

Original Research

Design of optimum solid oxide membrane electrolysis cells for metals production

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Received 17 September 2015; accepted 30 October 2015

Available online 24 December 2015

Abstract

Oxide to metal conversion is one of the most energy-intensive steps in the value chain for metals production. Solid oxide membrane (SOM) electrolysis process provides a general route for directly reducing various metal oxides to their respective metals, alloys, or intermetallics. Because of its lower energy use and ability to use inert anode resulting in zero carbon emission, SOM electrolysis process emerges as a promising technology that can replace the state-of-the-art metals production processes. In this paper, a careful study of the SOM electrolysis process using equivalent DC circuit modeling is performed and correlated to the experimental results. A discussion on relative importance of each resistive element in the circuit and on possible ways of lowering the rate-limiting resistive elements provides a generic guideline for designing optimum SOM electrolysis cells.

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Keywords: Electrolysis; Solid oxide; Membranes; Metals production; Environmentally benign

1. Introduction

Solid oxide membrane (SOM) electrolysis is an electrolytic technique for the production of metals (Me) directly from their respective oxides (MeO_x). For SOM electrolysis cells, the overall electrochemical reaction is given as



where x is the stoichiometric amount of oxygen in the metal oxide. To date, the SOM electrolysis process has been applied for the production of various technologically important metals, such as Mg, Al, Ti, Ta, Yb, and Si [1–9]. This process has also been adapted to produce alloys and intermetallics, such as Ti–Fe alloy, Ti–Si intermetallics, and CeNi₅ [10–13].

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Peer review under responsibility of Chinese Materials Research Society.

Fig. 1 shows a schematic illustration of a SOM electrolysis cell employing an inert oxygen anode [3]. An oxygen-ion-conducting SOM typically made of yttria-stabilized zirconia (YSZ) separates the inert anode from a molten salt (flux) electrolyte and a cathode. During electrolysis, an applied potential exceeds the dissociation potential of the metal oxide dissolved in the molten salt. The desired metal is reduced at the cathode while oxygen ions are transported through the SOM and are oxidized to pure O₂ gas at the inert anode.

2. Equivalent DC circuit

Equivalent DC circuit is a useful tool to gain an insight into the SOM electrolysis process. Fig. 2(a) presents a general equivalent DC circuit that takes into account all the known mechanisms associated with the current flow: (1) the dissociation of the desired oxide, (2) the dissociation of the impurity oxides (undesired oxides) dissolved in the flux, (3) electronic

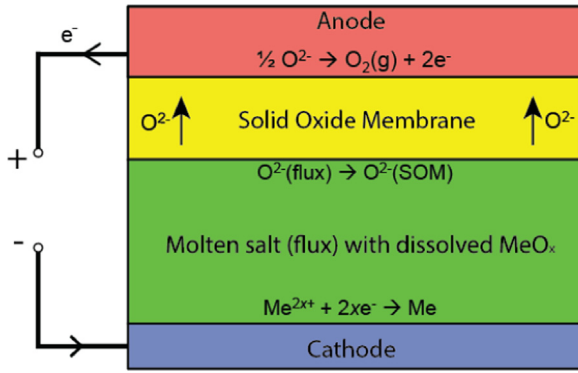


Fig. 1. Schematic illustration of a SOM electrolysis cell.

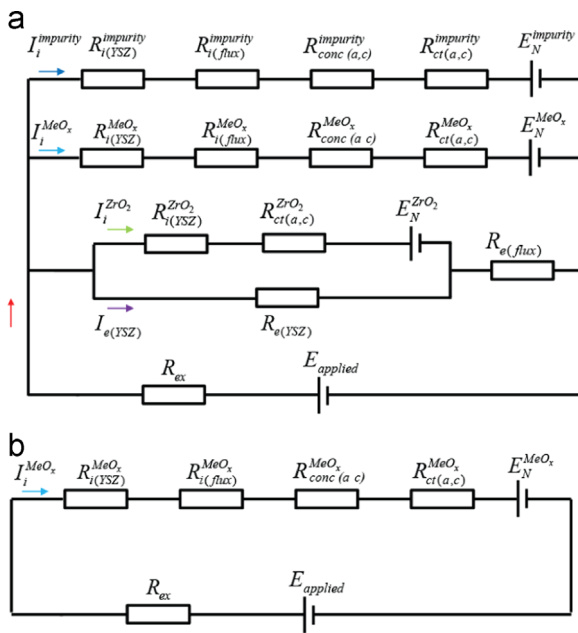


Fig. 2. Equivalent DC circuit of the SOM electrolysis cell: (a) general case and (b) ideal case.

