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Review

Mixed ionic–electronic conducting (MIEC) ceramic-based membranes for oxygen separation

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ABSTRACT

Although Nernst observed ionic conduction of zirconia–yttria solutions in 1899, the field of oxygen separation research remained dormant. In the last 30 years, research efforts by the scientific community intensified significantly, stemming from the pioneering work of Takahashi and co-workers, with the initial development of mixed ionic–electronic conducting (MIEC) oxides. A large number of MIEC compounds have been synthesized and characterized since then, mainly based on perovskites ($ABO_{3-\delta}$ and $A_2BO_{4\pm\delta}$) and fluorites ($A_\delta B_{1-\delta}O_{2-\delta}$ and $A_{2\delta}B_{2-2\delta}O_3$), or dual-phases by the introduction of metal or ceramic elements. These compounds form dense ceramic membranes, which exhibit significant oxygen ionic and electronic conductivity at elevated temperatures. In turn, this process allows for the ionic transport of oxygen from air due to the differential partial pressure of oxygen across the membrane, providing the driving force for oxygen ion transport. As a result, defect-free synthesized membranes deliver 100% pure oxygen. Electrons involved in the electrochemical oxidation and reduction of oxygen ions and oxygen molecules respectively are transported in the opposite direction, thus ensuring overall electrical neutrality. Notably, the fundamental application of the defect theory was deduced to a plethora of MIEC materials over the last 30 years, providing the understanding of electronic and ionic transport, in particular when dopants are introduced to the compound of interest. As a consequence, there are many special cases of ionic oxygen transport limitation accompanied by phase changes, depending upon the temperature and oxygen partial pressure operating conditions. This paper aims at reviewing all the significant and relevant contribution of the research community in this area in the last three decades in conjunction with theoretical principles.

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1. Introduction

Although Nernst [1] reported the ionic conductivity in a $ZrO_2(Y_2O_3)$ solid solution in 1899, novel application of these solid solution oxides for O_2 separation attracted the attention of the research community only in the last 30 years, stemmed from Takahashi et al.'s [2] initial development of mixed ionic–electronic conduction in oxide materials such as Bi_2O_3–BaO. The introduction of mixed conducting solid oxides concept for oxygen semi-permeable membranes was introduced by Cales and Baumard [3,4] over 25 years ago.

While past research concentrated on improvements of oxygen ionic conductor for use in fuel cells, recent efforts are motivated to introduce the electronic conductivity into materials that predominantly are ionic conductors. Defect-exclusive dense ceramic membranes show superior oxygen separation, ensuring that only pure oxygen exists in the permeate stream, thus resulting in theoretically infinite selectivity. The highest oxygen flux is generally observed for dense ceramic membranes having a perovskite structure at high temperature in excess of 800 °C [5]. In addition to producing high purity oxygen, dense ceramic membranes can also be integrated in catalytic membrane reactors for carrying out different petrochemistry processes such as oxidative coupling of methane to C_2 (ethylene and/or ethane) (OCM), partial oxidation of methane to syngas (POM), partial oxidation of heptane to hydrogen (POH), selective oxidation of ethane to ethylene (SOE), and selective oxidation of propane to propylene (SOP) [6–13], during which both separation and catalytic processes are achieved in a single step. The mixed conducting membrane technology is also commercially prospective for an air separation unit integrated with a hot turbine system [14]. Another interesting example of the coupling between oxygen separation and catalysis is the ammonia oxidation into NO in the nitric acid manufacturing process [15].

There are numerous reports investigating the modification of mixed ionic–electronic conducting materials and corresponding changes in oxygen flux. Several excellent reviews on mixed conducting ceramics focusing on catalytic processes have been published. For example Yang et al. [12] reviewed the development and challenges around perovskite-type materials for use in OCM, POM, POH, SOE and SOP; Liu et al. [16] outlined various doping scenarios centralized around perovskite compounds to boost its oxygen permeation flux including their outcomes and progressions for several class of perovskite compounds as well as a brief transport theory and summary of its potential applications in different areas. However, a review of mixed conducting compounds based on their limiting transport mechanism and structures in detail has yet to be written. Therefore, this review endeavors to highlight the development of dense ceramic-based membranes in the last 30 years, in particular mixed ionic–electronic ceramic membranes for oxygen separation application. In addition, novel materials with improved oxygen permeability are discussed together with the transport theory of oxygen.

2. General background

2.1. Current ceramic oxygen separation technology

The two main types of oxygen separation systems based on ceramic membranes are pure oxygen conducting membranes and mixed ionic–electronic conducting membranes. A driving force has to be provided for oxygen to permeate through the membrane. This driving force can be either an electrical potential gradient or a

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