CFD Models in the Development of Electrical Waste Recycling Technologies

Attila Egedy*,[a], Szabolcs Fogarasi[b], Tamás Varga[a], Árpád Imre-Lucaci[b],
Tibor Chován[a]

[a]Department of Process Engineering, University of Pannonia, Egyetem Str. 10, H-8200 Veszprém, Hungary
[b]Faculty of Chemistry and Chemical Engineering, Babes-Bolyai University, Arany János Str. 11, RO-400028, Cluj
Napoca, Romania
egedya@fmt.uni-pannon.hu

Nowadays electrical waste (EW) recycling became a practical way to provide raw material for new devices. Computer parts such as memory, motherboard or other part contains large amount of metals of which the recovery of precious metals and copper represents the highest economical potential. With a proper chemical treatment these metals can be efficiently extracted and separated from the actual waste. For this task a specially designed leaching reactor, equipped with a perforated rotating drum, was used.

This work is aimed at investigating if Computational Fluid Dynamics CFD tools can be efficiently applied to model the chemical reactor used to dissolve the metals from the EW. A hybrid CFD compartmental approach was developed to facilitate the chemical reactions in the leaching reactor. The CFD models were used to model the hydrodynamics, and a compartmental model based on the CFD model was used for modelling the reactions. Based on the velocity field the chemical reactions can be implemented, and the developed simulator can be used to enhance the performance of the existing reactor system. For the modelling studies COMSOL Multiphysics was be used as CFD software. Optimized construction and operating parameters of the leaching reactor, allow us to reduce the energy consumption and the environmental impact.

1. Introduction

Due to the constant development of computer technologies tons of electrical waste (EW) are produced. The EW contains different valuable metals for example: Au, Ag, Pt, Cu, Zn which can be leached with different methods (Yang et al., 2012).

Selective leaching is not an option in this case because the reagent leaches all the metals in a redox reaction. Traditionally different solvents and complex-forming compounds can be used for example (Fogarasi et. al 2012).

- H_2SO_4
- $(NH_4)_2SO_4$
- $(NH_4)_2S_2O_8$
- $FeCl_3$
- $CuCl_2$
- CN- salts (Au)

Since all the metals leached out, their order of leaching can be determined only based on the reactivity series of the metals. After the leaching part of the process was completed, the dissolved metals must be separated from the solution using an electro-chemical reactor. For this step of the process traditionally rotating electrode systems can be used (Van Parys et. al., 2008). With these reactors copper recovery was also modelled (Rivero et. al 2010).

In the leaching phase the proper phase contact must be crucial next to the reagent type and reagent concentration. The proper homogeneity of phases can be achieved easily in a laboratory scale device

using magnetic stirrers. However in larger scales, these methods cannot be used, so an impeller must be used to mix the fluid inside the reactor. The reactions can be faster if the reagent circulates in the reactor too, therefore a continuous reactor configuration is a good choice in this case. A new reactor was developed based on an existing reactor, and hydrodynamic and kinetic models were implemented to optimize the operation.

For the hydrodynamic modeling a CFD approach was used as an excellent tool to analyze whole technologies, even multiphase systems (Tamburini et al., 2011). A full 3D CFD model was built for modelling the hydrodynamic and the developing flow field in the leaching reactor. The geometry of the reactor and the impeller are quite complex, still with a validated CFD model the velocity field can be computed within the reactor. Besides, several studies show that CFD simulators can be excellent tools for modelling electro-chemical processes (Martinez-Delgadillo et al. 2012,).

After the hydrodynamic model was solved the velocity values derived from the obtained results can be used to compute the parameters of the compartment model. These models are called hybrid CFD-compartmental models, and widely used in design, modelling and development. The main advantage of the hybrid approach is that the momentum balance equations can be computed using the CFD software, and the component balances and the optimization steps can be solved in a different software, more suitable for optimization problems solving. A compartmental model can be more suitable for facilitating full leaching kinetics, crystallization or polymerisation reactions (Maggioris et al. 2008).

The connections between the CFD and compartment models mean the determination of the circulation ratios. The circulation ratios can be calculated from the variables of the turbulent model (diffusivity, or energy dissipation rate), or in a simpler case by using the averaged velocity field values (Guha et al., 2006). A laminar flow based model was used without any turbulent parameters; hence the velocity values could be used to calculate the circulation ratios.

Beside the connections the other important factor in the field of compartment modelling is the compartment model structure. The model structure can be determined using structure identification, zoning applying CFD, or with the application of some heuristics (Delafosse et al. 2010). Heuristics and structure identifications were combined to indentify the most appropriate model structure for the developed reactor.

