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Current Efficiency of Low Temperature Aluminum Electrolysis Studied by Neural Network

Huimin Lu[1], *Zuxian Qiu*[2], *Keming Fang*[1], *Fuming Wang*[1], *Yanruo Hong*[1]

1) Metallurgy School, University of Science and Technology Beijing, Beijing 100083, China
2) Department of Nonferrous Metallurgy, Northeastern University, Shenyang 110006, China
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Abstract: A prediction model for Current Efficiency (CE) of low temperature aluminum electrolysis (LTAE) with the low molar ratio electrolyte of Na_3AlF_6-AlF_3-CaF_2-MgF_2-LiF-Al_2O_3 system was investigated based on artificial neural network principles. The nonlinear mapping between CE of LATE and various electrolytic conditions was obtained from a number of experimental data and used to predict CE of LATE. The trained neural networks possessed high precision and resulted in a good predicting effect. As a result, artificial neural networks as a new cooperating and predicting technology provide a new approach to the further studies on low temperature aluminum electrolysis.

Key words: low temperature aluminum electrolysis; current efficiency; neural network; prediction model; low molar ratio electrolyte

Low Temperature Aluminum Electrolysis (LATE) is generally accepted to be a development tendency of aluminum production due to its potential of high Current Efficiency (CE), low energy consumption, long cell life and easier adoption of inert materials for electrodes and cell lining. Because CE is an important economic and technical index and high CE is also a marking of electrolytic technological parameter optimizing, it is very important to predict CE for conducting investigation on LATE in a thorough-going way.

Researches have shown that CE is affected by various electrolytic technological parameters, including bath composition, electrolytic temperature, cathodic current density, inter-polar distance and cell structure [1]. However, the relationships between CE and electrolytic technological parameters are very complex and nonlinear, therefore it is very difficult to predict CE by traditional mathematical statistical and regressive methods.

Artificial neural networks (ANN) is a new information processing system based on modeling the neural system of human brain. It has some remarkable properties such as self-learning and adaptation, which make it suitable for nonlinear problems with complex factors [2]. This paper aims at finding a practical method to predict CE by using ANN to correlate CE with various technological parameters of low temperature aluminum electrolysis.

1 Experimental

The low molar ratio electrolyte from the Na_3AlF_6-AlF_3-CaF_2-MgF_2-LiF-Al_2O_3 system was used. The anode of the laboratory cell was equipped with a central, vertical hole and two horizontal holes, while the underside had the shape of a cone. Research works indicated that such anode with holes could indeed improve the circulation of the electrolyte in the cell [3]. The electrolysis cell was contained in a closed furnace with argon atmosphere protection, the CE was determined from the gain of aluminum after carrying out 1h experiment, and the current was monitored by HA-1 type ampere-hour meter. **Tables 1–3** list the electrolytic constituent,

Table 1 Constituent variables of low ratio electrolyte

Parameter	Symbol	Value
Cryolite ratio(CR)	Z_1	1.5, 1.8, 2.0, 2.1, 2.2, 2.3, 2.4
$w(CaF_2)$ / %	Z_2	3, 4, 5, 6, 7
$w(MgF_2)$ / %	Z_3	0, 1, 2, 3, 4
w(LiF) / %	Z_4	0.5, 1, 2, 3

Table 2 Main operating variables

Parameter	Symbol	Value
Temperature t / ℃	Z_5	800, 850, 900, 920, 930,940, 950
Cathodic current density d_c / A·cm^{-2}	Z_6	0.50, 0.75, 0.85, 1.00
Inter-polar distance h / cm	Z_7	0.5, 1, 2, 3, 4,

Table 3 Electrolysis cell structure and experimental data

Inner diameter of the cell lining / cm	6.5
Inner depth of the cell / cm	8.0
Diameter of the anode / cm	2.6
Diameter of holes in anode / cm	0.8
Anode underside area / cm^2	4.8
$w(Al_2O_3)$ / %	3.0
Amount of electrolyte in a run / g	350.0
Electrolytic time / h	1.0

main operating variables and cell structure data respectively.

