

A CFD Model for Predicting the Fe Production in a Cylindrical Rotating Electro-winning Cell

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A CFD model for predicting the Fe production rate in a cylindrical rotating electro-winning cell is presented. The aim is to determine the production capacity for varying rotational speed and given cell dimensions. The model solves the transport of electrolyte (l), iron-ore (s), and O₂ (g) between the cell gap of two rotating co-centric cylinders, along with the current density between the anode and cathode. Taylor vortices are identified for angular frequencies larger than 0.5 rev/s.

The Fe production is initially raised for increasing angular frequency due to higher levels of turbulent fluctuations, promoting the removal of the produced O₂ gas from the anode and thus leading to a larger current density. Above a sufficiently large angular frequency, however, the Fe production is decreased due to the formation of Taylor vortices, hindering the removal of O₂ in the stagnation points of the counter rotating Taylor vortices.

These two counteractive effects lead to an optimum angular frequency in terms of Fe production at 0.25 rev/s for a rotating anode (inner cylinder) and at 0.75 rev/s for a rotating cathode (outer cylinder). Rotating the cathode (outer cylinder) leads to a 50% higher overall Fe production capacity.

Introduction

As restrictions on the emissions of industrially generated CO₂ become tighter, as a counter measure to climate change, it becomes increasingly important to study and develop alternative ways to produce metallic iron from its naturally occurring ores. One of the possible process routes is the reduction of iron oxides by means of electrolysis. Laboratory scale experiments, carried out in the ULCOS project, have shown promising results with an alkaline aqueous electrolysis process. With this technique, iron-ore particles are suspended in a OH⁻electrolyte solution and introduced between electrodes over which a potential difference is applied. At the anode, OH⁻ ions are oxidised into H₂O and O₂ gas, whereas at the cathode, iron oxide and water are reduced to metallic iron and OH⁻ ions. In such a system, care has to be taken that O₂ gas can not migrate from the anode to the cathode, as this may lead to the reversed oxidation reaction with the deposited iron. Several ways of preventing migration of O₂ to the cathode are possible, such as creating holes in the anode to allow the gas to escape, or by choosing a orientation of the electrodes such that the O₂ rises up due to gravity. The control of O₂ gas bubbles at the anode and the prevention of their migration to the cathode is a key factor in the successful design of a large scale electrolysis cell.

In the present research, the separation of O₂ is established by employing centrifugal forces generated by a rotating cylindrical cell (see Figure 1). The iron particles are denser than the electrolyte, and are thus forced to the outer cylinder, whereas the lighter O₂ gas bubbles are forced to the inner

cylinder. The ore suspension is made to flow upwards, leading to a Poiseuille flow in between the plates.

The flow in between concentric rotating cylinders is characterised by Taylor-Couette instabilities^[1-4]. These are counter rotating vortices stacked on top of each other which may interfere with the electro-winning process (see Figure 1). Depending on the rotational velocity, the vortices may be stable and laminar or unstable and turbulent. In order to be able to capture the formation of the Taylor vortices, it is necessary to use a 3D CFD model.

The commercial flow solver Fluent, release 6.3.26, is used to solve the transport equations. Using this approach, the influence of the rotational speed on the iron production is investigated. Finally, conclusions concerning the optimum configuration for high gas-phase removal and particle transfer to the cathode are given, and recommendations for further research are made.

Modelling assumptions

All process condition and geometrical layout parameters used in the present research are based on a standard ULCOS benchmark case such that comparisons with alternative electrolyse systems studied by other groups is allow afterwards. Electrolyte (l) enters the domain at the bottom of the cell at a speed of 1 mm/s. The density of the electrolyte is 1000 kg/m³, and its kinematic viscosity is 10⁻⁶ m²/s. Produced O₂ gas is treated as the dispersed phase with a void fraction α_{O_2} and a bubble diameter of 1 mm, corresponding to alternative CFD models developed with ULCOS⁵. The electrolyte contains a

volume fraction iron ore of $\alpha_{ore}=0.2$ at the inlet, as prescribed by the ULCOS benchmark.