Copper leaching reactions were chosen for detailed investigations because this metal is one of the most frequent materials in EW. The primary goal of this study was to create a hybrid CFD-compartmental model for examine the behaviour of the reactor and after model validation to use this model for optimizing the leaching process. COMSOL Multiphysics was used for solving the hydrodynamic model and MATLAB for the compartment and kinetics modelling, and optimization.

2. Modeling steps and results

In this chapter the modelling steps of the leaching reactor are discussed. In the first step of the model building the kinetic parameters were identified and hydrodynamic models were implemented in the chosen CFD software. The parameters of the compartment model were determined based on measurements, and by using the indentified kinetic parameters and the compartment model we optimized the process.

2.1 Kinetic identification
The following two reactions are considered as the main reaction steps of the leaching process of copper via Fe^{3+} ions and the copper leaching with Cu^{2+} ions:

$$Cu + 2Fe^{3+} \rightarrow Cu^{2+} + 2Fe^{2+} \tag{1}$$

$$Cu + 2Cu^{2+} \rightarrow 2Cu^{+} \tag{2}$$

Both reactions were identified as first order reaction. Three measurements were taken at different temperatures to identify the reaction kinetic parameters of these reactions (20-40-60°C). $FeCl_3$ was used as a reagent with concentrations (0.2 M), with 0.5 M HCl. Arrhenius-equations were used to calculate the reaction rate constants of each reaction. The kinetic parameters we have to identify are the pre-exponential constant, and the activation energies, as Eq.(3) and Eq.(4) shows.

$$k_1 = k_{01} \cdot e^{\frac{-Ea_1}{RT}} \tag{3}$$

$$k_2 = k_{02} \cdot e^{\frac{-Ea_2}{RT}} \tag{4}$$

Consequently there are four parameters have to be identified. For the kinetic parameter identification step the MATLAB built in fmincon function was used, which based on sequential quadratic programming. Mass balance was calculated for the four different substances (Cu and Fe ions). The concentrations were measured with an AAS spectrometer from 1 mL samples, so the results are corrected with the amount of copper in the samples too. Table 1 shows the identified model parameters.

Table 1: Reaction kinetic parameters for the leaching of copper

Parameter	k_{01}	Ea_1	k_{02}	Ea_2
Value	492.82	25965	718.13	23082
Unit	1/min	J/mol	1/min	J/mol

2.2 The hydrodynamic model of the leaching reactor
The hydrodynamic model of the leaching reactor was implemented in COMSOL Multiphysics. There is no need to apply any of the turbulence models because the maximal Reynolds number in the system is less than 300. The modelled reactor is a continuous reactor with one inlet, one outlet, and a perforated rotating drum. Navier-Stokes equations were used to model the momentum balance. Since there is fluid mass inside the drum multiple fluid domains must be defined. Figure 1 shows a photograph of the reactor.
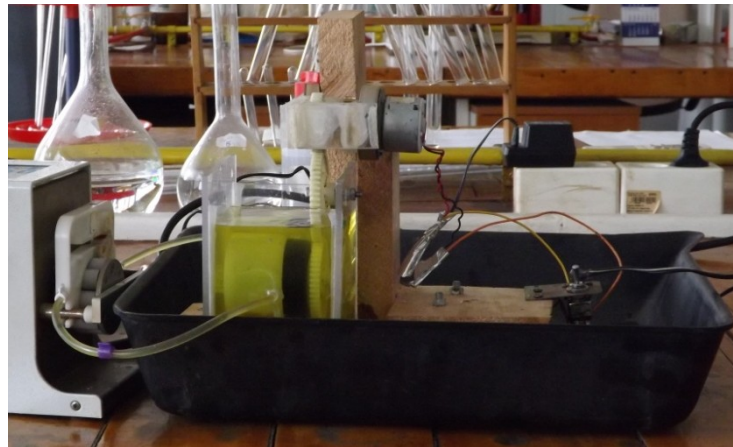