2 ANN Modeling of CE Prediction

So far, there are ANN models no less than 40 [4], but only the Bake Propagate (BP) network is the most useful neural network in the present day. The BP network is a feedforward type of overall linking multilayer ANN, which has stronger associative memory and popularizing abilities and may approach on any nonlinear continuous function in arbitrary accuracy.

As shown in **figure 1**, the ANN used in this study is a tree layer BP network system with $7 \times 10 \times 1$ nodes, in which $Z_i (i = 1, 2, \cdots, 7)$ represents the input layer nodes, in the order of the Cryolite Ratio (CR), $w(CaF_2)$, $w(MgF_2)$, $w(LiF)$, temperature (T), cathodic current density (d_c), inter-polar distance (h); O(CE) represents the output layer node and $Y_j (j = 1, 2, \cdots, 10)$ the hidden layer nodes; V_{ji} and W_j represent the connecting weights between the input layer and the hidden layer, the hidden layer and the output layer respectively.

Carrying out the algorithm procedure of ANN may be described as follows:

(a) Take the learning rate $\eta = 0.5$, the momentum coefficient $\alpha = 0.8$.

(b) Set V_{ji} and W_j with the random numeral value between -0.5 and 0.5.

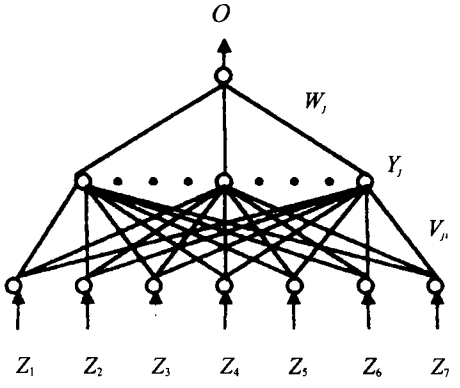

Figure 1 BP network structure of ANN.

(c) Calculate each neuron output successively from the input layer to the output layer:

$$\text{net}_j = \sum_{i=1}^{7} V_{ji} Z_i - \theta_j, \quad (j = 1, 2, \cdots, 10) \tag{1}$$

$$Y_j = f(\text{net}_j) \tag{2}$$

$$\text{net} = \sum_{j=1}^{10} W_j Y_j - \gamma \tag{3}$$

$$O = f(\text{net}) \tag{4}$$

$$f(x) = \frac{1 - e^{-x}}{1 + e^{-x}} \tag{5}$$

where, net is a summed function used to combine weighed incoming information; $f(x)$ is a transfer function which takes the form of sigmoid function; θ and γ are the threshold values of $f(x)$ to the hidden layer and the output layer respectively.

(d) Calculate system errors. Assume there are P learning samples, the mean squared system error is determined by

$$E = \frac{1}{2P} \sum_{n=1}^{P} (D_n - O_n)^2 \tag{6}$$

where, D_n represents the output of the nth learning sample, usually known as tutorial signal, O_n shows the corresponding actual network output.

(e) If E meets the error accuracy requirement or reaches the given learning times, then the learning is over.

(f) Calculate the learning signal δ of each neuron on successive layer.

$$\delta_0 = (D - O) f'(\text{net}) \tag{7}$$

$$\delta_j = W_j \delta_0 f'(\text{net}_j) \tag{8}$$

(g) Accordingly to the δ learning rule, the connecting weights on the $t+1$ iteration will be modified by

$$W_j(\tau + 1) = W_j(\tau) + \eta \delta_0 Y_j + \alpha [W(\tau) - W_j(\tau - 1)] \tag{9}$$

$$V_{ji}(\tau + 1) = V_{ji}(\tau) + \eta \delta_1 Z_j + \alpha [V_{ji}(\tau) - V_{ji}(\tau - 1)] \tag{10}$$

(h) Return to (c), repeating the network calculation until the network learning end.

