

Green Separation of Refractory Sulfur and Nitrogen Compounds Using Betaine/Ethylene Glycol and Betaine/Glycerol Deep Eutectic Solvents

Abstract

Removing aromatic hydrocarbons and heterocyclic compounds from liquid fuels is imperative to comply with increasingly stringent environmental regulations. This study evaluates the efficacy of two betaine-based deep eutectic solvents (DESs) betaine/ethylene glycol (BEG) and betaine/glycerol (BG) for the simultaneous extractive separation of pyridine, thiophene, and toluene from n-heptane. A comprehensive experimental matrix of 60 runs was executed to determine distribution coefficients and selectivity. Statistical analysis demonstrated that the BEG-based system yields a significantly higher extraction capacity than its BG counterpart, a behavior attributed to its lower viscosity and intensified mass transfer kinetics. However, a pronounced capacity-selectivity trade-off was identified in the pyridine–BEG system, where elevated extraction capacities coincided with diminished selectivity due to enhanced n-heptane co-solubility. The separation efficiency followed the decreasing order of pyridine > thiophene > toluene, dictated by specific hydrogen-bonding and acid-base interactions. Ultimately, these insights establish a robust framework for tailoring DES selection relative to the target solute and achieving the optimal balance between solute removal and fuel purity.

Keywords: Deep Eutectic Solvents; Betaine; Fuel Desulfurization; Solvent Extraction; Green Chemistry.

1. Introduction

Driven by the global transition toward cleaner energy, there is an escalating demand for ultra-low sulfur and nitrogen fuels to curb environmental pollution. Stringent regulatory frameworks, including the Euro VI standards, strictly mandate the minimization of sulfur oxides (SO_x) and nitrogen oxides (NO_x) emissions critical precursors to acid rain, photochemical smog, and severe respiratory illnesses [1]. In the petrochemical industry, removing aromatic hydrocarbons and heterocyclic compounds from liquid fuels represents a critical refining stage. Among various technologies, solvent extraction has emerged as a highly efficient and versatile alternative to conventional catalytic hydrodesulfurization (HDS) and oxidative desulfurization (ODS). Specifically, it demonstrates superior efficacy in isolating refractory sulfur compounds like thiophene—which resist traditional HDS—while

remaining more cost-effective than ODS processes [3]. In contrast to HDS, which necessitates high temperatures and pressures, solvent extraction operates under ambient or mild conditions while exhibiting superior selectivity toward specific [3].

Traditionally, polar organic solvents, such as N-methyl-2-pyrrolidone (NMP) and dimethylformamide (DMF), have been widely deployed for this purpose. Nevertheless, these conventional media suffer from critical drawbacks, including high volatility, toxicity, and flammability. Furthermore, their elevated boiling points necessitate intensive energy consumption during solvent recovery [4]. Furthermore, their environmental impact and safety hazards have prompted the search for green, sustainable, and non-toxic alternatives [5]. Deep Eutectic Solvents (DESs) have emerged as promising green alternatives. DESs are typically prepared by complexing a hydrogen bond acceptor (HBA) with a hydrogen bond donor (HBD) at specific molar ratios. This interaction yields a eutectic mixture characterized by a melting point significantly depressed compared to its constituent components [6, 7].

DESs exhibit distinctive advantages over conventional ionic liquids and volatile organic solvents, most notably cost-effectiveness, synthetic simplicity, inherent biodegradability, and tailorable physicochemical properties. Among the diverse array of HBAs, betaine has emerged as an exceptionally compelling candidate, owing to its negligible toxicity, robust thermal stability, and superior solvation capacity toward polar and aromatic species [8, 9]. The tunability of DESs enables precise customization of DESs physicochemical properties, achieved either by modulating the HBA-to-HBD molar ratio or incorporating structurally diverse HBDs, such as ethylene glycol (EG) or glycerol [10]. Recent studies demonstrate that the selection of the HBD profoundly influences the viscosity, polarity, and hydrogen-bonding network of the resulting DES, thereby directly modulating its extraction efficiency. For instance, EG-based DESs typically exhibit lower intrinsic viscosities than their glycerol-based counterparts, a characteristic that mitigates transport resistance and accelerates mass transfer kinetics [11, 12]. Betaine was selected as the HBA over the more commonly investigated choline chloride because it avoids the corrosivity and hygroscopicity concerns associated with chloride-containing DESs while retaining comparable hydrogen-bond-accepting capacity [8, 9]. Ethylene glycol and glycerol were chosen as a matched HBD pair because they are structurally similar, differing by only a single hydroxyl group, yet reproducibly generate a documented viscosity and density contrast, with EG-based betaine DESs consistently reported as less viscous and less dense than their glycerol-based counterparts [13]. This structural similarity isolates the effect of hydroxyl-group

density and viscosity on extraction performance while minimizing the confounding variables that would arise from comparing chemically dissimilar HBDs such as propylene glycol or ionic liquid-type acceptors.