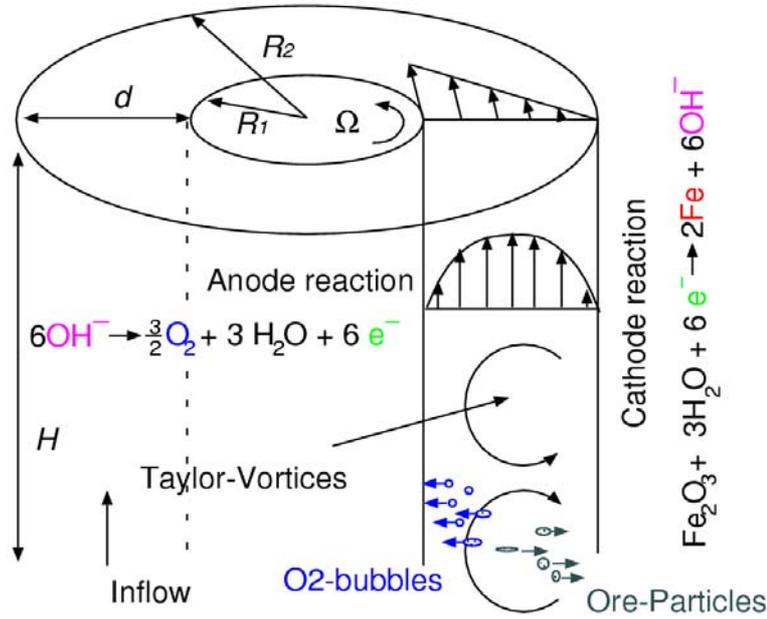


Figure 1. Diagram of concentric rotating cylinders with electrolyte flowing in between anode (inner cylinder) and cathode (outer cylinder). Due to the centrifugal forces, oxiden gas bubble are moving inward, and the iron-ore particles are moving outward.

In order to solve the transport of the phases (ore/gas/liquid), two approaches can be used. The first approach (used by Serna) disregards any variations in ore density and solves a single phase flow of a uniform ore-suspension, whereas the influence of the O_2 gas bubbles is modeled with a Discrete Particle Model (DPM). In this approach, each gas bubble is tracked individually within the single phase system, and the influence of the bubble is coupled back as a momentum source term in the transport equation of the electrolyte. Although this approach is applicable for the low density used in this benchmark, DPM modeling can not be applied on the iron-ore, and also becomes computationally demanding in a full three dimensional flow (the model of Serna^[5] is 2D and does not solve the iron-ore phase). The formation of Taylor vortices can only be predicted in a 3D simulation, and on top of that, also the iron-ore concentration is not necessarily uniform. In order to be able to solve the transport of all the three phase in a three dimensional cylinder, the computationally expensive DPM model is now replaced with a Euler approach that solve the transport equation of each phase individually by describing the gas and ore phase in terms of the volume fractions α_{O_2} and α_{ore} , respectively. The momentum transfer between the phases is described by the Schiller-Neumann model, which assumes spherical particles. The full description of the Euler model falls outside the scope of the present article, and can be found in any standard fluid mechanics textbook, e.g. the Fluent manual^[6].

The focus in the paper is in solving the gas distribution over a rotating cell. Therefore, as a first attempt, only the primary current distribution is calculate, following

$$\vec{j} = \kappa \vec{\nabla} \phi \quad (1)$$

where ϕ is the local electrical potential and κ the local electrical conductivity of the electrolyte. A constant voltage difference of 0.07 V is applied between the electrodes (based on the prescribed ULCOS standard benchmark case). This approach neglects any kinetic and concentration effects.

This electrical conductivity depends on the volume fraction of O_2 gas and iron ore according to^[3]

$$\kappa = \kappa_0 (1 - \alpha_{O_2} - \alpha_{ore})^{3/2} \quad (2)$$

Here κ_0 , the conductivity of pure electrolyte is a function of the local concentration of OH^- ions acting as charge carriers:

$$\kappa_0 = F^2 \cdot \frac{D_{OH}}{RT} C_{OH} \quad (3)$$

where F is Faraday's constant, R is the gas constant, T is the temperature, and C_{OH} is the OH^- concentration.

