



The new evolution of protic ionic liquids: Antielectrostatic activity correlated with their surface properties



Joanna Feder-Kubis^a, Małgorzata Musiał^b, Marzena Dzida^b,
Monika Geppert-Rybczyńska^{b,*}

^a Faculty of Chemistry, Wrocław University of Science and Technology, Wybrzeże Wyspiańskiego 27, 50-370 Wrocław, Poland

^b Institute of Chemistry, University of Silesia, Szkolna 9, 40-006 Katowice, Poland

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ABSTRACT

The primary aim of this study was to explain the antielectrostatic activity of a series of protic 3-(alkoxymethyl)-1*H*-imidazol-3-ium salicylate ionic liquids as a function of their surface properties. The surface tension and contact angle on chosen surfaces were measured and discussed. The results were investigated as a function of the alkyl chain length of the alkoxymethyl substituent in the imidazolium cation. Despite the complex structure of the ions, the estimated surface tension was correlated with the corresponding experimental results. It was found that the alkyl chain length has a significant impact on the surface properties and wetting abilities of salicylates.

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Introduction

Since the 1990s, when interest in ionic liquids (ILs) was piqued [1,2], the general picture of this class of compound has been evaluated based from more-or-less accidental physicochemical characteristics to intentional investigations of the properties of selected compounds.

Nowadays, the unusual properties of ILs are thought of as attractive if they can be turned into concrete applications. The term “task-specific ionic liquids” is not new, but it refers to the next generations of materials that are being synthesized for use in special projects (e.g., catalysis, synthesis, gas absorption, analysis, lubricants, antiwear properties, medicine, pharmacology, electrochemistry, and energetic materials) [2–4]. However, in order to determine new applications it is necessary to reveal the physicochemical properties of a given material.

The surface tension of ILs has been frequently investigated [5]. Some examples of interfacial tension in mixtures of ILs with alkanols and alkanes (when the miscibility gap occurs) are also known [6–9]. These parameters are necessary to use ILs as solvents in synthesis [10,11] or for constructing the proper liquid-liquid

system for extraction [12–14]. On the other hand, contact angle measurements of ILs are very limited and are largely available only for commercial ILs (e.g., bis(trifluoromethylsulfonyl)imides [NTf₂⁻], tetrafluoroborates [BF₄⁻], hexafluorophosphates [PF₆⁻], ethyl sulfates [EtSO₄⁻] or dicyanamides [N(CN)₂⁻]) and surfaces such as glass, poly(tetrafluoroethylene) (PTFE), or silicone, or modified Si materials [15–17]. As a matter of fact, contact angles can be useful especially in heterogeneous systems or transport processes in capillary and porous surfaces, but there have been no attempts to correlate this quantity with such property, as is done in this work.

Here, this manuscript discusses one of the series of protic ILs with a salicylate anion, namely 3-(alkoxymethyl)-1*H*-imidazol-3-ium salicylates, [H-im-C₁OC_{*n*}][Sal]. The syntheses and thermal properties of considered salts have been previously described in the literature [18,19]. Another study reported the excellent results of 3-(alkoxymethyl)-1*H*-imidazol-3-ium salicylates as wood-preservative agents [20], where the salicylate ILs penetrate the wood surface and act effectively against fungi. In the same study, the authors noted the antielectrostatic effect of salicylates on some surfaces. This aspect seems to be extremely interesting, and our work accordingly focuses on it more specifically.

Thus, some ILs have been shown to be effective antielectrostatic agents [21–24]. For instance, a series of imidazolium ILs containing

* Corresponding author.

E-mail address: mong@o2.pl (M. Geppert-Rybczyńska).

alkoxymethyl substituent were tested by Pernak et al. to determine their capacity to drain a surface electric charge [21]. These authors found that the type of anion strongly influenced the strength of the resulting antielectrostatic effect.

This study presents not only the antielectrostatic activity of 3-(alkoxymethyl)-1*H*-imidazol-3-ium salicylates but also their surface and wetting behavior. The goal of this work was to determine a relation between these two kinds of properties. In particular, our investigation focuses on the influence of the alkyl chain length of the alkoxymethyl substituent in the imidazolium ring on these properties. From the point of view of applications, this question may be important since it allows for a reduction in the effort necessary to select the best candidate to improve the antielectrostatic properties of any material after simple surface tension and contact angle experiments. Despite the fact that the dependence between antielectrostatic activity and surface properties appears to be interesting, this subject has not been researched by other scientists yet.

Experimental

Chemicals, materials and synthesis

The syntheses and thermal properties of 3-(alkoxymethyl)-1*H*-imidazol-3-ium salicylate have been described in a previous publication [19]. Additional information about the material used for synthesis and preparation and the structure and purity of all of the investigated substances confirmed by spectral analysis (Table S.1) are collected in the Supporting Information.

