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ACCEPTED MANUSCRIPT

Corrosion of metal electrodes in deep eutectic solvents

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Abstract

Deep eutectic solvents (DESs) represent a green and non-expensive substitute for ionic liquids as nonaqueous electrolytes (NAEs). Electrochemical applications in NAEs benefit from the extended potential windows, due to the absence or minimization of water. However, interactions between electrode and electrolyte, as well as their degradation, embody a relevant but unexplored territory for the implementation in electrochemical systems. Electrochemical characterization of six electrode–electrolyte couples (three electrodes in two different DESs) were conducted by means of cyclic voltammetry and chronoamperometry. Besides conventional electrochemical characterization, ICP-OES, TOC, SEM and EDX measurements were used to track the transformations in the system. This work represents a first attempt to develop a methodology for the characterization of electrode corrosion in DESs.

Keywords: metal corrosion, deep eutectic solvents, electrochemistry, electrodes' stability

1. Introduction

The use of aqueous electrolytes in electrochemical applications is mainly limited by their operating potential window since the thermodynamic limit is at 1.2 V [1]. Furthermore, the use of NAEs such as ionic liquids and DESs can lead to modification of the redox kinetics [2,3,4], influence the formation of the passivation layers due to absence of dominant water splitting reaction [5,6] and expand the temperature and voltage range of application [7, 8]. Therefore, the interest in new NAEs increased [9]. One innovative group of NAEs are the so called deep eutectic solvents (DESs). DESs are eutectic mixtures consisting of two or more components which form homogeneous liquids through hydrogen bonding [9, 10]. Their importance in research is growing due to their superior properties over ionic liquids (ILs). Precisely, DESs are considered non-toxic, green and low-cost compared to ILs [10, 11]. The main applications of DESs include energy storage [12, 13], biomass-electroconversion [14, 15], electrodeposition [16, 17] and homogeneous catalysis [18]. Although passivation and corrosion behaviors in agueous solutions have been thoroughly investigated [19, 20, 21], only few studies were conducted in the field of NAEs. A pioneering study was conducted by Haerens regarding the electrochemical stability of a DES made of ethylene glycol-choline chloride (ethaline) over eight days of electrolysis. As a result, the formation of decomposition products was observed [22]. Hartley used extended X-ray absorption to determine the speciation for 25 metal salts in different DESs [23]. The study by Abbott focused on the interaction between the metal electrode and ethaline. Nine different metals were electrochemically characterized in ethaline using linear sweep voltammetry [24]. However, the electrode-DES interactions and the possible degradation products that can be formed during this interaction are not fully investigated. Here, we present the investigation of the electrochemical corrosion of three metal electrodes (titanium, nickel and iron) and their interactions in two different DESs, namely ethaline and proline-lactic acid (PL). with an unprecedented insight into the stability of the electrochemical system. Ethaline is one of the most utilized DES due to high conductivity and PL was chosen as an example of chlorine-free DES. The electrochemical behavior of the electrodes is investigated using cyclic voltammetry (CV) and chronoamperometry. The changes in the electrolyte were analysed using inductively coupled plasma optical emission spectrometry (ICP-OES) and total organic carbon (TOC). The changes in the electrodes' surface were studied using scanning electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDX).

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