The diffusivity D_{OH} is written as

$$D_{OH} = D_{OH,0} (1 - \alpha_{O_2} - \alpha_{ore})^{3/2} \quad (4)$$

The OH^- concentration is solved as a passive scalar in the flow domain with a source at the cathode and a sink at the anode (see Figure 1). The electrical current density which follows from equation (1) is then directly converted to a Fe production rate per area unit according to

$$r_{FE} = j_{cat} M_{FE} / (3F) \quad (5)$$

Where j_{cat} is the current density at the cathode, $M_{FE} = 0.5585$ kg/mol is the molar mass of Fe, and the factor 3 reflects the number of electrons required per produced Fe atom. This production rate model merely is an upper limit disregarding any effect electrical chemical effects at the molecular scale. The model focuses therefore on the effect of formed O_2 gas and hydrodynamics on the Fe production. For a more complete model, a more sophisticated model than equation (4) should be applied.

Finally, standard cell dimensions are chosen (i.e.: according to the ULCOS standard benchmark) such that the results can be compared with other cells investigated within the electrolyses sub-project: height $H=500$ mm, inner radius $R_1=190$ mm, and outer radius $R_2=200$ mm (i.e. gap width=10 mm).

Results

Figure 2 shows distribution of the velocity magnitude and the turbulent kinetic energy of the cell gap for increasing rotational speed for both the rotating cathode (outer cylinder at 10mm) and the rotating anode (inner cylinder at 0 mm). For zero rotational speed, the contour plots for the rotating anode and cathode are (of course) identical. An increase of both the velocity magnitude and the turbulent kinetic energy is observed for increasing rotational speed of both the anode and the cathode. For the rotating cathode, a clear periodic horizontally aligned pattern of increasing velocity and turbulent kinetic energy is found at 1 rev/s, reflecting the formation of Taylor vortices as anticipated earlier. This pattern is more profound for the rotating anode case, as Taylor instabilities are more likely to occur for a rotating outer cylinder [1-4].