Total 23 sets of experimental samples were investigated in this study, but only 18 sets, known as the learning sample selected randomly from the experimental samples, were used to train the neural networks. The rest 5 sets, known as the testing sample, being treated as "unknowns", were put aside temporarily for the later examination of predictive ability of the ANN modeling. After trained for 6000 times, the summed square residual errors were satisfied. **Figure 2** shows the comparison of the network-predicted CE with the measured CE.

From figure 2, the ANN actual output O and desired

output D approached considerably, the maximum relative error was 2.2% and most of the relative error (ε) were less than 1%, which showed that the ANN had founded a correct model and it had high accuracy. The linear correlation coefficient between the measured and the predicted CE was 95.8%. Good agreement from the test samples was also obtained. Therefore, the trained neural networks could be used to predict CE with a new input (beyond training) according to the knowledge it acquired during training.

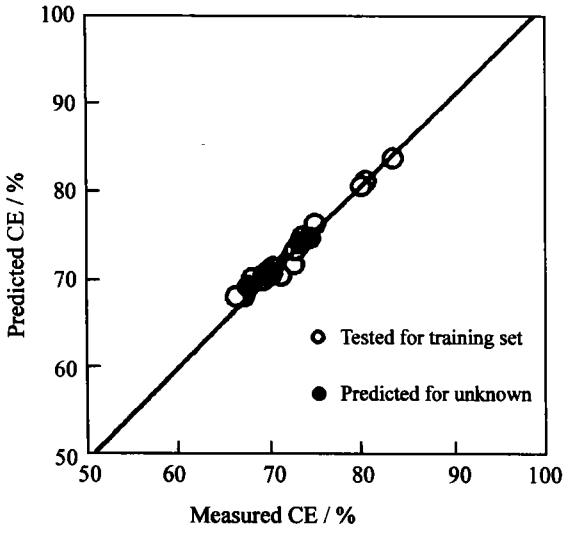

Figure 2 Results of ANN training and prediction.

3 Predicting Results and Discussion

The resulted ANN model may be used to predict CE, *i.e.*, simulating the whole process of LTAE, thereby realizing the comprehensive prediction of CE. The effective relationships of the various electrolytic parameters with the CE are given in **figure 3**.

Figure 3 (a) shows the CE as a function of the NaF/AlF_3 cryolite ratio (CR) at 20℃ above liquidus and constant 950℃. The CE roughly follows the curves 1 and 2 with the slope $\frac{dCE}{dCR} = -16.7\%$ and -5.2% per unit ratio respectively. The reason that these curves have different slopes is that curve 1 expresses the com-

