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Application of ionic liquids for metal dissolution and extraction

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ABSTRACT

This review summarizes the results of studies on the selective dissolution and extraction of Fe, Cr, Cu, and Zn by ionic liquids, as an alternative to the use of conventional molten salts or pickling agents, for various types of steel. Ionic liquids are classified according to the metals, metal ions, and metal oxides by which they can be extracted or dissolved. The results of the metal extraction efficiency per unit time presented in the literature are summarized in a simple unified graphic format. This provides a comparative understanding of the most efficient ionic liquid for the extraction of specific metals.
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Introduction

The steel manufacturing process is subjected to various heat treatments, which essentially produce scales (mostly oxides) on a metal surface [1]. Such oxides need to be removed before and/or during the following manufacturing industrial operations. The various methods of descaling can be broadly classified into physical methods and chemical methods [2,3]. Typical physical methods include mechanical cleaning, such as scraping, abrasive blasting, and waterjet spraying [4]. Two major chemical methods include aqueous alkaline cleaning (with some mild alkaline solutions, such as sodium hydroxide, sodium phosphate, and sodium carbonates) and acid cleaning, known as pickling (with various strong acids and/or solutions of acid mixture), which are often environmentally hazardous [5–9]. In many cases, for most metal-rich scale removal methods, a very aggressive action needs to be used, with a very strong acid mixture, which is often preceded by pretreatment such as an alkaline salt bath or shot blasting process [4,10].

The choice of acid solution for the pickling process depends on the type of steel and product style. For general carbon steel, hydrochloric acid is mainly used. Also, for a thick stainless steel plate (hot rolled steel sheet, 3–8 mm), sulfuric acid and a mixed acid (nitric acid + hydrofluoric acid) solution are used. In general, for a thin steel plate (cold rolled steel plate, 0.3–3 mm), molten salt ($NaOH + NaNO_3$), sulfuric acid, and mixed acid are sequentially applied [11,12]. For processes with nitric acid, a $DeNO_x$ system needs to be used to remove NO_x, while the removal of nitrogen from the waste acid is critical to avoid possible environmental problems [2]. Recently, technologies have been developed in which nitric acid is replaced with sulfuric acid, and have been commercialized as an environmentally friendly pickling process [2,13–16]. Non-nitric acid pickling solutions are often composed of sulfuric acid, hydrofluoric acid, and hydrogen peroxide. While hydrogen peroxide, as a substitute for nitric acid, plays a role similar to nitric acid in the pickling process, it is expensive, and easily decomposes into water and oxygen at the surface of metal at temperatures higher than 45 °C [14].

Molten salts are very strong basic compounds, and generally require a high operating temperature of 350–550 °C, which makes the process very effective for pickling or oxide removal, particularly for stainless steel [17]. When Fe or Cr metal reacts with NO_3^- at high temperature, it produces a bulky material, which generates fine cracks between the oxide and the reactant upon immediate water-cooling, due to thermal shock [18]. The permeation of the mixed acid (nitric acid + hydrofluoric acid) into these cracks facilitates the removal of the oxide layer, by dissolving the boundary between the oxide and the base metal. While many environmentally harmful substances are being replaced with other materials, no alternatives have yet been found that outperform the oxide removal ability of toxic molten salts, particularly in a moderate temperature range.

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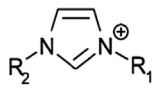
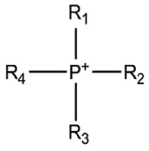
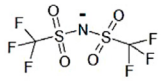
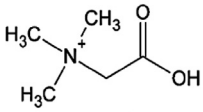
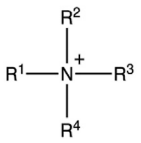
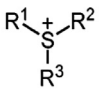
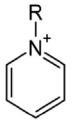
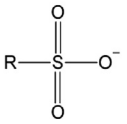
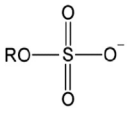
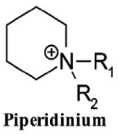
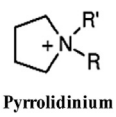
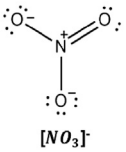
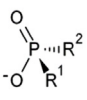
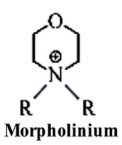
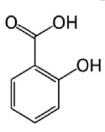
We considered the use of an ionic liquid as a substitute for molten salts, as well as some acidic pickling solutions. Ionic liquids are of interest, since they can dissolve some metal oxide even in ambient condition. We therefore investigated the possibility of using some ionic liquids and their mixtures to replace the molten salt system for pickling a number of metal oxides. Selective extraction and dissolution of many metals from ores and various oxidized forms by ionic liquids have attracted significant attention in the steel industry over the last decade [15,16,19–21], and a variety of ionic liquids have been synthesized for this purpose. Further, the use of ionic liquids is more practical and environmentally friendly than molten salt, because of the negligible vapor pressure and recyclability. The main aim of this study was to review the possibilities of (1) a method of dissolving the metal oxide directly in the ionic liquid, (2) a method in which the oxide does not need to be directly dissolved in the ionic liquid, while other acids can transform the oxide layer into a special form that is easily removable, and (3) an efficient method of dissolving metal oxides by an additional secondary process, such as electrochemical anodization. The first point is the main theme of this review, while the second and third points are of continuous ongoing interest as additional strategies with some efficient ionic liquids for pickling. As a first stage, we focused on collecting ionic liquids with dissolution and extraction capabilities for Fe and Cr, which are two major components in stainless steels, as well as Cu and Zn, which are major components in brass.

Ionic liquids for the extraction and dissolution of metal and metal oxides

Ionic liquids are substances that exist in a liquid state, where cations and anions are not in a crystalline state at temperatures below 100 °C [22,23]. Many ionic liquids are present in a liquid state even at room temperature (~25 °C); these ionic liquids have the significant advantage of being able to be used in a relatively freely synthesized structure of cations and anions according to the purpose. Table 1 summarizes the structures of several cations and anions commonly used in ionic liquids. It is known that the combinations of cations and anions of these ionic liquids can theoretically synthesize approximately 10^{18} new ionic liquids. Ionic liquids are used as solvents, refrigerants, absorbents, electrochemical electrolytes, and lubricants because of their low volatility, high thermal stability, and good conductivity. The first ionic liquid synthesized was ethanolammonium nitrate, reported by Gabriel and Weiner in 1888 [24]. Studies on the extraction and separation of metals and metal oxides using ionic liquids started in earnest in the 1980s [25]. In early 1980, Seddon and Hussey reported on the dissolution of transition metal compounds using the ionic liquid chloroaluminate as a non-aqueous polar solution [26]. In the 1990s, several ionic liquids were reported to have been used as solvents for transition metal catalysts [27]. Up to the present day, various ionic liquids containing various cations and anions have been synthesized, and studies on the extraction efficiency of metal and metal oxides using the synthesized ionic

Table 1
General cation and anion structures of ionic liquids that are widely used for the extraction and dissolution of metals and metal oxides.

Cation		Anion	
 Imidazolium	 Phosphonium	 $[Tf_2N]^-$	 $[PF_6]^-$
 Betanium	 Ammonium	 $[BF_4]^-$	 Halide anion
 Sulfonium	 Pyridium	 Sulfonate anion	 Alkyl sulfate anion
 Piperidinium	 Pyrrolidinium	 $[NO_3]^-$	 Phosphinate anion
 Morpholinium		 Salicylate	

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