conductivity of the flux caused by either the intrinsic electronic conductivity or the metal solubility in the flux, and (4) the various resistive contributions of the SOM [5]. The symbols used in Fig. 2 are defined in Table 1. Contributions related to the impurity oxides and the electronic conductivity of the flux are undesirable. The presence of impurity oxides in the flux can lower the purity of the metal product, and the electronic conductivity of the flux can reduce the Faradaic current efficiency for metals production. The electronic conductivity of the flux also provides a pathway for the applied potential to reduce ZrO_2 in the YSZ membrane. The issues related to impurity oxides can be mitigated by performing pre-electrolysis at lower applied potentials or through careful selection of the feed material. The issues related to the electronic conductivity of the flux needs to be mitigated by removing the sources that contribute to generating the

Table 1

Definitions of symbols in the SOM equivalent DC circuits shown in Fig. 2.

Symbol	Definition
$R_{i(YSZ)}^{impurity}$	Ionic resistance of YSZ membrane involved for impurity oxides dissociation
$R_{i(YSZ)}^{MeO_x}$	Ionic resistance of YSZ membrane involved for MeO_x dissociation
$R_{i(YSZ)}^{ZrO_2}$	Ionic resistance of YSZ membrane involved for ZrO_2 dissociation
$R_{i(flux)}^{impurity}$	Ionic resistance of flux involved for impurity oxides dissociation
$R_{i(flux)}^{MeO_x}$	Ionic resistance of flux involved for MeO_x dissociation
$R_{conc(a,c)}^{impurity}$	Concentration polarization resistance at the anode and cathode for impurity oxides dissociation
$R_{conc(a,c)}^{MeO_x}$	Concentration polarization resistance at the anode and cathode for MeO_x dissociation
$R_{ct(a,c)}^{impurity}$	Charge transfer resistance at the anode and cathode for impurity oxides dissociation
$R_{ct(a,c)}^{MeO_x}$	Charge transfer resistance at the anode and cathode for MeO_x dissociation
$R_{ct(a,c)}^{ZrO_2}$	Charge transfer resistance at the anode and cathode for ZrO_2 dissociation
$R_{e(YSZ)}$	Electronic resistance of the YSZ membrane
$R_{e(flux)}$	Electronic resistance of the flux between YSZ and bubbling tube
R_{ex}	Resistance of external lead wires and the current collectors
$E_N^{impurity}$	Nernst potential for impurity oxides dissociation
$E_N^{MeO_x}$	Nernst potential for MeO_x dissociation
$E_N^{ZrO_2}$	Nernst potential for ZrO_2 dissociation
$E_{applied}$	Applied potential
$I_i^{impurity}$	Ionic current for impurity oxides dissociation
$I_i^{MeO_x}$	Ionic current for MeO_x dissociation
$I_i^{ZrO_2}$	Ionic current for ZrO_2 dissociation
$I_{e(YSZ)}$	Electronic current passing the YSZ membrane

electronic carriers or by creating an electron blocking layer around the SOM [4,14].

Fig. 2(b) shows the equivalent DC circuit for an ideal SOM cell where the circuit branches for the impurity oxides and the electronic conductivity of the flux have been removed from the general equivalent DC circuit.

3. Polarization model for an ideal SOM cell

According to the equivalent circuit shown in Fig. 2(b), $E_{applied}$ can be expressed by

$$E_{applied} = \left| E_N^{MeO_x} \right| + \eta_{ohm} + \eta_{ct(a,c)} + \eta_{conc,c} + \eta_{conc,a} \quad (2)$$

where $\left| E_N^{MeO_x} \right|$ is the absolute value of the Nernst potential for MeO_x dissociation, η_{ohm} is the ohmic polarization of the SOM cell, $\eta_{ct(a,c)}$ is the charge transfer polarization, $\eta_{conc,c}$ is the cathodic concentration polarization, and $\eta_{conc,a}$ is the anodic concentration polarization.

3.1. Ohmic polarization, η_{ohm}

Literature related to SOM electrolysis for Mg production reports that the ohmic polarization dominates the total polarization [5]. Therefore, it is critical to reduce η_{ohm} to improve

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