Figure 1: The modelled leaching reactor

Rotating wall boundary specification was applied to the walls of the rotating drum, calculating the velocity field using moving mesh method. A velocity inlet boundary was applied to the inlet, and an outlet for the outlet of the reactor. All the other boundaries were defined as walls. Figure 2a shows the solution of the momentum balance model, i.e. the velocity field within the reactor.
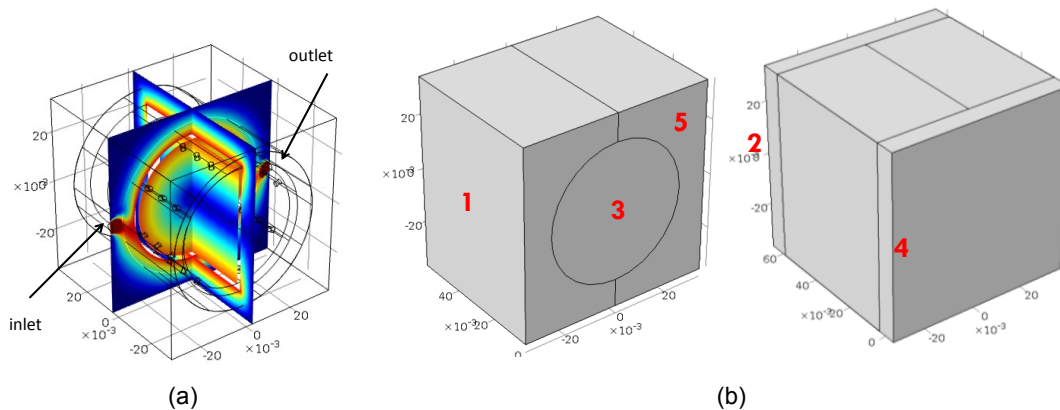

(a)   (b)

Figure 2: (a) The velocity field in the reactor, (b) The reactor compartments

The CFD model was solved at three different revolution speeds (10-20-30 1/min). The calculated velocities are higher near the inlet and the outlet; however the revolution speeds have the highest effect on the homogeneity inside the drum.

2.3 The compartment model

The real geometry was divided into five subsections to define the compartment model. Figure 2b shows the reactor compartments based on the reactor geometry. The drum in the middle section (3), the other two parts are from the inlet to the drum (1) and from drum to the outlet (5). The reactions only took place within the drum because only the rotated drum contains leachable metal which cannot be escaped from it. The fourth and fifth compartments are the compartments near the drum (2, 4), without any connection to it. Hence, they presumably will act as dead zones. Figure 3 shows the structure of the compartment model, extended with mixers and distributors to each compartment. To identify unknown model parameters a residence time distribution experiment was conducted. We compared four different cases, based on the degree of freedom in the compartment model parameters.

- Parameter identification for all parameters (1);
- Parameter identification with fixed volumes (2);
- Parameter identification with fixed volumes and based on the symmetries of the circulation numbers (3);
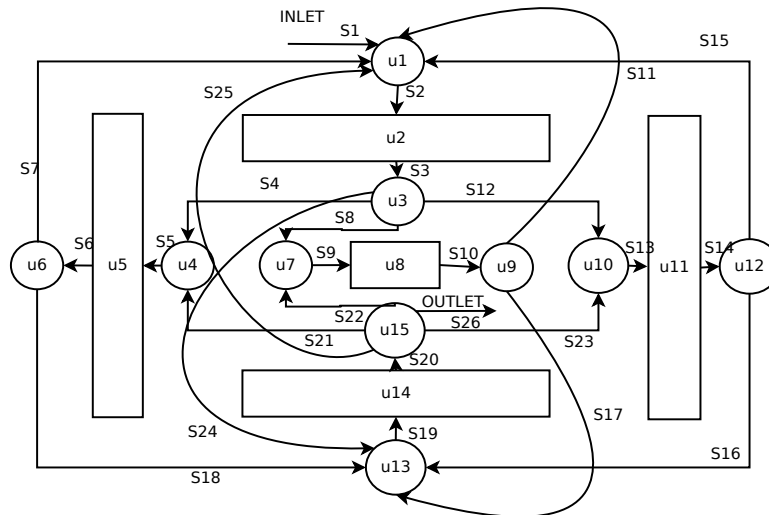- Simulation used to circulation number identification (4).

Figure 3: The proposed compartment model structure

Concerning the circulation numbers, the following simplifications can be applied in the third approach (based on heuristic assumptions):

- The fluid flows between the inlet compartment and the side compartments are the same
- The fluid flows between the outlet compartment and the side compartments are the same.

Table 2: Results of the compartment model parameter identification step in the case of 30 1/min revolution speed measurements