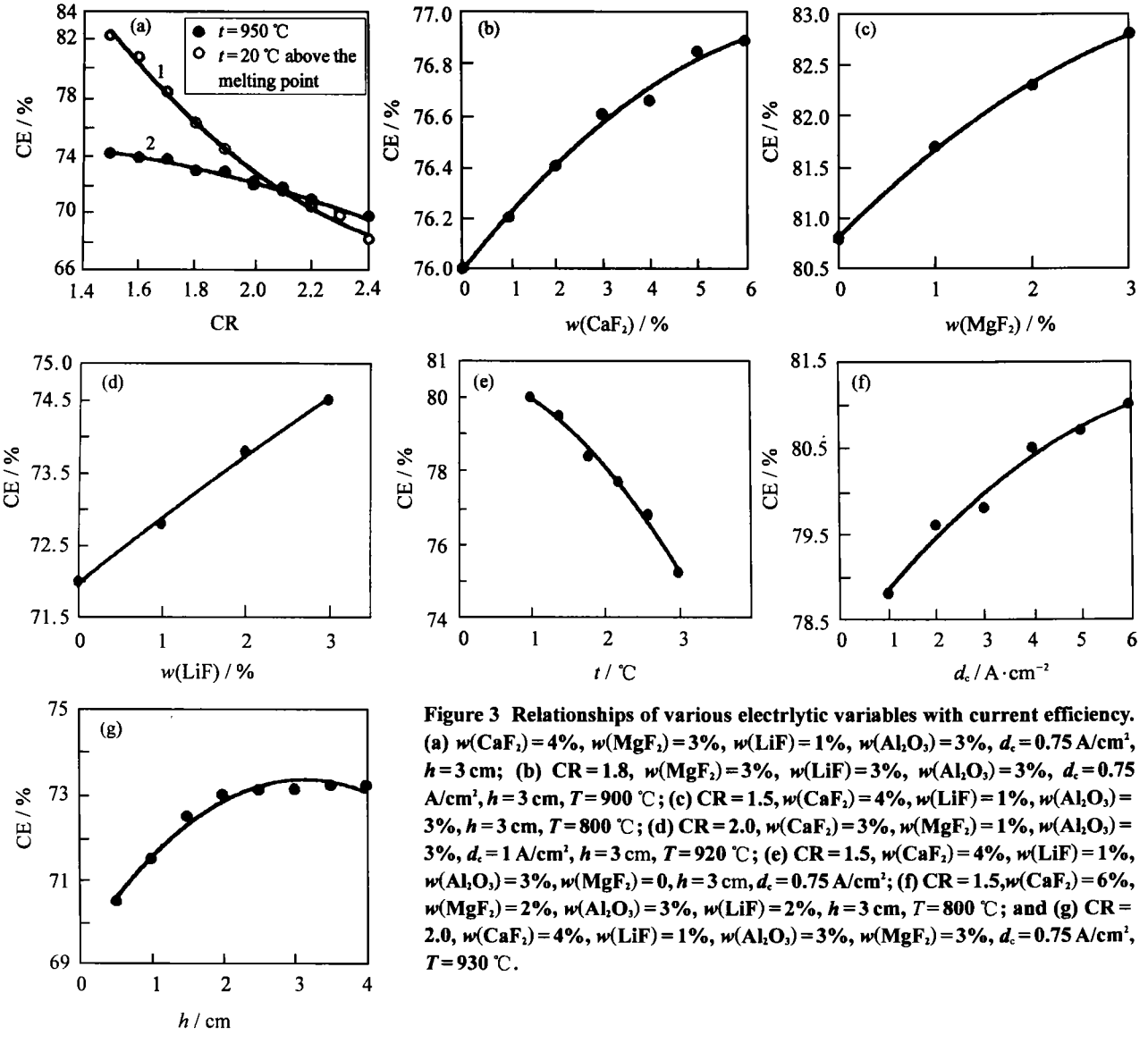

Figure 3 Relationships of various electrlytic variables with current efficiency. (a) $w(CaF_2)=4\%$, $w(MgF_2)=3\%$, $w(LiF)=1\%$, $w(Al_2O_3)=3\%$, $d_c=0.75$ A/cm², $h=3$ cm; (b) CR = 1.8, $w(MgF_2)=3\%$, $w(LiF)=3\%$, $w(Al_2O_3)=3\%$, $d_c=0.75$ A/cm², $h=3$ cm, $T=900$ ℃; (c) CR = 1.5, $w(CaF_2)=4\%$, $w(LiF)=1\%$, $w(Al_2O_3)=3\%$, $h=3$ cm, $T=800$ ℃; (d) CR = 2.0, $w(CaF_2)=3\%$, $w(MgF_2)=1\%$, $w(Al_2O_3)=3\%$, $d_c=1$ A/cm², $h=3$ cm, $T=920$ ℃; (e) CR = 1.5, $w(CaF_2)=4\%$, $w(LiF)=1\%$, $w(Al_2O_3)=3\%$, $w(MgF_2)=0$, $h=3$ cm, $d_c=0.75$ A/cm²; (f) CR = 1.5, $w(CaF_2)=6\%$, $w(MgF_2)=2\%$, $w(Al_2O_3)=3\%$, $w(LiF)=2\%$, $h=3$ cm, $T=800$ ℃; and (g) CR = 2.0, $w(CaF_2)=4\%$, $w(LiF)=1\%$, $w(Al_2O_3)=3\%$, $w(MgF_2)=3\%$, $d_c=0.75$ A/cm², $T=930$ ℃.

