4} \quad \text{and} \quad n = 21/22,$$

(b) the flow through the channel containing the eddy promoters (perturbing inserts), placed at a distance of $\Delta l = 10 \text{ cm}$ from each other. The measurement of the mass transport in this type of channel has been made by Sonin and Isaacson [7]. The coefficients β and n are

$$\beta = 1 \times (Sc)^{1/3} (h/\Delta l)^{0.38} \quad \text{and} \quad n = 0.6,$$

where (Sc) is the Schmidt number, defined as

Table 1. Optimum current density j_{opt} and cost coefficient k_{opt} values for different interelectrode distances h with and without eddy promoters: [$\sigma = 100 \Omega^{-1} m^{-1}$, $Q_d/A = 200 J m^{-2} s^{-1}$, $k_c/k_e = 800 \$ m^{-2} s^{-1} (\$ J^{-1})^{-1}$]

h (cm)	j_{opt} (A m ⁻²)		$k_{opt} \times 10^9$ ($\$ C^{-1}$)	
	Empty channel	Channel with eddy promoters	Empty channel	Channel with eddy promoters
1	1930	2690	2.29	1.50
2.5	1480	1700	3.20	2.38
5	1130	1200	4.34	3.36
7.5	948	981	5.25	4.12
10	830	849	6.03	4.76

$$(Sc) = \frac{\mu}{\rho D}$$

In Table 1 the values j_{opt} and k_{opt} for different electrode distances have been given for both of the above cases. The figures contained in the table show that usage of eddy promoters decreases the electrorefining costs by about 1/3. Therefore further analysis has been carried out for an electrolyser equipped with flow perturbbers.

Figs. 2 and 3 present the optimum current density j_{opt} versus the electrode distance h for different values of σ and for various constant cost coefficients k_c/k_e ratios. The influence of the interelectrode distance on the optimum current density value is evident, especially in the case of high values of the k_c/k_e ratio. It is interesting that

the optimum current density value equals the current densities used in industrial practice only for the lowest values of the k_c/k_e ratios. It has to be noted that the value of the k_c/k_e ratio decreases if the refinery construction costs (capital costs) decrease, or if the unit cost of electrical energy increases. On the other hand the change of the solution conductivity (parameter σ) influences the j_{opt} insignificantly.

Figs. 4–6 present the electrolysis cost coefficient k_{opt} ($\$ C^{-1}$) versus the electrode distance h for different values of other parameters. As previously, the influence of the capital costs on the unit electrical energy cost (k_c/k_e) ratio is most significant and k_{opt} is practically independent of the thermal loss coefficient Q_d/A . Figs. 7 and 8

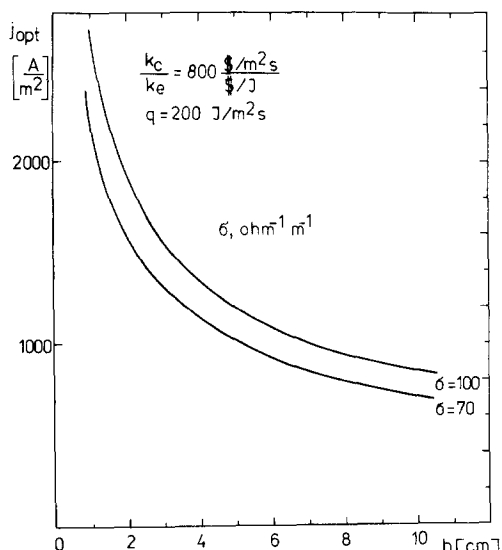


Fig. 2. Optimum current density j_{opt} versus the electrode distance for different electric conductivity values σ .

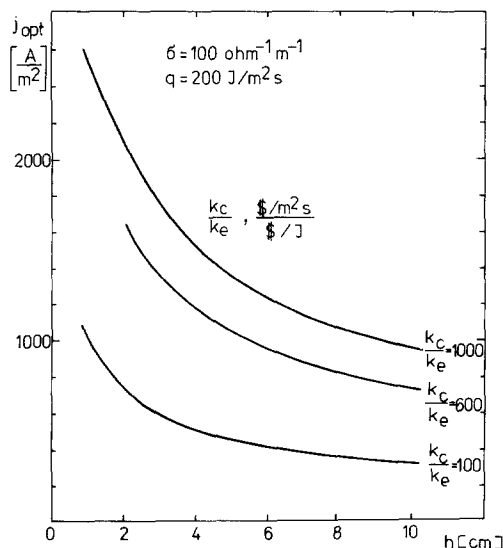


Fig. 3. Optimum current density j_{opt} versus h for different values of capital costs to electrical energy costs k_c/k_e ratio.