Parameter	1	2	3	4	Parameter	1	2	3	4
V_1	2.08e-5	8.08e-5	8.08e-5	8.08e-5	α_{25}	5.23e-1	6.38e-1	9.96e-1	5.10e-1
V_2	1.08e-5	3.68e-5	3.68e-5	3.68e-5	α_{31}	3.56e-1	9.72e-1	9.35e-1	4.97e-1
V_3	6.40e-5	7.48e-5	7.48e-5	7.48e-5	α_{35}	6.44e-1	2.80e-2	6.50e-2	5.03e-1
V_4	1.35e-5	3.68e-5	3.68e-5	3.68e-5	α_{41}	7.40e-1	4.25e-1	4.70e-3	4.39e-1
V_5	7.94e-5	8.08e-5	8.08e-5	8.08e-5	α_{45}	2.60e-1	5.75e-1	9.95e-1	5.61e-1
α_{12}	2.31e-1	4.01e-4	4.70e-3	3.31e-1	α_{51}	9.93e-2	8.44e-2	2.90e-2	1.59e-1
α_{13}	3.36e-1	9.25e-1	9.59e-1	7.40e-2	α_{52}	2.06e-1	9.05e-2	1.18e-1	1.06e-1
α_{14}	6.95e-2	3.30e-3	4.70e-3	9.14e-2	α_{53}	9.00e-2	4.89e-2	2.90e-2	3.23e-2
α_{15}	3.63e-1	7.12e-2	3.12e-2	5.04e-1	α_{54}	8.10e-1	1.84e-1	2.73e-1	2.87e-2

| α_{21} | 4.77e-1 | 3.62e-1 | 4.00e-3 | 4.90e-1 | α_{5out} | | 5.23e-1 | 5.93e-1 | 5.51e-1 | 6.74e-1 |

Practically in the fourth case the parameter identification step does not need because the circulation numbers and volumes were calculated from the CFD simulation. Table 2 shows the volumes and the identified circulation ratios. V refers to the columns and α refers to the circulation ratios.

All cases show a relatively small squared error value between the measured and the simulated cases, however, there are significant differences between the circulation ratios in each case. The CFD based methods has a significant advantage compared to the other methods, namely the effect of the revolution speed can be calculated, hence the circulation numbers derived from the CFD model is revolution speed dependent. After we validated our model against measurements with different revolution speed and initial concentrations, we used the validated compartment models for optimization. Figure 4a shows the results based on CFD simulation, and Figure 4b and c, the validation with different reagent concentration, and revolution speed.
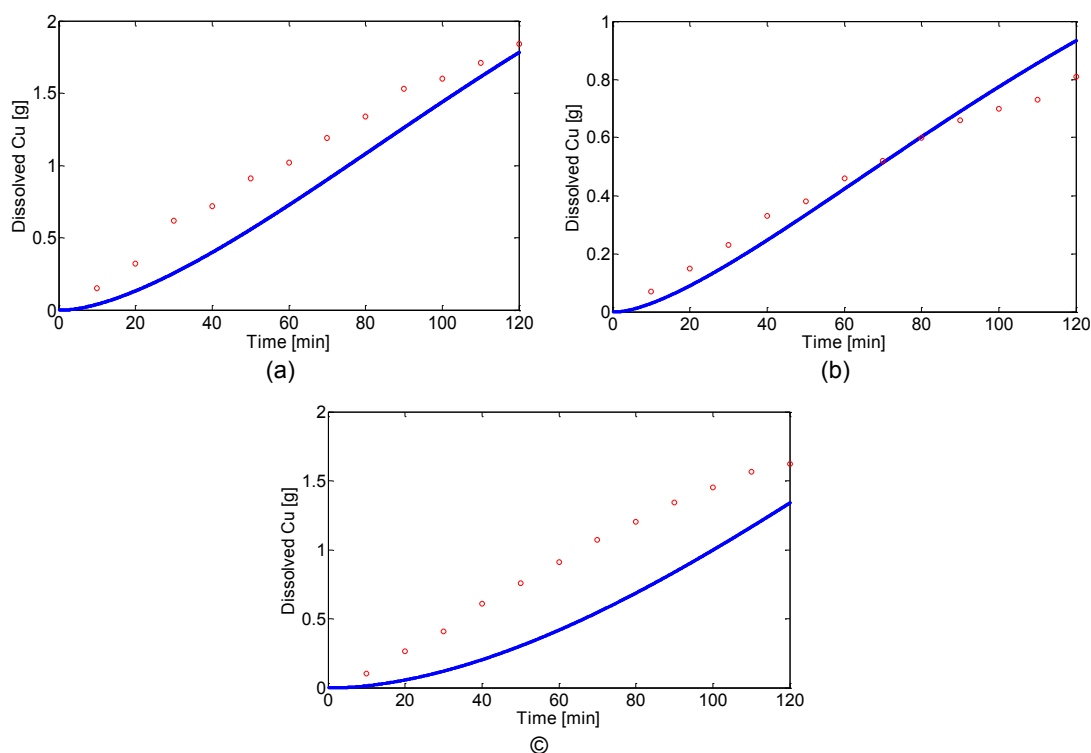

Figure 4: (a) CFD based results 0.2 M reagent (30 1/min), (b) Validation with 0.1M reagent (30 1/min), (c) validation with 0.2 M reagent (10 1/min)

Although there are differences between the measured and the simulated curves, the simulator calculates the leaching process reasonable accurately. The different cases has the following square error of a – 726.9, b – 77.2, c – 1538. However, a more detailed tuning step has to be evaluated.

