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Synthesis of Natural Deep Eutectic Solvent from EFB Waste and Used Cooking Oil for Cathode Recovery

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Abstract

This study develops a biomass-derived natural deep eutectic solvent (NADES) by combining potassium carbonate-rich oil palm empty fruit bunch (EFB/TKKS) ash as the hydrogen bond acceptor, and polyol-rich used cooking oil as the hydrogen bond donor for cobalt recovery from spent lithium-ion battery cathodes. The novelty of this work lies in converting two abundant local waste streams into a reusable green leaching medium for battery recycling. The HBA and HBD precursors were prepared through ash extraction, used-oil purification, and hydroxylation, followed by thermal mixing at molar ratios of 1:1, 1:2, and 2:1. The products were evaluated using density, viscosity, water-solubility, FTIR, GC-MS, AAS, UV-Vis, and qualitative cobalt confirmation tests. The 1:2 ratio produced the most stable homogeneous NADES, with a density of 0.8895 g/mL and viscosity of 55.57 mPa·s. FTIR and GC-MS analyses confirmed hydroxyl-bearing components and hydrogen-bond interactions that support eutectic solvent formation. During cobalt recovery, the first three regeneration cycles produced apparent cobalt-containing solid recovery values of 53.41%, 51.11%, and 49.30%, with solid recovery yields of 99.82%, 95.52%, and 92.15%, respectively. These findings indicate that EFB- and used-cooking-oil-derived NADES is a promising waste-based solvent platform for more sustainable lithium-ion battery cathode recycling, although further optimization is required to improve cobalt recovery efficiency and long-term solvent stability.

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INTRODUCTION

Lithium-ion batteries (LIBs) are advanced energy-storage systems that are widely used in portable electronic devices, electric vehicles, and renewable-energy storage because of their high energy density, good charge–discharge efficiency, and long service life (Perdana, 2020; Harper et al., 2019). The rapid growth of LIB consumption has consequently increased the amount of spent LIB waste. If not properly managed, spent LIBs may release hazardous components and cause environmental contamination. At the same time, these wastes contain valuable strategic metals, including cobalt (Co), lithium (Li), nickel (Ni), and manganese (Mn), which are essential for the battery and energy industries. Among the main components of LIBs, the cathode is one of the most important metal-bearing fractions and commonly consists of $LiCoO_2$ (LCO), $LiNiMnCoO_2$ (NMC), or $LiNiCoAlO_2$ (NCA) materials (Peeters et al., 2022; Wang et al., 2024). Therefore, cobalt recovery from spent LIB cathodes is important not only to reduce environmental pollution but also to sup-port circular management of critical metals in the energy-storage sector (Hanada and Goto, 2022).

Several recycling technologies have been developed to recover valuable metals from spent LIBs, particularly pyrometallurgical and hydro-

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metallurgical processes. Pyrometallurgical methods are relatively simple and suitable for large-scale processing, but they generally require high operating temperatures and high energy consumption. Hydrometallurgical methods can provide higher metal selectivity and recovery efficiency, but they often involve concentrated inorganic acids, large amounts of water, and additional purification steps that may increase environmental burden and processing cost (Schaeffer et al., 2018; Zante and Boltoeva, 2020). These limitations have encouraged the development of alternative leaching media that are more selective, less volatile, and potentially reusable.

Deep eutectic solvents (DESs) have emerged as promising alternatives for metal leaching and separation because their physicochemical properties can be adjusted through the selection of hydrogen bond acceptors (HBAs) and hydrogen bond donors (HBDs). Parameters such as polarity, viscosity, acidity, metal-complexation ability, and phase behavior can be tuned to improve dissolution and recovery performance (Tran et al., 2019; Padwal et al., 2022). In particular, hydrophobic eutectic solvents are attractive for metal recovery because they can form a separate organic phase, reduce aqueous wastewater generation, and allow solvent reuse after stripping or regeneration (Zainal-Abidin et al., 2021; van Osch et al., 2019; Hanada and Goto, 2022). Previous studies have shown the potential of polyol-based DESs for biomass processing and DES or hydrophobic eutectic solvent systems for the recovery of valuable metals from spent LIBs (Chen and Mu, 2019; Hossain et al., 2021; Peeters et al., 2020; Liu et al., 2023). However, many reported DES formulations still depend on synthetic, relatively expensive, or less locally available chemical components. This condition creates a need for cheaper, renewable, and locally abundant solvent precursors.