Prior to use, all of the chemicals were carefully dried under vacuum for approximately 24–48 h with heating ($T = 333$ K). The drying procedure was maintained until the water content, detected using a TitroLine 7500 Karl Fischer trace coulometer (SI Analytics GmbH, Germany), was $w_{\text{H}_2\text{O}} \cong (3\text{--}6) \cdot 10^{-4}$. The calibration and characteristics of the work of the coulometric Karl Fischer titrator have been already described by Skowronek et al. [25]. The samples were stored in an argon atmosphere in vessels closed with septa. Immediately prior to the experiments, the samples were degassed by heating them up to 323 K. The results of the water determinations of the samples after drying are listed in Table 1 together with the molar masses of the investigated ILs.

Higher homologues of [*H*-im- C_1OC_n][Sal] with $n = 9\text{--}10$, as well as higher-melting-point polymorphic types of [*H*-im- C_1OC_{11}][Sal],

have a T_m of at least at 295 K [19]. Therefore, these compounds were measured in a super-cooled state.

The materials that were used for the antielectrostatic investigation included polyethylene (LDPE) film (POLITEN II/003/GO produced by Chemical Institute Blachownia in Kędzierzyn-Koźle, Poland) and polypropylene (PP) nonwoven fabric (WIGOFIL/MALEN P F-401 produced by the Wigolen joint-stock company in Częstochowa, Poland). These materials were free of lubricants and antioxidants. Disks 12.5 mm in diameter were cut from the LDPE and PP and then washed in acetone and dried by placing them in an air-conditioned room. A disk was rubbed with a cotton swab soaked with a 0.5% chloroform solution of each of the salts that were studied. Next, each disk was hung so that the chloroform could evaporate spontaneously. The disks, covered with protic ILs, were stored for 24 h in an air-conditioned room at 294 ± 2 K and a relative humidity of $55 \pm 5\%$.

The materials used for contact angles measurements included PTFE film with a thickness of 0.1 mm and a density of 0.35 g cm^{-3} (Bisan LLC, Poland) and clear glass PP with a thickness of 0.04 mm (Office Products, Poland). The substrates were only rinsed with distilled water and dried; they were then stored in an atmosphere with the same humidity as was used for the contact angle measurements (*i.e.*, around 40%).

Antielectrostatic properties

The surface resistance R_s (Ω), half charge decay time $\tau_{1/2}$ (s) and maximum voltage U_{ind} (V) induced on the disk surface of the polymers were determined. The surface resistance was measured using a Tralin III Statron electrometer, and the maximum voltage induced with a Type V 531 digital voltmeter (Meratronik, Poland). This apparatus and measurement procedure have been previously described in the literature [21]. The experiment was performed in duplicate. In each test, the surface resistance was measured at least three times on each side of the disk, and the other parameters (*i.e.*, the half charge decay time and the maximum voltage) were measured twice. The relative error in the determination of these three quantities did not exceed 5%.

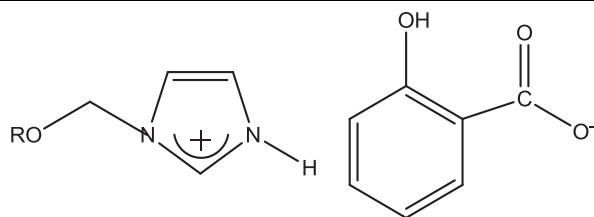
Density, surface tension and contact angles measurements

The density, ρ , necessary for carrying out the surface tension measurements according to the pendant drop method, was

Table 1

List of 3-(alkoxymethyl)-1*H*-imidazol-3-ium salicylate, [*H*-im- C_1OC_n][Sal], ILs with $n = 3\text{--}11$, molar masses, and initial and final mass fraction water content based on coulometric Karl Fisher titration.

IL	R	$M \cdot 10^{-3}$ (mol kg $^{-1}$)	$w_{\text{H}_2\text{O}} \cdot 10^4$ (initial)	$w_{\text{H}_2\text{O}} \cdot 10^4$ (final)
[<i>H</i> -im- C_1OC_n][Sal]				
[<i>H</i> -im- C_1OC_3][Sal]	C_3H_7	278.30	98.6	6.0
[<i>H</i> -im- C_1OC_4][Sal]	C_4H_9	292.33	6.1	5.7
[<i>H</i> -im- C_1OC_5][Sal]	C_5H_{11}	306.36	191.5	6.3
[<i>H</i> -im- C_1OC_6][Sal]	C_6H_{13}	320.38	81.1	5.4
[<i>H</i> -im- C_1OC_7][Sal]	C_7H_{15}	334.41	87.6	4.7
[<i>H</i> -im- C_1OC_8][Sal]	C_8H_{17}	348.44	29.9	6.3
[<i>H</i> -im- C_1OC_9][Sal]	C_9H_{19}	362.46	154.9	5.5
[<i>H</i> -im- C_1OC_{10}][Sal]	$\text{C}_{10}\text{H}_{21}$	376.49	41.5	4.9
[<i>H</i> -im- C_1OC_{11}][Sal]	$\text{C}_{11}\text{H}_{23}$	390.52	77.0	3.1



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