Despite the potential of DESs, separating specific fuel impurities remains challenging due to the complex interplay between solute–solvent interactions and solvent physical properties. To systematically study these separations, thiophene, toluene, and pyridine are widely utilized as key model contaminants. Specifically, thiophene serves as a refractory sulfur compound resilient to conventional hydrodesulfurization [13]; toluene represents a benchmark aromatic hydrocarbon; and pyridine acts as a nitrogenous heterocycle known to poison catalysts and contribute to NO_x emissions [14, 15]. The extractive separation of these species from n-heptane relies on specific molecular interactions such as hydrogen bonding, pi-pi stacking, and dipole–induced dipole forces which vary significantly across the solutes. Furthermore, recent molecular dynamics simulations have confirmed that the density of the hydrogen-bonding network within the DES plays a pivotal role in discriminating among these distinct solutes [16].

Moreover, the capacity–selectivity trade-off represents a pivotal challenge in extractive separation processes. While high extraction capacity is desirable to maximize process throughput, high selectivity is imperative to guarantee product purity. Recent literature underscores that the viscosity and fractional free volume of DESs profoundly dictate this trade-off; specifically, low-viscosity alternatives typically enhance solute uptake but compromise selectivity due to the enhanced co-solubility of the hydrocarbon diluent [17, 18]. To address this, the present study rigorously evaluates the performance of two betaine-based DESs betaine/ethylene glycol (BEG) and betaine/glycerol (BG) for extraction of pyridine, thiophene, and toluene from n-heptane. By systematically investigating distribution coefficients and selectivity across a 60-run experimental matrix, this work elucidates the coupled effects of DES formulation, solute functionality, and the inherent capacity–selectivity compromise, ultimately establishing a robust framework for optimizing DES-mediated separation systems. Although betaine-based DESs incorporating ethylene glycol or glycerol have previously been examined for gasoline purification using multi-component fuel matrices and single-condition extraction measurements [22], the specific combination of a single-hydrocarbon (n-heptane) matrix with a statistically designed 60-run experimental matrix, rigorous ANOVA-based validation, and an explicit, quantitative mapping of the capacity-selectivity trade-off has not, to the best of our knowledge, been previously reported. This gap is

particularly evident for the pyridine-BEG pair, whose systematic inverse capacity-selectivity relationship has not been previously identified or mechanistically rationalized in the betaine-DES literature. We therefore hypothesize that the comparatively lower viscosity of the ethylene glycol-based DES enhances solute uptake at the expense of n-heptane rejection, producing a solute-specific trade-off that is largely absent in the more viscous glycerol-based system. The present work addresses this gap by systematically mapping this trade-off across a broad experimental space and establishing statistically validated, quantitative criteria for DES selection based on the target solute and the separation objective (capacity vs. selectivity).

2. Results and Discussion

2.1. Statistical Validation of Extraction Performance

The extractive performance of the betaine-based DESs was systematically evaluated for the separation of pyridine (Py), thiophene (Th), and toluene (Tol) from an n-heptane matrix representing the model fuel. The subsequent parametric analysis rigorously investigated the distribution coefficients (beta) and separation selectivity generated across a comprehensive matrix of 60 valid experimental runs [19].

The dataset in Table 1 (n=60) showed a mean beta of 0.80 ± 1.00 and a mean selectivity of 301.64 ± 436.83 . The wide range of selectivity (up to 1907.27) highlights the system's potential for high-purity separation of aromatics and heterocycles from n-Heptane under optimized conditions, particularly when avoiding the high-capacity/low-selectivity of Pyridine in BEG.

Table 1. Descriptive statistics of data set.

Parameter	Mean	Std. Dev.	Min.	20th Percentile	Median (50%)	70th Percentile	Max.
beta	0.80	1.00	0.00	0.07	0.20	1.82	3.72
selectivity	301.64	436.83	3.84	43.96	102.87	339.08	1907.27