Indonesia, including West Kalimantan, has strong potential for the development of waste-derived eutectic solvents because of the abundance of palm-oil-related biomass residues and household used cooking oil. West Kalimantan has approximately 1,497,841 ha of oil palm plantation area (Wibowo, 2023), and palm oil processing can generate approximately 230 kg of empty fruit bunches (EFB/TKKS) per ton of processed palm oil (Salamiyah and Hizriani, 2023). EFB ash contains potassium-based compounds, particularly potassium carbonate (K_2CO_3), which can function as a hydrogen bond acceptor in eutectic solvent formation (Sanjaya et al., 2018; Ananda et al., 2023). Meanwhile, used cooking oil is a common domestic and culinary waste stream that mainly contains triglyceride-based compounds. Through purification and hydroxylation, used cooking oil can be converted into hydroxyl-bearing polyols that may act as hydrogen bond donors in eutectic solvent systems (Nofiyanti and Wardani, 2018; Turco et al., 2021; Fithry et al., 2024). Thus, EFB ash and used cooking oil are relevant local waste resources for developing low-cost biomass-derived natural deep eutectic solvents (NADESs). The valorization of biomass-derived materials has also been widely explored for environmental applications, including pollutant adsorption and remediation, indicating that biomass waste can be converted into functional materials with added environmental value (Sivakumar & Lee, 2022).

The main research gap addressed in this study is the limited availability of a simple waste-to-solvent route that integrates EFB-derived K_2CO_3 and used-cooking-oil-derived polyols as eutectic solvent precursors for cobalt recovery from spent LIB cathode materials. Although DES-based systems have been investigated for metal recovery, the combined use of agricultural ash-derived potassium carbonate as the HBA and used-cooking-oil-derived polyols as the HBD remains underexplored, especially in the context of hydrophobic NADES development for LIB cathode leaching. Therefore, the scientific novelty of this study lies in the synthesis of a biomass-derived hydrophobic NADES from two locally available waste streams, namely EFB/TKKS ash and used cooking oil, and its application as a leaching, recovery, and regenerable solvent system for cobalt from spent LIB cathodes.

Based on this gap, this study aims to synthesize and characterize biomass-derived hydrophobic NADES using EFB/TKKS-derived K_2CO_3 and used-cooking-oil-derived polyols, and to evaluate its ability to leach, recover, and regenerate cobalt from spent lithium-ion battery cathode materials. The expected contribution of this study is the development of a locally relevant green-solvent platform that supports circular economy strategi-es by connecting agricultural waste valorization, used-oil management, and critical-metal recycling.