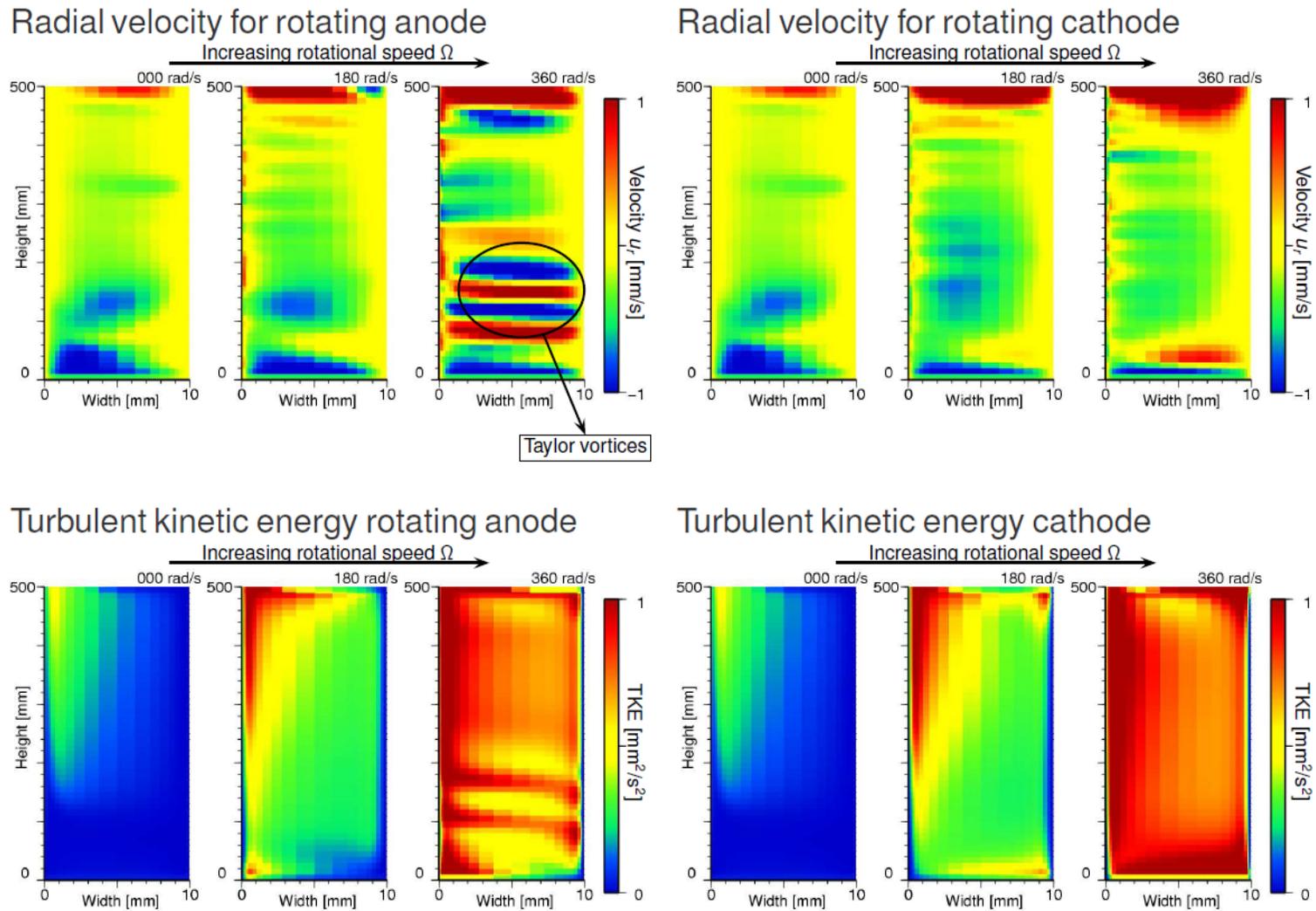


Figure 2. Effect of increasing rotational speed on radial velocity magnitude (top) and turbulent kinetic energy (bottom) for rotating inner electrode (left) and outer electrode (right).

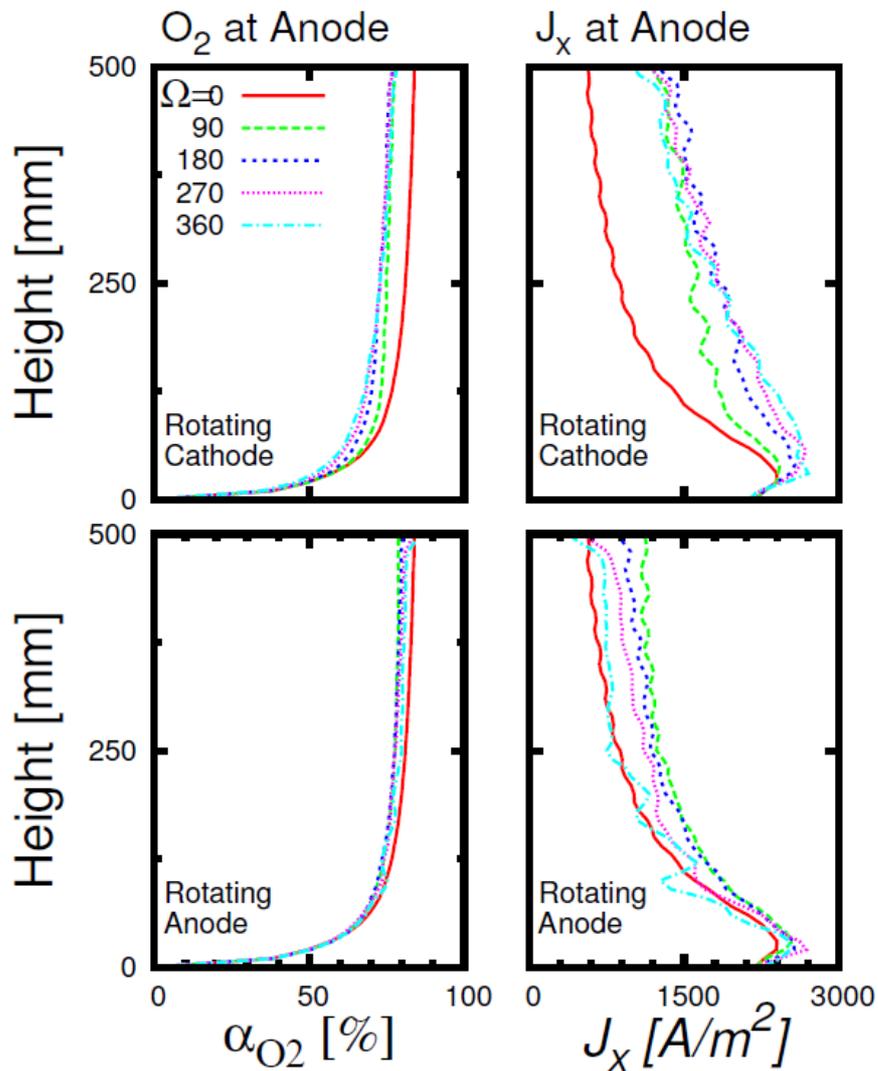


Figure 3 . Effect of rotational speed Ω (in degrees/second) on O_2 distribution and current density