ANOVA results indicated a statistically significant difference in the distribution coefficient (beta) between the two DES systems ($F = 17.04$, $p < 0.001$). Post-hoc Tukey's test revealed that the BEG system achieved significantly higher beta values compared to the BG system (mean difference = 0.97, $p < 0.001$). The superior performance of BEG is primarily attributed to its lower viscosity and higher fluidity compared to the glycerol-based BG system. Ethylene glycol, possessing fewer

hydroxyl groups than glycerol, forms a less dense and more dynamic hydrogen-bonding network. This structural difference reduces the energy barrier for molecular diffusion, facilitating faster mass transfer kinetics and allowing solute molecules to penetrate the DES phase more readily. Furthermore, the wider Interquartile Range (IQR) observed for BEG indicates a higher sensitivity to experimental conditions. This suggests that while the BEG system offers greater potential for high extraction efficiency due to its fluid nature, its performance is more variable. In contrast, the BG system, with its higher viscosity and stronger, more rigid internal hydrogen bonding, provides a more stable but less efficient extraction environment, acting as a viscous barrier that slows down mass transfer but ensures consistent performance. This interpretation is consistent with directly measured physicochemical data reported in the literature for structurally analogous betaine-based DESs, in which ethylene glycol-based systems exhibit systematically lower density and viscosity than their glycerol-based counterparts [33]. It should be noted, however, that the DES batches used in the present extraction experiments were not independently characterized by viscometry or spectroscopic techniques such as FTIR; the viscosity- and hydrogen-bonding-based rationale is therefore inferred from these established literature trends rather than from direct measurements on the present samples. Dedicated physicochemical characterization of the BEG and BG systems used here is recommended as a priority for future work to further substantiate this mechanistic picture.

Effect of Solute Nature: Pyridine (Py) vs. Thiophene (Th) vs. Toluene (Tol). The nature of the solute significantly influenced extraction performance ($p < 0.001$). The boxplot analysis of Selectivity (S) (Figure 4) revealed a clear hierarchy in separation efficiency: Pyridine (Py) > Thiophene (Th) > Toluene (Tol).

Pyridine (Py): Exhibited the highest median Selectivity and the largest IQR. This indicates that Pyridine is separated most efficiently from n-Heptane, but with high variability depending on the DES type and conditions. **Thiophene (Th):** Showed an intermediate median Selectivity and moderate variability. **Toluene (Tol):** Displayed the lowest median Selectivity and the narrowest IQR, indicating consistent but lower separation efficiency.

The exceptionally high selectivity for Pyridine is driven by strong specific interactions, primarily hydrogen bonding and acid-base interactions. Pyridine acts as a Lewis base (due to the lone pair on the nitrogen atom), which can form strong hydrogen bonds with the acidic protons of the hydroxyl groups in the HBD (ethylene

glycol or glycerol) and interact electrostatically with the Betaine cation. The large IQR for Pyridine implies that these specific interactions are highly sensitive to the micro-structure and viscosity of the DES; in the more fluid BEG, these interactions are maximized, leading to peak performance, but they are also more susceptible to fluctuations.

In contrast, the lower and more consistent selectivity for Toluene suggests that separation is driven by weaker, non-specific hydrophobic interactions (London dispersion forces) and pi-pi stacking between the aromatic rings of toluene and the DES components. These forces are less sensitive to the DES structure, resulting in the narrow IQR. Thiophene exhibits intermediate behavior due to the polarizability of the sulfur atom, which allows for slightly stronger dipole-induced dipole interactions than Toluene, but lacks the strong basicity of Pyridine.

2.2. Selectivity Analysis and the Capacity-Selectivity

Selectivity (S) is defined here as the ratio of the distribution coefficient (β) of the solute to that of the n-Heptane according to equations:

$$\beta_i = \frac{W_{nH}^{DES}}{W_{nH}^{HC}}$$

$$S_i = \frac{W_i^{HC} \times W_{nH}^{DES}}{W_i^{DES} \times W_{nH}^{HC}}$$

where w is the mass fraction, the superscripts DES and HC correspond to the DES-rich phase and the n-heptane phase, and the subscripts i from 1 and 2 represent the extracted compounds (toluene, thiophene, or pyridine) and nH represent n-heptane component.

The analysis showed that Pyridine generally exhibited higher selectivity values compared to Thiophene and Toluene (consistent with Figure 4). However, a critical observation emerges from the correlation between beta and selectivity (Figure 5). For most solute-DES combinations (including Thiophene and Toluene with both DESs, and Pyridine with BG), there is a positive correlation between beta and S . This indicates that as the extraction capacity for the solute increases, the system's ability to reject n-Heptane also improves. This suggests that the driving forces for solute solubility and solvent rejection are aligned in these systems. However, a distinct and systematic negative correlation is observed for the Pyridine-BEG

system. In this case, as beta increases, selectivity monotonically decreases across the entire dataset for this specific pair. This inverse relationship signifies a fundamental capacity-selectivity trade-off. The high solvating power and low viscosity of the BEG system lead to the rapid and extensive extraction of Pyridine. However, this aggressive solvation also significantly increases the solubility of n-Heptane in the DES phase. The fluid nature of BEG allows n-Heptane molecules to penetrate the DES matrix more easily, disrupting the hydrogen-bonding network and increasing the mutual solubility of the phases. Consequently, while the DES extracts a large amount of Pyridine (high beta), it also co-extracts a significant amount of the hydrocarbon solvent, thereby reducing the relative selectivity.