METHOD

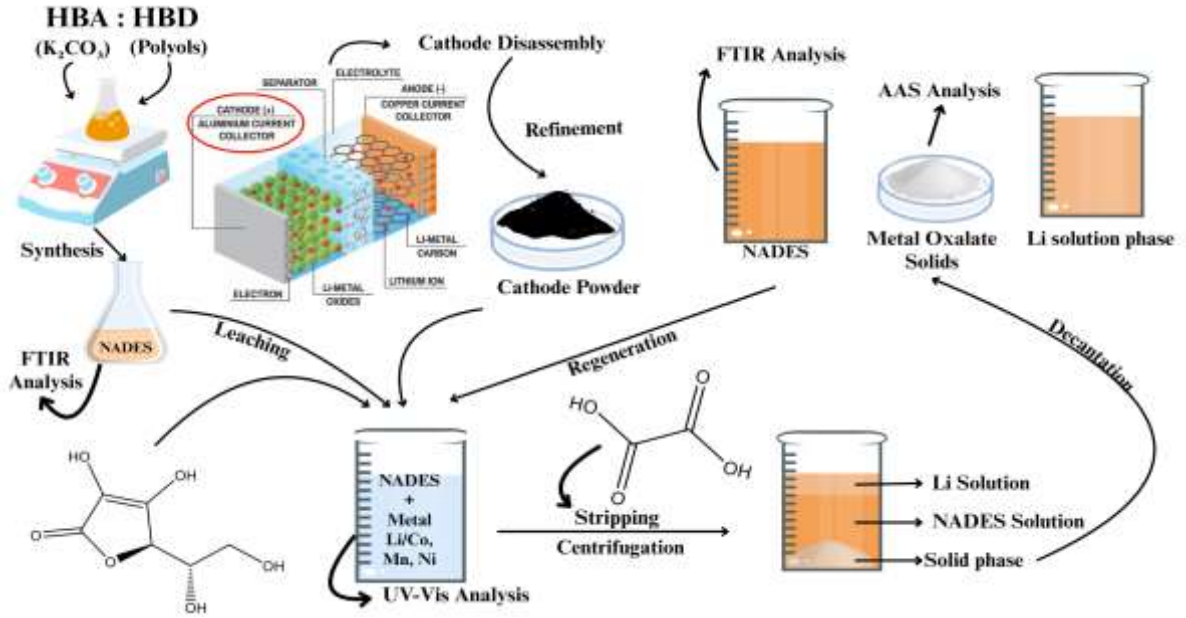

Figure 1. Research Scheme

Instruments and Materials

Analytical-grade reagents, including distilled water (H_2O), ammonium thiocyanate (NH_4SCN), ascorbic acid ($C_6H_8O_6$), hydrochloric acid (HCl), oxalic acid ($H_2C_2O_4$), 90% formic acid (CH_2O_2), ethanol (C_2H_5OH), and 30% hydrogen peroxide (H_2O_2), were purchased from Merck and used without further purification. Spent lithium-ion batteries, oil palm empty fruit bunches (EFB/TKKS), and used cooking oil were collected as waste materials for this study. The main instruments used were an analytical balance, magnetic stirrer-hotplate, oven, furnace, Büchner filtration setup, separatory funnel, centrifuge, FTIR spectrophotometer, GC-MS, AAS, UV-Vis spectrophotometer, density measurement apparatus, and viscometer.

Analytical data were interpreted using a combination of qualitative and quantitative approaches. AAS was used to determine potassium concentration in the EFB ash extract by standard calibration. FTIR spectra were interpreted from changes in O–H, C–H, C=O, and C–O absorption bands to verify hydrogen bonding and eutectic formation, while GC-MS was used to identify hydroxyl-bearing compounds in the used-cooking-oil-derived HBD fraction. UV-Vis spectra and cobalt spot tests were used to verify the presence of dissolved cobalt species and recovered cobalt solids. Solvent performance was evaluated from apparent cobalt-containing solid recovery, solid recovery yield, and changes in density and viscosity across regeneration cycles (Svehla, 1979; Nandiyanto et al., 2019; Zante and Boltoeva, 2020).

Extraction of HBA from EFB

EFB waste was cut into small pieces, dried at 105 °C for 60 minutes, and carbonized in a closed combustion system for 180 minutes. The resulting EFB carbon was ground and passed through a 120-mesh sieve, followed by calcination at 700 °C for 300 minutes to obtain dry ash rich in potassium compounds. This procedure was adapted from EFB ash preparation and potassium extraction methods reported by Sanjaya et al. (2018) and Ananda et al. (2023).

A total of 50 g of dry EFB ash was mixed with 250 mL of distilled water, stirred at 500 rpm, and heated at 90 °C for 60 minutes, then filtered using a Büchner funnel. The residue was re-extracted twice using the same water volume to maximize recovery of potassium-containing species. The combined filtrate was analyzed for potassium concentration using AAS to estimate the K_2CO_3-rich HBA fraction (Sanjaya et al., 2018; Ananda et al., 2023).

Extraction of HBD from Used Cooking Oil

A total of 100 mL of used cooking oil was mixed with 4.5 g of EFB ash as an adsorbent, and the mixture was stirred at 70 °C for 180 minutes. It was then filtered and stored in a dark container to minimize oxidation and preserve the purified

triglyceride fraction. This purification stage was adapted from used-cooking-oil adsorption methods using biomass-derived adsorbents (Al Qory et al., 2021; Fithry et al., 2024).

