

# A New Class of Magnetic Fluids: bmim[FeCl<sub>4</sub>] and nbmim[FeCl<sub>4</sub>] Ionic Liquids

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The responses to a magnet of two room-temperature ionic liquids containing tetrachloroferrate(III) ions, 1-butyl-3-methylimidazolium tetrachloroferrate (bmim[FeCl<sub>4</sub>]) and 1-butyronitrile-3-methylimidazolium tetrachloroferrate (nbmim[FeCl<sub>4</sub>]) are compared. Although their magnetic susceptibilities are similar, the observed responses are distinct from each other, suggesting that the response is determined not only by the magnetic susceptibility but also by the other factors including density, viscosity, and surface tension. The two magnetic ionic liquids constitute a new class of magnetic fluids that hold many attractive physical properties for practical applications.

**Index Terms**—1-butyl-3-methylimidazolium, 1-butyronitrile-3-methylimidazolium, ionic liquid, magnetic fluid, tetrachloroferrate(III) ion.

## I. INTRODUCTION

**I**ONIC LIQUID (IL) refers to a class of liquids that are composed solely of ions [1]. It is a synonym of molten salt. IL in a narrow sense often means room-temperature ionic liquid (RIL) that exists as liquid at room temperature. Although the dipolar and/or higher order multipolar interactions are dominating in ordinary molecular liquids, Coulomb interaction plays a major role in ILs. Owing to the dominating Coulomb interaction, specific liquid structures may exist in ILs that are not expected for molecular liquids. In fact, recent spectroscopic as well as diffraction studies have revealed several new structural properties that are indicative of ordered local structures in 1-butyl-3-methylimidazolium (bmim<sup>+</sup>) based ILs [2]–[6]. Raman spectra of liquid bmimCl show that two rotational isomers (*trans-trans* and *gauche-trans* forms) of the butyl group of the bmim<sup>+</sup> cation co-exist in the liquid state and that they are likely to be included in two distinct local structures that can only be interconverted with an unusually long equilibration time [2]. A wide-angle X-ray scattering experiment [6] on liquid bmimCl shows a local ordering of the iodide ions having a spacing that is close to those found in the crystal structures of bmimCl and bmimBr [3]–[5]. If such structural orderings are significantly extensive, they may well cause extraordinary physical properties unique to ILs. For instance, if magnetic anions are aligned locally in an IL, it would show interesting magnetic properties.

In a previous paper, we reported that a RIL, 1-butyl-3-methylimidazolium tetrachloroferrate (bmim[FeCl<sub>4</sub>]), exhibited an unexpectedly strong response to a magnet [7]. This liquid contained in a glass sample tube was attracted strongly by a small Nd magnet (0.55 T) with its shape highly distorted to a concave shape (Fig. 1). In order to gain more insight into this newly found phenomenon, we here extend our study to another RIL, 1-butyronitrile-3-methylimidazolium tetrachloroferrate (nbmim[FeCl<sub>4</sub>]). Nbmim[FeCl<sub>4</sub>] is obtained by replacing the butyl group of the bmim<sup>+</sup> cation with the butyronitrile group.

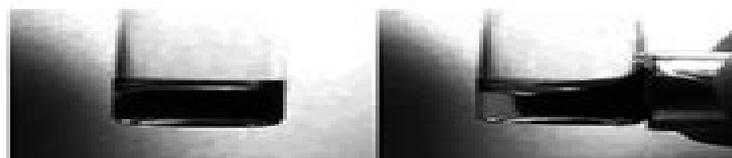


Fig. 1. Pictures showing the response of bmim[FeCl<sub>4</sub>] to a small Nd magnet (0.55 T) [7]. In order to show the displacement and distortion of bmim[FeCl<sub>4</sub>] more clearly, water is added to the sample liquid. Left: two layers of liquids, water (upper) and bmim[FeCl<sub>4</sub>] (lower), are seen when no magnetic field is applied. Right: bmim[FeCl<sub>4</sub>] is attracted and moves towards the magnet and the liquid is distorted to a concave shape.

## II. PREPARATION AND CHARACTERIZATION OF nbmim[FeCl<sub>4</sub>]

The preparation and characterization of nbmim[FeCl<sub>4</sub>] was carried out in a similar way as bmim[FeCl<sub>4</sub>] [7]. 1-butyronitrile-3-methylimidazolium chloride, nbmimCl, was synthesized from redistilled N-methylimidazole (Aldrich Chemicals), and 1-chlorobutyronitrile (Wako Chemicals) and purified with repeated recrystallization from dry acetonitrile. Nbmim[FeCl<sub>4</sub>] was obtained by a solid-phase reaction of crystalline nbmimCl with FeCl<sub>3</sub>·6H<sub>2</sub>O. The resultant liquid was separated from water and dried under vacuum. Thus prepared nbmim[FeCl<sub>4</sub>] was characterized by Raman and visible absorption spectroscopy. Raman spectra were recorded on a laboratory made near-infrared multi-channel Raman system. The 1064 nm line of a Nd:YAG laser was used as the excitation source in order to avoid fluorescence from impurities. As shown in Fig. 2, the observed Raman spectrum of the nbmim[FeCl<sub>4</sub>] shows the same pattern as that of nbmimCl, indicating that the nbmim<sup>+</sup> cation dominantly exists. The strong band assigned to the totally symmetric stretch mode of FeCl<sub>4</sub><sup>-</sup> [8] is observed at 330 cm<sup>-1</sup> only in the spectrum of the nbmim[FeCl<sub>4</sub>].

The visible absorption spectrum was recorded on a Hitachi U-3500 UV-VIS spectrometer. The visible absorption spectrum of the nbmim[FeCl<sub>4</sub>] is compared with that of bmim[FeCl<sub>4</sub>] in Fig. 3. The two absorption spectra resemble each other and show three characteristic bands of the [FeCl<sub>4</sub>]<sup>-</sup> ion [9] at 534, 619,

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