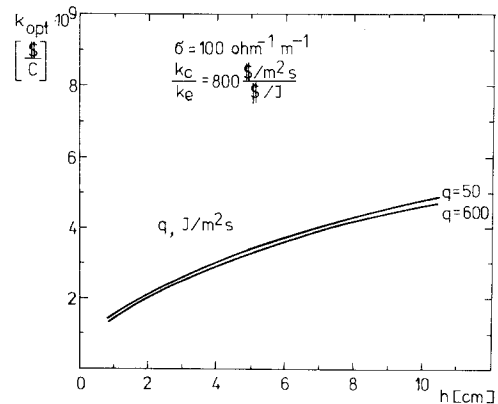
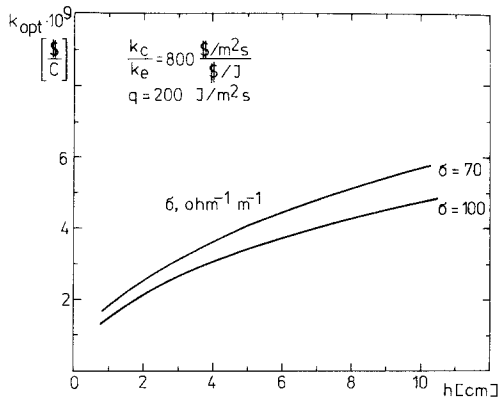


Fig. 4. Optimum coefficient of unit mass production costs k_{opt} ($\$ C^{-1}$) versus interelectrode distance h for different electric conductivity, σ , values.

Fig. 6. k_{opt} versus h for different values of the heat loss coefficient $Q_d/A = q$.

represent the curves k_{opt} and j_{opt} versus the cost coefficients k_c/k_e ratio for different interelectrode distances.

Fig. 7 shows that the value of the cost coefficient k_{opt} strongly decreases with the decrease of the interelectrode distance. Simultaneously, the small distance h corresponds to the high values of optimum electrolysis current density j_{opt} . Therefore, there are economic reasons for a tendency to decrease the interelectrode distances and to operate at relatively high current densities. The decrease of the interelectrode distances required technological changes, with higher precision of the electrolyser. Obviously it results in an increase of the capital costs k_c . However, using Fig. 7 the electrolysis parameters could be chosen so as not to increase the cost of electrorefining of the

copper mass unit. For example, let the interelectrode distance $h = 10$ cm, the conductivity of the solution $\sigma = 100 \text{ohm}^{-1} \text{m}^{-1}$, the electrolyser's heat loss coefficient $Q_d/A = 200 \text{J m}^{-2} \text{s}^{-1}$ and the ratio $k_c/k_e = 100 (\text{\$/m}^2 \text{s}^{-1})/(\text{\$/J})$. These assumptions correspond to a value of the production cost coefficient of $k_{opt} = 1.77 \times 10^{-9} \text{\$/C}$ and the optimum current density $j_{opt} = 316 \text{A m}^{-2}$. Let the technology change associated with the process intensification result in a three-fold increase of the value of the k_c/k_e ratio; nevertheless, if the mean interelectrode distance is kept at the level of 2.5 cm (which corresponds to $k_{opt} = 1.43 \times 10^{-9} \text{\$/C}$ and $j_{opt} = 1020 \text{A m}^{-2}$) the costs of electrorefining may be decreased. At the same time the output of the refinery under discussion will increase.