For hydroxylation, 250 mL of purified used cooking oil was heated under reflux at 40 °C while 380 mL of 90% formic acid and 30 mL of 30% hydrogen peroxide were gradually added under stirring. The reaction mixture was transferred into a separatory funnel and washed with distilled water until two distinct layers formed. The upper yellow polyol-rich phase was separated and washed three times with water (1:1, v/v), then characterized by GC-MS and FTIR to confirm the formation of hydroxyl-bearing HBD compounds. The procedure was adapted from the used-oil-to-polyol conversion and in situ performic acid epoxidation/hydroxylation approaches (Nofiyanti and Wardani, 2018; Turco et al., 2021).

Synthesis of Natural Deep Eutectic Solvent (NADES)

The K_2CO_3-rich HBA fraction and polyol-rich HBD fraction were mixed at molar ratios of 1:1, 1:2, and 2:1 using a thermal mixing method commonly applied in DES/NADES synthesis (Chen and Mu, 2019; Maduka et al., 2025). Each mixture was heated at 80 °C and stirred at 500 rpm until a homogeneous liquid phase was formed. The resulting NADES was cooled to room temperature and stored in a sealed container for density, viscosity, water solubility, and FTIR characterization.

Preparation of Lithium-Ion Battery (LIB) Samples

Spent lithium-ion batteries were immersed in a 5% NaCl solution for 24 hours to reduce the voltage to below 0.5 V before disassembly, following battery-safety pretreatment protocols used in LIB recycling studies and patent applications (Badan Riset dan Inovasi Nasional [BRIN], 2025; Harper et al., 2019). After discharge, the batteries were air-dried at room temperature and carefully dismantled to separate the cathode (+) and anode (–) components. The active cathode powder was collected and used as the primary sample for the metal leaching stage.

Leaching of LIB Cathodes Using NADES

A total of 70 mL of NADES was homogenized with 5–10% distilled water and 0.15 mol/dm³ ascorbic acid as a mild reducing agent to facilitate the reduction of Co^{3+} to Co^{2+} during leaching (Peeters et al., 2020; Hanada and Goto, 2022). Subsequently, 1% cathode powder was added to the homogeneous mixture, which was stirred at 500 rpm and 60 °C for 240 minutes. The leached solution in the NADES phase was analyzed using UV-Vis spectrophotometry to evaluate cobalt-containing species formed during the leaching process.

Recovery of Cobalt (Co) and Regeneration of Organic NADES over Repeated Cycles

The cobalt recovery and solvent regeneration procedure was adapted from Hanada and Goto (2022), with slight modifications. A 0.5 M oxalic acid solution was added to the organic NADES phase obtained after leaching at a 1:1 volume ratio. The mixture was stirred at 500 rpm for 1 hour at room temperature, then centrifuged at 3500 rpm for 10 minutes to separate the phases. The resulting two layers were separated using a separatory funnel to isolate the regenerated organic NADES phase from the cobalt-containing precipitate. The solid pellet was rinsed with distilled water, filtered, and dried at 80 °C for 1 hour. Apparent cobalt containing solid recovery and solid recovery yield were calculated to evaluate the recovery performance of the NADES system. Because the recovered cobalt-containing solid was confirmed qualitatively rather than fully quantified by ICP-OES or AAS, the recovery value was expressed as an apparent recovery indicator. Apparent cobalt-containing solid recovery was calculated based on the recovered cobalt-containing precipitate relative to the initial cathode sample.

$$\textit{Apparent cobalt-containing solid recovery (\%)} = (\textit{mass of cobalt-containing precipitate} / \textit{initial mass of cathode powder}) \times 100$$

Meanwhile, solid recovery yield was calculated using the following equation:

$$\textit{Solid recovery yield (\%)} = (\textit{mass of recovered solid} / \textit{initial mass of cathode powder}) \times 100$$

The recovered metal solid was qualitatively tested with 37% HCl and NH_4SCN using the classical cobalt spot test described by Svehla (1979). The regenerated NADES phase was then reused in subsequent leaching cycles to evaluate solvent stability, while density and viscosity were measured after repeated use to assess changes in solvent physicochemical properties that could

affect diffusion and cobalt recovery efficiency (Zante and Boltoeva, 2020; Liu et al., 2023).

