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DESIGN OF SALICYLATE-BASED IONIC LIQUID AQUEOUS BIPHASIC SYSTEMS FOR THE EXTRACTION OF A SYNTHETIC FOOD COLORANT E133

J. Mušović, D. Tekić, A. Jocić, S. Marić and A. Dimitrijević

Vinča Institute of Nuclear Sciences – National Institute of the Republic of Serbia, University of Belgrade, Mike Petrovića Alasa 12-14, 11351 Belgrade, Serbia (jasmina.musovic@vin.bg.ac.rs)

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ABSTRACT

Food colorants play an important role in the food industry; however, their accurate determination, separation, and control are essential to ensure food quality and safety. Brilliant Blue FCF (E133) is a synthetic dye widely used in food products due to its intense color and high stability. In this study, ionic liquid-based aqueous biphasic systems (ABSs) composed of salicylate-based ionic liquids (ILs) and potassium carbonate (K_2CO_3) were investigated as alternative extraction platforms for the separation of E133. Initially, phase diagrams were constructed to evaluate the biphasic behavior of the investigated systems and to assess the influence of IL cation structure on phase formation. Subsequently, the extraction performance of selected IL-ABSs toward E133 was evaluated. The tested systems demonstrated excellent extraction efficiency, reaching 99.48% and 99.55%. These findings highlight the potential of salicylate-based IL-ABSs as efficient, tunable, and sustainable platforms for the extraction and separation of food-related compounds.

INTRODUCTION

Synthetic food colorants are widely used in the food industry to enhance the appearance and consumer acceptance of various food products. Among them, Brilliant Blue FCF (E133) is one of the most commonly applied water-soluble dyes due to its intense color, high stability, and compatibility with different food matrices [1]. However, the increasing use of synthetic colorants has highlighted the importance of reliable separation and extraction approaches for their determination, purification, and potential recovery from complex systems, contributing to improved food quality and safety.

In recent years, ILs have emerged as promising compounds for the development of sustainable separation processes due to their negligible vapor pressure, high chemical stability, and highly tunable physicochemical properties. Their ability to establish specific interactions with target molecules, including

hydrogen bonding, electrostatic interactions, and π-π interactions, enables their application as efficient extraction media.

Among different IL-based separation approaches, ABSs have gained considerable attention as environmentally favorable platforms for the extraction and separation of various compounds. Aqueous biphasic systems are liquid-liquid separation systems composed mainly of water and two phase-forming components, which, under appropriate concentrations and conditions, spontaneously separate into two distinct aqueous phases [2]. Due to differences in the affinity of solutes toward these phases, target compounds can be selectively distributed between them, enabling efficient extraction and separation. By incorporating ionic liquids as one of the phase-forming components, IL-ABSs combine the advantages of aqueous biphasic media with the tunability of ILs, allowing the selective partitioning of target molecules between coexisting aqueous phases.

In this study, aqueous biphasic systems composed of salicylate-based ILs (tetrabytilphosphonium - TBP and tetrabutylammonium - TBA) and K_2CO_3 were investigated for the extraction of E133. Initially, phase diagrams were constructed to evaluate the phase-forming ability of the investigated systems and to determine the influence of IL cation structure on biphasic behavior. Subsequently, selected ABSs were applied for the extraction of E133. The obtained results demonstrate the effectiveness of salicylate-based IL-ABSs for the extraction of E133 and highlight the importance of IL structure in designing efficient dye separation systems.

EXPERIMENTAL

Salicylate-based ILs were prepared through a neutralization reaction between the corresponding hydroxide precursors, tetrabutylphosphonium hydroxide or tetrabutylammonium hydroxide (40 wt% aqueous solutions, Sigma-Aldrich, USA), and salicylic acid (≥99% purity, Sigma-Aldrich, USA). The reactants were combined in an equimolar ratio, and the resulting mixtures were subjected to continuous stirring at ambient conditions to promote complete reaction. After the reaction, water was removed from the obtained mixtures under reduced pressure using a rotary evaporator (R-210 Rotavapor System, Büchi Labortechnik AG, Switzerland) to obtain the desired ILs.