2.4 Process optimization

The previously defined hybrid CFD-compartment model was used for process optimization. In the optimization step the effect of the revolution speed, and initial reagent concentration were examined. Besides, we examined the effect of the drum revolution direction, but it has almost no effect to the circulation numbers. Figure 5 shows the results of the optimization of operational parameters.

Figure 5 show that there are multiple possible optimums in this case and the optimal revolution speed is between 20 and 40 1/min, and concentrations 0.4 M and 0.5 M. The more detailed optimum search must contain the economic functions of motor power and reagent cost too.

3. Conclusions

A detailed model of the electrochemical reactor was developed. The model contains a CFD based hydrodynamic model, and a compartment model based kinetic and component balance model. The

hydrodynamic conditions was transferred to the compartment model using a hybrid approach, and based on averaged velocity values. The kinetics of the leaching process was identified, and then the hybrid model was validated against multiple measurements at different conditions. With the validated model we are able to optimize and intensify the process, as the optimization step of the study shows.
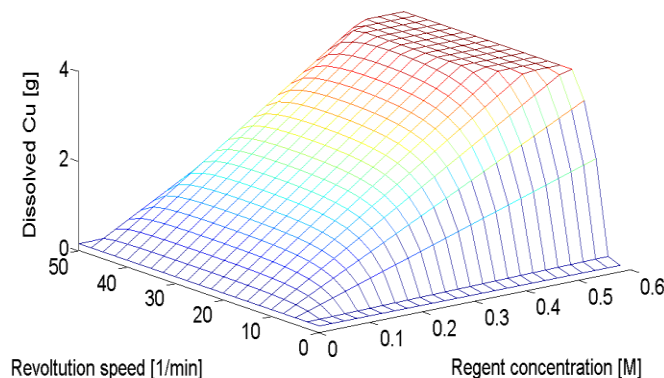

Figure 5: The effect of the revolution speed and initial reagent concentration on the leaching process

Acknowledgements

References

Delafosse A., Delvigne F., Collignon M-L., Thonart P., Crine D., 2010, Stochastic modeling of a micro-organism displacements in a stirred-tank bioreactor, Proceedings of CHISA 2010, Prague, P5.76

Fogarasi Sz, Imre-Lucaci F., Ilea P., 2012, Metals leaching from waste printed circuit boards. Part I: Efficiency and selectivity in FeCl$_3$ and CuCl$_2$ acidic solutions", Part II: Influence of thiourea, thiosulfate and thiocyanate concentration on the leaching process, Studia Universitatis Babes-Bolyai, LVII, 3, 31-49

Guha D., Dudukovic M.P., Ramachandran P.A., Mehta S., Alvare J., 2006, CFD based compartmental modelling of single phase stirred tank reactors, AIChE Journal, 52, 1836-1846

Maggioris D., Goulas A., Alexopoulos A.H., Chatzi E.G., Kiparossides C., 2008, Use of CFD in Prediction of Particle Size Distribution in Suspension Polymer Reactors, Computers and Chemical Engineering, 22, 315-322

Martinez-Delgadillo S., Mollinedo-Ponce H., Mendoza-Escamilla V., Gutiérrez-Torres C., Jiménez-Bernal J., Barrera-Diaz C., 2012, Performance evaluation of an electrochemical reactor used to reduce Cr(VI) from aqueous media applying CFD simulations, Journal of Cleaner Production, 34, 120-124

Rivero E. P., Granados P., Rivera F. F., Cruz M., González I., 2010, Mass transfer modelling and simulation at a rotating cylinder electrode (RCE) reactor under turbulent flow for copper recovery, Chemical Engineering Science, 65, 3042-3049

Tamburini A., Cipollina A., Micale G., 2011, CFD Simulation of Solid Liquid Suspensions in Baffled Stirred Vessels Below Complete Suspension Speed, Chemical Engineering Transactions, 24, 1435-1440

Van Parys H., Tourwé E., Breugelmans T., Fepauw M., Deconick J., Hubin A., 2008, Modeling of mass and charge transfer in an inverted rotating disc electrode (IRDE) reactor, Journal of Electroanalytical Chemistry, 622, 44-50

Yang J., Wu Y., Li J., 2012, Recovery of copper particles from metal components of waste printed circuit boards, Hydrometallurgy, 121-124, 1-6