RESULTS AND DISCUSSION

Extraction of Potassium Carbonate (HBA) from Oil Palm Empty Fruit Bunch (EFB)

The potassium (K) content in 50 g of EFB ash extract was analyzed using AAS with the K-flame method and standard calibration solutions (Figure 2). The calibration curve produced a linear regression equation of y = 0.1639x + 0.0033 with a correlation coefficient of R^2 = 0.9988 (Table 1), indicating good linearity for potassium determination. The analysis showed a potassium concentration equivalent to 37% (w/w), confirming that the EFB ash extract was rich in potassium. Since AAS determines elemental potassium rather than directly identifying its chemical form, the presence of K_2CO_3 was inferred based on previous reports showing that potassium species in EFB ash are predominantly present in carbonate form (Sanjaya et al., 2018; Ananda et al., 2023). Therefore, the EFB ash extract was treated as a K_2CO_3-rich hydrogen bond acceptor (HBA) precursor for NADES synthesis. The high potassium content supports the feasibility of using local EFB waste as a low-cost inorganic HBA source for biomass-derived NADES development.

Table 1. Potassium Standard Solution Data

ID Standard	Concentration (mg/L)	Mean Absorbance	%RSD	SD
Standard 1	0.5000	0.0870	1.4	0.0012
Standard 2	1.0000	0.1735	1.5	0.0026
Standard 3	1.5000	0.2499	1.4	0.0035
Standard 4	2.0000	0.3270	1.8	0.0060
Sample	9.7436	0.2116	1.2	0.0025
Auto Dilution		7,67x		
Manual Dilution		1000x		

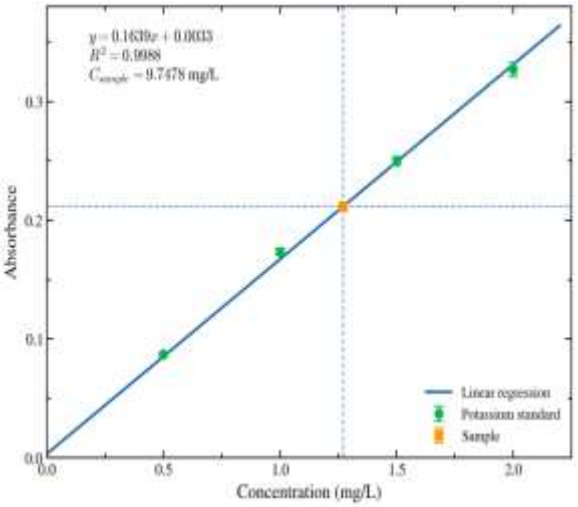

Figure 2. Standard Solution Curve

Extraction of Polyols from Used Cooking Oil

The polyol extract was analyzed using FTIR to identify functional groups in the used-cooking-oil-derived fraction. FTIR spectroscopy is commonly used as a qualitative method to assign characteristic absorption bands of organic functional groups in complex matrices, including O–H, C–H, C=O, and C–O vibrations (Kowalski et al., 2018; Nandiyanto et al., 2019). As shown in Figure 3, the spectrum exhibited a broad absorption band at 3384.59 cm^{-1}, which corresponds to O–H stretching vibrations from alcohol groups in the polyol-rich extract. This finding is consistent with Nofiyanti and Wardani (2018), who reported that alcohol O–H stretching in used-oil-derived polyols appears around 3380 cm^{-1}. Similar hydroxyl absorption near 3300 cm^{-1} has also been reported for glycerol- and diol-containing materials (Mitrea et al., 2019; Moura et al., 2019).