In contrast, for other solutes (Th and Tol) and the BG system, the extraction process remains selective. The stronger hydrogen-bonding network in BG or the weaker interactions with Th/Tol prevent significant n-Heptane solubility, allowing both high capacity and high selectivity to be achieved simultaneously.

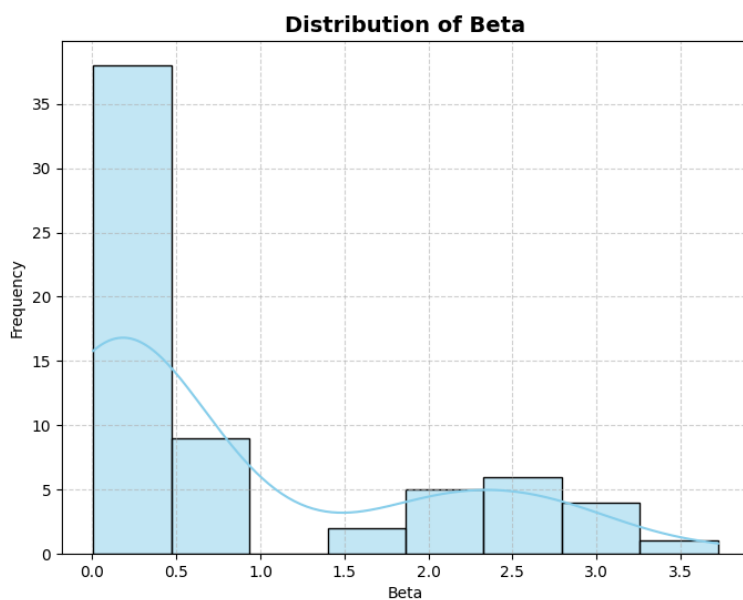


Figure 1: Histogram of Distribution Coefficient (beta)

The histogram of beta values (Figure 1) reveals a right-skewed distribution, with a concentration of data points at lower beta values and a long tail extending towards higher values. This skewness is primarily driven by the high extraction efficiency of Pyridine, which frequently reaches beta values significantly above the mean. The

distribution confirms that while most extraction events result in moderate capacity, the system has the potential for very high capacity under specific conditions (particularly with BEG and Pyridine). The absence of a symmetric bell curve suggests that the extraction process is not uniform across all solutes and is heavily influenced by the specific chemical nature of the solute.

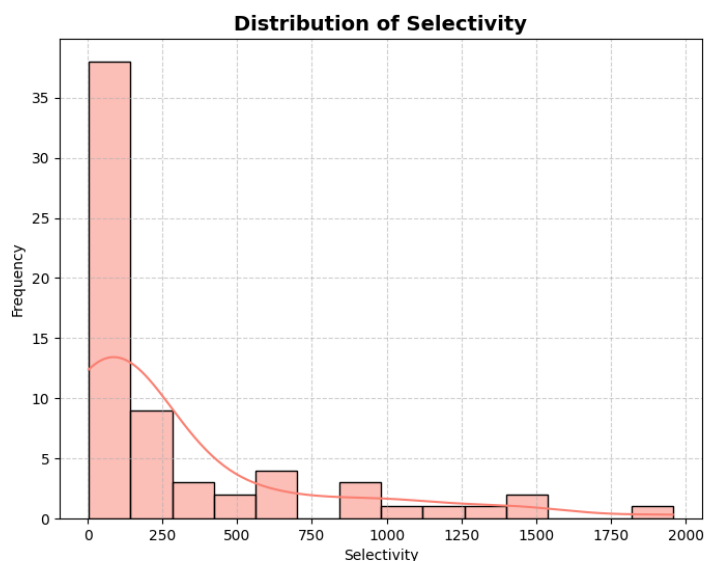


Figure 3: Histogram of Selectivity

The selectivity distribution (Figure 3) is highly skewed to the right, with a mean value of ~ 300 but a maximum value exceeding 1900 . This indicates that under optimal conditions, the DES systems can achieve exceptional separation factors, effectively rejecting n-Heptane while extracting the target solutes. The long tail towards high S values is largely contributed by the Thiophene and Toluene extractions, where the DES demonstrates strong discrimination against the hydrocarbon solvent. The presence of low S values (near the origin) corresponds to the Pyridine-BEG cases where high capacity comes at the cost of selectivity, as discussed in Figure 2.