Phase diagrams, representing the mass fraction of IL as a function of the mass fraction of potassium carbonate, were determined using the cloud point titration method. Aqueous mixtures of known amounts of IL and potassium carbonate were prepared and titrated with water until a clear single-phase system was obtained, followed by titration with potassium carbonate solution until turbidity appeared, indicating the onset of phase separation. The compositions at the cloud point were recorded and used to construct the phase diagrams.

The concentration of E133 was quantified using a LLG-uniSPEC 2 UV-Vis spectrophotometer (LLG Labware, Germany) by measuring the absorbance at 625 nm.

RESULTS AND DISCUSSION

Phase diagrams (Figure 1.) were determined to evaluate the phase-forming ability of the investigated ABSs with potassium carbonate. Both ILs readily formed biphasic systems; however, [TBP][Sal] showed a stronger salting-out tendency than [TBA][Sal], as indicated by its binodal curve being closer to the coordinate origin. This behavior can be attributed to the higher hydrophobicity of the $[TBP]^+$ cation compared with $[TBA]^+$, resulting from the larger atomic radius and lower electronegativity of phosphorus. The more diffuse charge distribution of the phosphonium cation reduces its hydration ability, facilitating IL salting-out and phase separation at lower component concentrations [3].

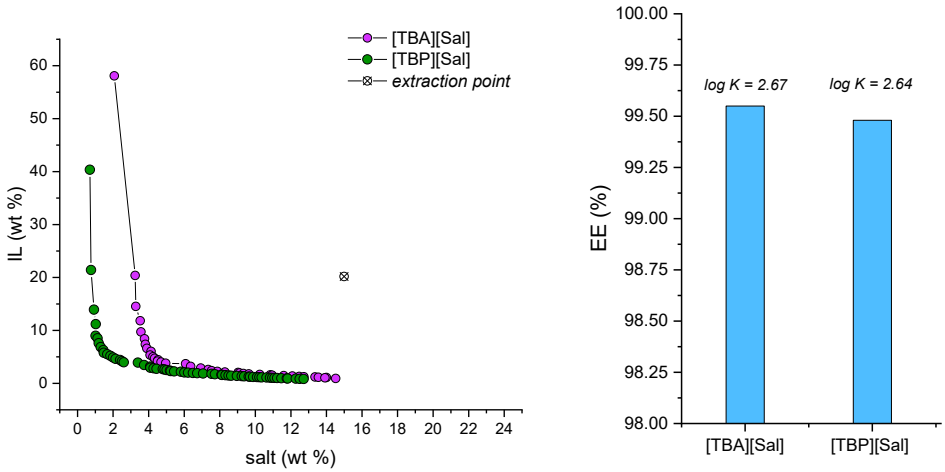


Figure 1. Phase diagrams for [TBP][Sal]/ [TBA][Sal] + K_2CO_3 at ambient conditions and extraction efficiencies of E133 obtained for [TBP][Sal]/ [TBA][Sal] + K_2CO_3 systems.

An extraction point within the biphasic region, consisting of 20 wt% IL and 15 wt% potassium carbonate, was selected for further investigation. ABSs with this composition were prepared and evaluated for the extraction of E133 dye. The obtained parameters are presented in Figure 1.

The obtained results showed that E133 dye migrated almost completely into the IL-rich phase, indicating a strong affinity of the dye toward the IL phase. This behavior can be attributed to the presence of specific interactions between the dye molecules and ILs. Consequently, high extraction efficiencies of 99.48% and 99.55% were achieved, with corresponding distribution coefficients (log K) of 2.67 and 2.64 for the ABSs containing [TBP][Sal] and [TBA][Sal], respectively.

CONCLUSION

Salicylate-based IL-ABSs combined with K_2CO_3 demonstrated excellent capability for the extraction of E133. Both investigated systems enabled efficient phase separation and almost complete dye transfer into the IL-rich phase, achieving extraction efficiencies above 99%. The higher hydrophobicity of [TBP][Sal] resulted in a slightly enhanced salting-out tendency compared with [TBA][Sal], highlighting the role of IL cation structure in tuning ABS performance. These results confirm the potential of salicylate-based IL-ABSs as efficient and sustainable platforms for the extraction of food colorants.

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