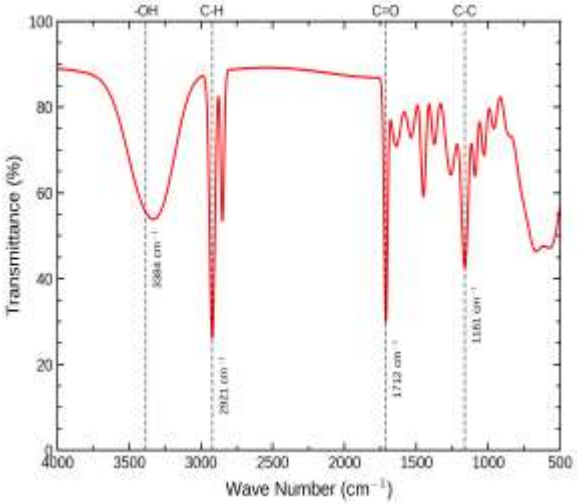

Figure 3. FTIR spectrum of the polyol extract

In addition, the bands associated with C–H, C=O, and C–O vibrations indicate that triglyceride-derived structures were still present while hydroxyl-containing groups were

introduced during the conversion process. These spectral features confirm that the HBD component contains active hydroxyl groups capable of forming hydrogen-bond networks in NADES.

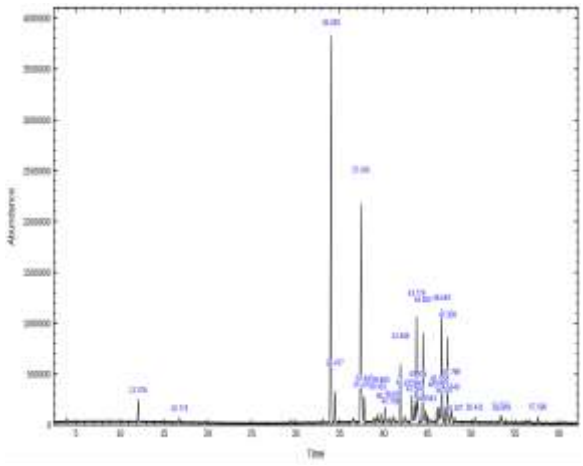

Figure 4. GCMS analysis of polyol extract

The GC-MS analysis (Figure 4) revealed a glycerol-derived diol compound at a retention time of 46.649 minutes, identified as 1-cis-vaccenoylglycerol. The detection of this compound supports the FTIR result by confirming that the used-cooking-oil extract contains hydroxyl-bearing molecules that can function as HBD components. Hydroxylated glyceride derivatives are important in eutectic solvent formation because they provide O–H donor sites that interact with carbonate or potassium-containing acceptor species through hydrogen bonding and ion-dipole interactions (Chen and Mu, 2019; Turco et al., 2021). Therefore, the GC-MS and FTIR results jointly demonstrate that the used-cooking-oil-derived fraction is chemically suitable for NADES synthesis and subsequent metal-complexation interactions.

Synthesis of Natural Deep Eutectic Solvent (NADES)

The synthesis of 70 mL of NADES was successfully achieved at an optimal HBA:HBD molar ratio of 1:2, producing a homogeneous and stable liquid phase (Figure 5c).