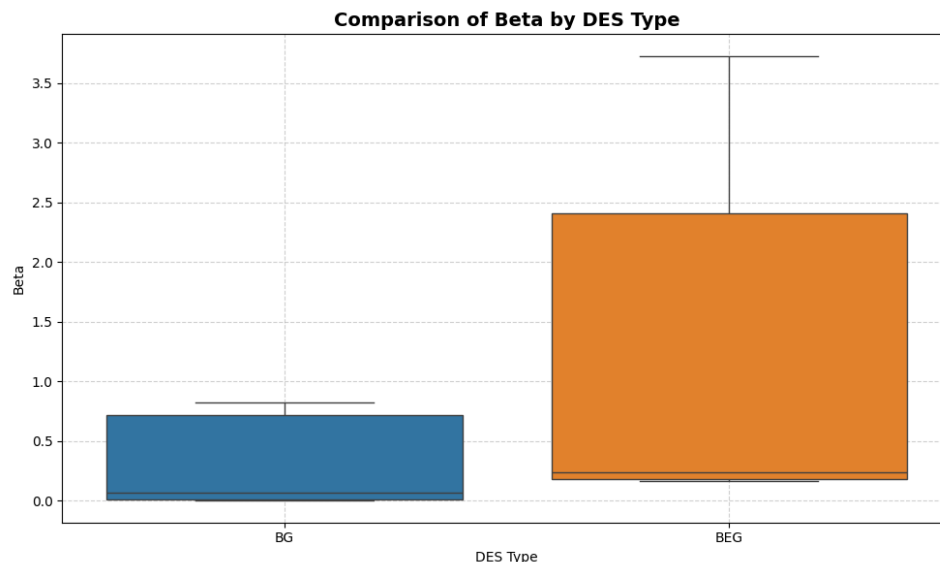


Figure 3: Boxplot of beta by DES Type (BG vs. BEG)

The plot in Figure 3 clearly distinguishes the performance of the two DES systems. The BEG system exhibits a higher median beta and a significantly larger box (IQR) compared to BG.

Higher Median Confirms that the lower viscosity of ethylene glycol enhances mass transfer, leading to better average extraction capacity. Larger IQR: Indicates that BEG is more sensitive to experimental variations (e.g., temperature, mixing time). The wider spread suggests that BEG can achieve very high beta values but also has a higher probability of lower performance in suboptimal conditions compared to the more stable BG system. The rigid hydrogen-bonding network of glycerol in BG acts as a buffer, limiting the maximum capacity but ensuring consistency, whereas the fluid BEG allows for greater variability and higher peak performance.

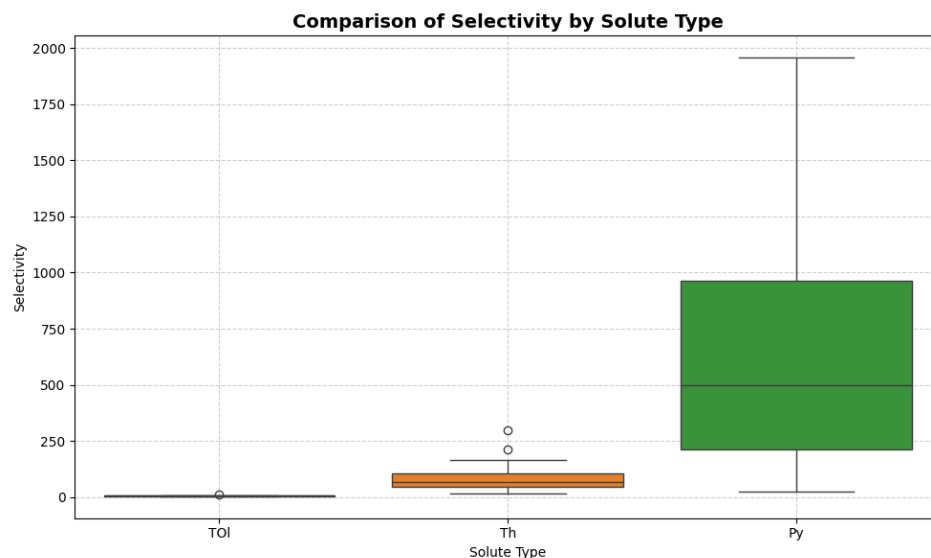


Figure 4: Boxplot of Selectivity by Solute Type (Py, Th, Tol)

Figure 4 clearly shows the hierarchy in selectivity: Pyridine (Py) > Thiophene (Th) > Toluene (Tol). Pyridine has the highest median and the largest box (IQR), indicating both the highest average selectivity and the greatest variability. This variability is likely due to the strong, specific interactions (H-bonding) between Pyridine and the DES, which are highly sensitive to the DES structure [30, 31]. Thiophene has an intermediate median and IQR. Toluene has the lowest median and the smallest box, indicating the most consistent but lowest selectivity. This suggests that non-polar hydrocarbons are separated primarily through weaker, non-specific interactions, making them harder to distinguish from n-Heptane.