prehensive effect of CR and temperature on CE but the curve 2 only shows that of CR on CE. The liquidus of predicting points on the curve 1 is determined only by the CR. Therefore, The temperature (t) of predicting points on the curve 1 increases with the CR increasing. The effect of cryolite ratio on CE reflects the rate change of cathodic side reactions, mainly due to the significant increase of equilibrium sodium concentration on the cathode aluminum, with the NaF concentration in the electrolyte increasing. Most of the experimental work previously carried out on CE as a function of NaF/AlF_3 cryolite ratio shows, in agreement with this present work, a substantial increase of CE with the AlF_3 concentration in electrolyte increasing [5–7]. The predicting results also show that the trained ANN model can predict not only the effect of single variable on CE, but also the effect of multivariable on CE.

Figure 3(b–d) shows that CEs increase as the CaF_2, MgF_2 and LiF concentration increase and the slope $\frac{dCE}{dw(CaF_2)}$, $\frac{dCE}{dw(MgF_2)}$ and $\frac{dCE}{dw(LiF)}$ of these curves are 0.15, 0.7 and 0.81 respectively. As shown in figure 3(e), the CE decreases as the electrolytic temperature rises, and the slope $\frac{dCE}{dt} = -0.052\%/℃$. It is generally acknowledged that the CE is chiefly related to the loss of aluminum produced in the electrolysis and the loss of aluminum increases with the electrolytic temperature increasing. Additives CaF_2, MgF_2 and LiF can all improve the physico-chemical properties of molten electrolyte in aluminum electrolysis, such as lowering liquidus and decreasing aluminum solubility. In addition, the favorable effect of LiF on CE is mainly caused by the increasing of electrical conductivity and thereby the transport of current through the melt. The influence of temperature on CE is due to the effect of temperature on the equilibrium concentration of sodium, and thus the effect on the rate of cathodic side reactions increasing electronic conduction.

Figure 3(f) shows the ANN prediction of the CE as a function of the cathodic current density. It can is seen that the CE increases as the cathodic current density increases and the slope $\frac{dCE}{dc} = 4\%/(A \cdot cm^{-2})$. The effect of cathodic current density on CE is due to the metal aluminum production on unit area of cathode surface increasing when the current density rising. When the metal loss from the unit area of cathode surface keeps constant, the CE must be increased.

The interpolar distance was varied between 0.6 and 4.0cm. The predicted results given in figure 3 (g) show that the CE is constant for the interpolar distance greater than 2.0 cm. The influence of interpolar distance on CE can be explained from the view of aluminum dissolution in electrolyte. During the electrolysis, the dissolved aluminum was transferred to the anode region by diffusion, vection and bath circulation. An increase in the interpolar distance means an elongation of the path of transference, a decrease in the concentration gradient of the dissolved aluminum, and a weakening of the bath circulation in the interpolar space, so that the reoxidation reaction can be lessened. In contrary, at the very low interpolar distance the rate of recombination will be greatly enhanced.

4 Conclusions

CE was correlated with electrolytic technological parameters by means of back propagation neural networks based on the low temperature aluminum electrolysis experiments with low cryolite ratio electrolyte of Na_3AlF_6-AlF_3-CaF_2-MgF_2-LiF-Al_2O_3 system. The trained neural networks possessed high precision and good predicting effect. It is concluded that artificial neural networks as a new cooperating and predicting technology provide some new ways and means for the further studies on low temperature aluminum electrolysis.

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