Both the gas hold-up and the current density over the anode are plotted in Figure 3. The O_2 distribution over the anode shows a gradual increase in downstream direction (i.e. flowing upward). The O_2 gas that is formed has the tendency to stay near the wall and this is affected by two counter-acting factors. Firstly, the increasing turbulent kinetic energy generated by the rotation of the cell tends to enhance the removal of the gas away from the wall. This effect is shown in Figure 3 for the rotating cathode case by the lower O_2 concentration for higher rotational speeds. The second factor is the formation of Taylor vortices which interfere with this removal, as the counter rotating vortices tend to push the bubbles to the wall in the stagnation points. Taylor vortices are formed at a higher rotational velocity, which is reflected by the increase of O_2 near the wall for the higher rotational speeds. The O_2 concentration near the anode remains higher for the rotating anode

(inner cylinder) as Taylor vortices are formed more easily, as shown in Figure 2.

The current density shown in Figure 3 is linked to the O_2 concentration profiles via the relations (1-5) presented in the previous section. As a result, the current density remains higher for the rotating cathode, as the lower gas hold up allows high current densities. Rotating the anode leads to more pronounced Taylor vortices, and consequently a high gas hold up at the anode side. The local increases and decreases in O_2 concentration caused by the Taylor vortices are reflected in the oscillations of the current density along the anode shown in Figure 3 for the higher rotational speeds.

The total amount of Fe produced at the cathode (calculated by integrating the local Fe production at the cathode over the cell height) is plotted against the rotational velocity of both the anode and cathode

in Figure 4. Since the Fe production is linearly related to the current density via equation (5), the same maximum in Fe production as in the current density is observed. For the rotating anode, a maximum Fe production of 60 mg/s is found at 0.25 rev/s, and for the rotating cathode a maximum Fe production of about 87 mg/s is found at 0.75 rev/s. The production of Fe is, therefore, related to the efficiency of removal of O₂ gas from the anode: the more bubbles that are removed, the lower the void fraction and consequently the higher the current density and Fe production rate. Or in other words: the variation in current density on the cathode (and therefore the Fe production rate) is linked to the inhomogeneous

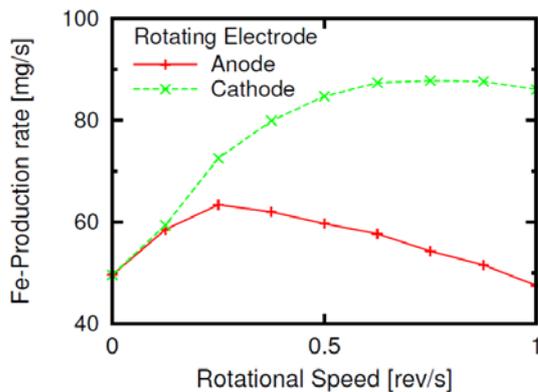


Figure 4. Fe production rate vs rotational speed for rotating anode and cathode, respectively

oxygen distribution.

Conclusion

A multiphase CFD model for predicting the iron production in a rotating cylindrical electro-winning cell has been developed. The model solves the transport of electrolyte (l), iron-ore (s), and O₂ (g) between the cell gap of two rotating concentric cylinders, along with the current density between the anode and cathode. Taylor vortices are identified for angular frequencies > 0.5 rev/s. The Fe production is initially increased for increasing angular frequency due to increase of turbulent fluctuations level, promoting the removal of the produced O₂ gas from the anode and thus leading to a larger current density. Above a sufficiently large angular frequency, however, the Fe production is decreased due to the formation of Taylor vortices, hindering the removal of O₂ in the stagnation points of the counter rotating Taylor vortices. These two counteractive effects lead to an optimum angular frequency in terms of Fe production: at 0.25 rev/s for a rotating anode (inner cylinder) and at 0.75 rev/s for a rotating cathode (outer cylinder). Rotating the cathode (outer cylinder) leads to a 50% higher overall Fe production capacity.

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¹ Priority 3 of the 6th Framework Programme in the area of "Very low CO₂ Steel Processes", in co-ordination with the 2003 and 2004 calls of the Research Fund for Coal and Steel