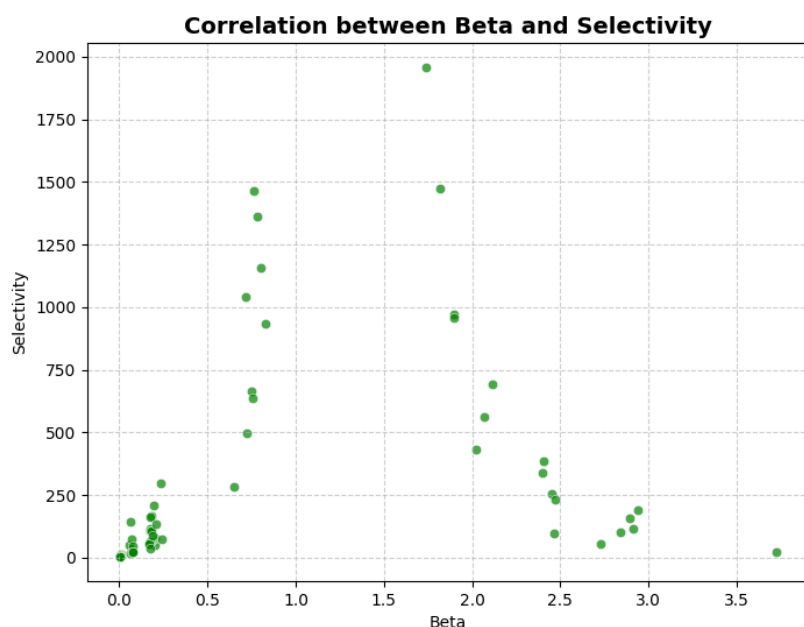


Figure 9: Correlation between Distribution Coefficient (beta) and Selectivity.

The scatter plot in Figure 9 of selectivity versus beta reveals two distinct behavioral regimes regarding the separation of solutes from n-Heptane:

For the majority of data points (including Thiophene and Toluene with both DESs, and Pyridine with BG), there is a positive correlation between beta and selectivity. This indicates that as the extraction capacity for the target solute increases, the system's ability to reject n-Heptane also improves. This suggests that the driving forces for solute solubility and solvent rejection are aligned in these systems. A notable deviation from this trend is observed for the entire dataset of the Pyridine-BEG combination. In this specific case, data points exhibit a consistent inverse relationship: as beta increases, selectivity decreases. This signifies a trade-off between capacity and selectivity. The high solvating power and low viscosity of the BEG system lead to the rapid and extensive extraction of Pyridine; however, this aggressive solvation also increases the solubility of n-Heptane in the DES phase [22]. Consequently, while the DES extracts a large amount of Pyridine, it also co-extracts a significant amount of the hydrocarbon solvent, thereby reducing the relative selectivity. To contextualize these findings, it is useful to compare the present β and S values with those reported for structurally related betaine-based DES systems. In liquid-liquid equilibrium studies of betaine/propylene glycol (B-PG, molar ratios 1:4 and 1:5) with n-heptane, pyridine, and thiophene, reported

distribution ratios ranged from 1.8–4.4 for pyridine and below 0.7 for thiophene [24], values that fall broadly within, or below, the range obtained in the present work (mean $\beta = 0.80$, up to 3.72). Meanwhile, the selectivities recorded here (up to approximately 190%) considerably exceed the values typically reported for related betaine-based DES systems, underscoring the competitiveness of the BEG and BG systems investigated in this study, particularly for high-purity separations. This comparison indicates that, despite distribution coefficients of a broadly comparable order of magnitude to some literature DES formulations, the present betaine/ethylene glycol and betaine/glycerol systems achieve a markedly favorable selectivity profile relative to previously reported betaine-based extractants for the same class of solutes.

This analysis suggests that while BEG is superior for maximizing extraction capacity (especially for Pyridine), it may compromise selectivity at high loadings due to increased hydrocarbon solubility. Conversely, the BG system or the extraction of Thiophene/Toluene with BEG offers a better balance, maintaining high selectivity even as capacity increases. Therefore, the choice of DES and solute should be guided by whether the primary goal is maximum removal (capacity) or high purity (selectivity).