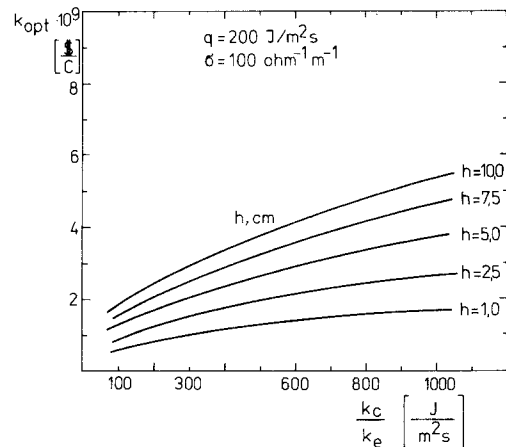
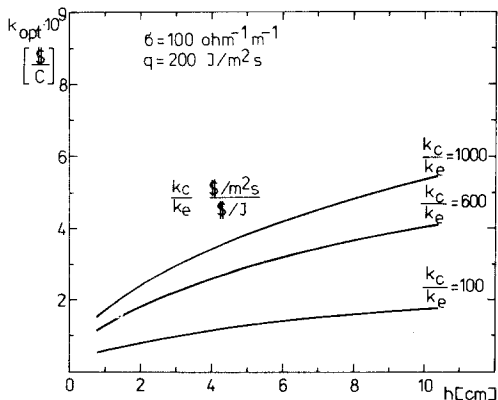


Fig. 5. k_{opt} versus h for different values of the ratio k_c/k_e .

Fig. 7. k_{opt} versus cost coefficient ratio k_c/k_e for different interelectrode distances h .

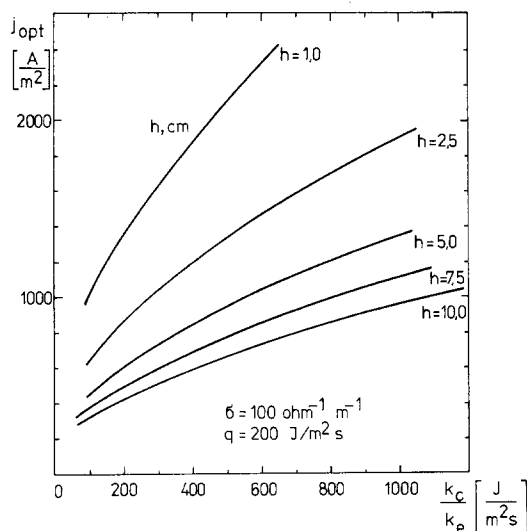


Fig. 8. Optimum current density j_{opt} versus costs coefficient ratio k_c/k_e for different electrode distances h .

It follows from the data shown in Table 1 and Figs. 7 and 8 that for the technological parameters used in commercial practice, the optimum current density is confined within the 800–900 A m^{-2} range.

To obtain this current density, an increase of the electrolyte circulation rate is necessary in order to prevent anode passivation and increase of noble metal losses and a deterioration of the quality of the cathode deposit. The cost of pumping equipment is a relatively small percentage of the capital costs, and therefore this technology change and the costs related to it seem to be economic. Similar conclusions concerning optimal current density and a contribution of pumping costs to total process costs were reached by Ibl [11, 12] in a different way.

6. Conclusions and recapitulation

The paper presents the method of determination of the copper electrorefining current density and of the mass unit production cost k_{opt} , using the simplified model of the electrolyser. All the costs have been presented in the form of the unit coefficients in order to achieve a more general analysis,

and therefore the coefficients obtained do not depend on the scale of the plant.

It has been assumed that the total production cost depends on the following factors:

(a) Capital costs represented by the capital costs coefficient k_c ($\text{\$ m}^{-2} \text{ s}^{-1}$), which to a first approximation does not depend on the electrolysis current density.

(b) Electrical energy cost expressed by the coefficient k_e ($\text{\$ J}^{-1}$).

(c) Costs of heating by steam or cooling by water expressed by the coefficient k_s ($\text{\$ J}^{-1}$).

(d) Cost of pumping the electrolyte represented by the coefficient k_p ($\text{\$ J}^{-1}$).

A nonlinear equation has been derived relating the above coefficients to the optimum current density and to the unit production cost. This equation has been solved assuming broad ranges of variability of the above parameters.

The results of the analysis of the electrorefining cost of the copper mass unit show that for the present technological parameters, the optimum current density, from the economical point of view, should range from 800–900 A m^{-2} . To obtain this value some changes in present technology are necessary. The discussion shows that these changes may result in even a decrease in production cost and the simultaneous intensification of the process.

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