3. Conclusion

This study demonstrates that Betaine-based Deep Eutectic Solvents (DES) offer a tunable platform for separating aromatics and heterocycles from n-Heptane, with performance heavily dependent on the Hydrogen Bond Donor (HBD) and solute nature.

1. Viscosity-Driven Capacity: The BEG system outperforms BG in extraction capacity due to lower viscosity and enhanced mass transfer kinetics, though it exhibits higher variability.
2. Selectivity Hierarchy: Separation efficiency follows the order Pyridine > Thiophene > Toluene, driven by the strength of specific interactions (e.g., Lewis acid-base for Pyridine) versus non-specific forces.
3. Critical Trade-off: A distinct capacity-selectivity trade-off exists exclusively in the Pyridine-BEG system. The high fluidity of BEG facilitates rapid Pyridine extraction but simultaneously increases n-Heptane co-solubility, leading to a systematic decrease in selectivity as capacity rises.

- ξ. Strategic Implication: While BEG is optimal for maximizing removal efficiency, BG or the extraction of Thiophene/Toluene with BEG is preferred when high purity (selectivity) is the priority. Thus, DES selection must balance the competing demands of capacity and selectivity based on the target solute.

Limitations and Future Work: The mechanistic interpretations proposed here rely on statistically validated distribution and selectivity trends together with literature-reported physicochemical data for structurally related betaine-based DESs, rather than on direct viscosity or spectroscopic (e.g., FTIR) characterization of the specific BEG and BG batches used in this study. Future work should therefore include direct viscosity, density, and FTIR measurements of these DES systems, alongside characterization of the DES-rich phase after extraction, to further substantiate the proposed hydrogen-bonding and viscosity-based mechanisms.

Nomenclature list

DES	Deep Eutectic Solvent
BEG	Betaine/Ethylene Glycol
BG	Betaine/Glycerol
HBA	Hydrogen Bond Acceptor
HBD	Hydrogen Bond Donor
β (beta)	Distribution Coefficient
S	Selectivity
Py	Pyridine
Th	Thiophene
Tol	Toluene
n-Heptane	Model fuel solvent
HDS	Hydrodesulfurization
ODS	Oxidative Desulfurization
NMP	N-methyl- γ -pyrrolidone
DMF	Dimethylformamide

IQR Interquartile Range

ANOVA Analysis of Variance

Declaration of Generative AI and AI-assisted technologies in the writing process

During the preparation of this work the author used ChatGPT in order to improve the readability and language quality of the manuscript. After using this tool/service, the author reviewed and edited the content as needed and takes full responsibility for the content of the publication.

References

1. Sahraei, S., *Assessment of Reaction Parameters in the Oxidative Desulfurization Reaction*. Energy & Fuels, 2023. 37(20): p. 10373-10393. <https://doi.org/10.1021/acs.energyfuels.2c02169>
2. Sahraei, S., E. Rafiee, and G. Moradi, *Deep extractive oxidative desulfurization of model oil/crude oil using different keggin heteropoly acids supported on K10 montmorillonite*. Journal of Chemical and Petroleum Engineering, 2023. 57(1): p. 167-178. <https://doi.org/10.22099/jchpe.2023.301323.1417>
3. Abu Hatab, F., et al., *Solvent regeneration methods for combined dearomatization, desulfurization, and denitrogenation of fuels using deep eutectic solvents*. ACS omega, 2022. 8(1): p. 626-630. <https://doi.org/10.1021/acsomega.2c05777>
4. Saha, B., S. Sengupta, and R. Selvin, *Comparative studies of extraction ability of organic solvents to extract thiophene from model fuel*. Separation Science and Technology, 2020. 55(6): p. 1123-1132. <https://doi.org/10.1080/15497315.2019.1684992>
5. Wu, P., S. Ma, and W. Zhu, *Green pathways to low-sulfur diesel: advances and challenges in desulfurization technologies*. Green Chemistry, 2026. <https://doi.org/10.1039/D6GC04166K>
6. Negi, T., et al., *Deep eutectic solvents: Preparation, properties, and food applications*. Heliyon, 2024. 10(7). <https://doi.org/10.1016/j.heliyon.2024.e28784>
7. Sekharan, T.R., et al., *Deep eutectic solvents as an alternate to other harmful solvents*. Biointerface Res. Appl. Chem, 2022. 12(10,33263). <https://doi.org/10.33263/BRIAC1210,44786>
8. Fuad, F.M. and M.M. Nadzir, *The formulation and physicochemical properties of betaine-based natural deep eutectic solvent*. Journal of Molecular Liquids, 2022. 360: p. 119392. <https://doi.org/10.1016/j.molliq.2022.119392>
9. Nowacki, K., M. Wysokowski, and M. Galiński, *Synthesis and characterization of betaine-based natural deep eutectic solvents for electrochemical application*. Journal of Molecular Liquids, 2020. 424: p. 127071. <https://doi.org/10.1016/j.molliq.2020.127071>
10. Usman, M.A., et al., *Ternary liquid-liquid equilibrium data for n-Hexane-Benzene-DES (choline chloride/ethylene glycol, choline chloride/glycerol, choline chloride/urea) at 303 K and 101.3 kPa*.

Applied Petrochemical Research, 2020. 10(3): p. 120-137. <https://doi.org/10.1007/s13203-020-00202-w>

11. Ijardar, S.P., V. Singh, and R.L. Gardas, *Revisiting the physicochemical properties and applications of deep eutectic solvents*. *Molecules*, 2022. 27(4): p. 1368. <https://doi.org/10.3390/molecules27041368>
12. Hirpara, D., et al., *Extraction and Separation Processes Using Deep Eutectic Solvents*, in *Deep Eutectic Solvents: Volume One: Fundamentals and Applications*. 2022, Springer. p. 127-163. https://doi.org/10.1007/978-3-032-3381-2_6
13. Pham, D.D., et al., *Advancing hydrodesulfurization in heavy Oil: Recent developments, challenges, and future prospects*. *Fuel*, 2022. 372: p. 132082. <https://doi.org/10.1016/j.fuel.2022.132082>
14. Lu, P., et al., *Performance of toluene oxidation over MnCe/HZSM-5 catalyst with the addition of NO and NH3*. *Applied Surface Science*, 2021. 567: p. 150836. <https://doi.org/10.1016/j.apsusc.2021.150836>
15. Yang, Y., et al., *Study on anti-sulfur dioxide poisoning of palladium-based catalyst for toluene catalytic combustion*. *International Journal of Hydrogen Energy*, 2021. 46(9): p. 6329-6341. <https://doi.org/10.1016/j.ijhydene.2020.11.126>
16. Hansen, B.B., et al., *Deep eutectic solvents: a review of fundamentals and applications*. *Chemical reviews*, 2020. 121(3): p. 1232-1280. <https://doi.org/10.1021/acs.chemrev.1c00380>
17. Suthar, K.J., *Natural deep eutectic solvents in extraction science: progress, challenges, and future prospects*. *Separation Science Plus*, 2020. 8(6): p. e70070. <https://doi.org/10.1002/sscp.70070>
18. Botella, M.B., et al., *Recent advances of extraction and separation of emerging organic contaminants through the application of natural deep eutectic solvents*. *TrAC Trends in Analytical Chemistry*, 2022. 171: p. 117018. <https://doi.org/10.1016/j.trac.2023.117018>
19. Islam, S., et al., *Desulfurization and denitrogenation using betaine-based deep eutectic solvents*. *Journal of Chemical & Engineering Data*, 2022. 69(6): p. 2244-2254. <https://doi.org/10.1021/acs.jced.1c00052>
20. Hizaddin, H.F., et al., *Extractive denitrogenation of diesel fuel using ammonium-and phosphonium-based deep eutectic solvents*. *The Journal of Chemical Thermodynamics*, 2016. 95: p. 164-173. <https://doi.org/10.1016/j.jct.2015.12.009>
21. Sosa, F.H., et al., *Deep eutectic solvents for sustainable separation processes*. *Sustainable Separation Engineering: Materials, Techniques and Process Development*, 2022: p. 60-62. <https://doi.org/10.1002/9781119741117.ch17>
22. Kučan, K.Z., et al., *Betaine+(glycerol or ethylene glycol or propylene glycol) deep eutectic solvents for extractive purification of gasoline*. *ChemistrySelect*, 2018. 3(44): p. 12082-12090. <https://doi.org/10.1002/slct.201803201>
23. Rodrigues, L.A., et al., *Deep eutectic systems from betaine and polyols – Physicochemical and toxicological properties*. *Journal of Molecular Liquids*, 2021. 330: p. 116201. <https://doi.org/10.1016/j.molliq.2021.116201>

٢٤. Rogošić, M., A. Krišto, and K. Zagajski Kučan, *Deep eutectic solvents based on betaine and propylene glycol as potential denitrification agents: A liquid-liquid equilibrium study*. Brazilian Journal of Chemical Engineering, ٢٠١٩. ٣٦(٤): p. ١٧٠٣-١٧١٦. <https://doi.org/10.1090/0.104-7632,2019.364S2019.0.49>

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