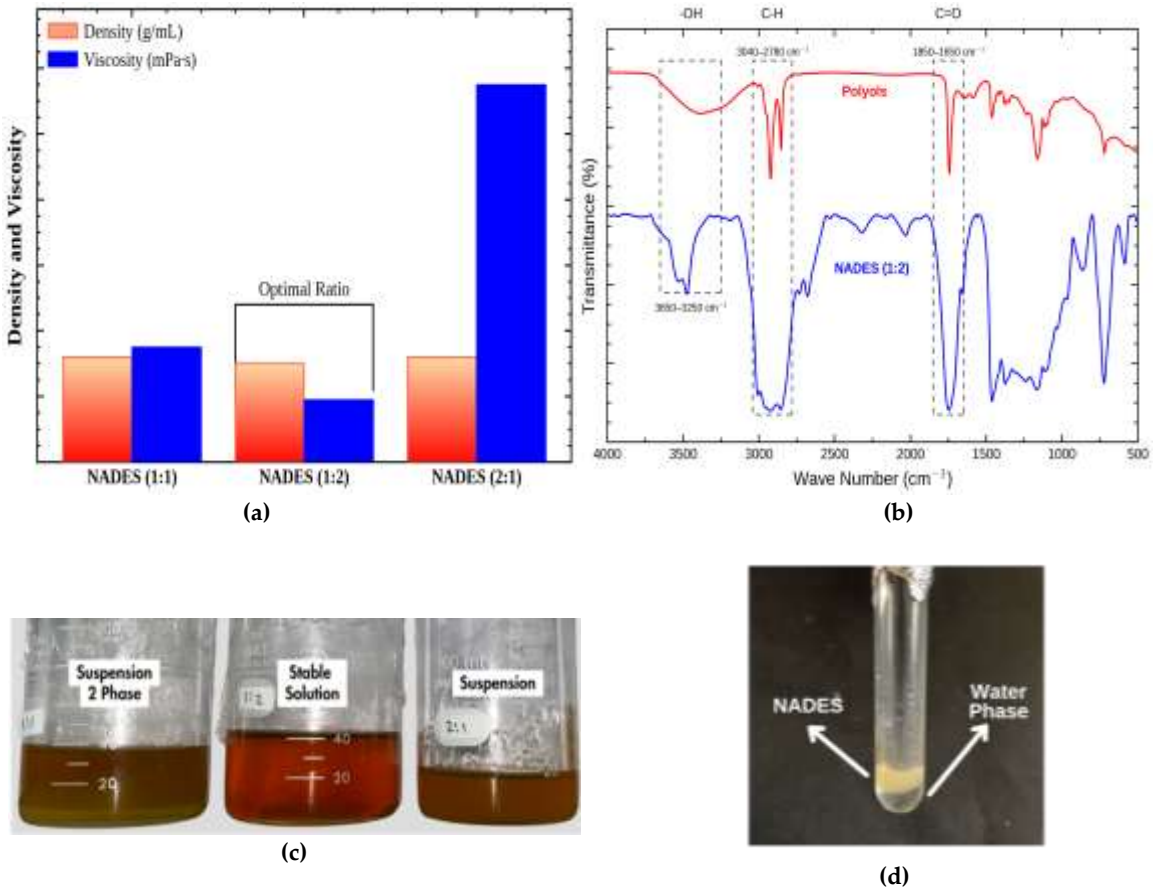

Figure 5. (a) Physicochemical of NADES at molar ratios 1:1, 1:2 and 2:1; (b) FTIR spectrum of NADES (1:2); (c) Visual of NADES solutions at ratios 1:1, 1:2 and 2:1 and (d) water solubility test of NADES.

The 1:1 and 2:1 systems formed suspensions, suggesting incomplete eutectic interaction or an imbalanced hydrogen-bonding composition. Physicochemical analysis (Figure 5a) showed that NADES (1:2) had a density of 0.8895 g/mL and a viscosity of 55.5694 mPa·s, both lower than those of the pure polyol fraction (0.93496 g/mL and 59.8085 mPa·s). The decrease in density and viscosity indicates the formation of a new liquid arrangement with weaker molecular packing and improved mobility. Similar behavior has been reported for potassium-based ionic eutectic systems, where interactions between potassium ions and donor groups alter the liquid microstructure and reduce intermolecular resistance (Gomes da Silva et al., 2025).

The FTIR spectrum of the synthesized NADES (Figure 5b) exhibited a broad O–H absorption band around 3473 cm^{-1} and a shoulder near 3527.80 cm^{-1}. Compared with the polyol extract band at 3384.59 cm^{-1}, the O–H region became broader and shifted, indicating a rearranged hydrogen-bond network after mixing the K_2CO_3-rich HBA and polyol-rich HBD components. The broadening of the O–H band is a key indicator of eutectic formation because it reflects stronger and more diverse hydrogen-bond interactions in the liquid phase (Kowalski et al., 2018; Nandiyanto et al., 2023; van Osch et al., 2019). Unlike the polyol precursor, K_2CO_3 does not contribute an organic O–H band; therefore, the spectral change is mainly attributed to interactions between hydroxyl groups, carbonate species, and potassium ions.

The absorption band at 2924 cm^{-1} corresponds to aliphatic C–H stretching from methyl and methylene groups in long hydrocarbon chains, while the carbonyl (C=O) band around 1745.58 cm^{-1} indicates the presence of ester or glyceride-derived carbonyl groups. These functional groups are relevant to cobalt leaching because hydroxyl and carbonyl oxygen atoms can participate in coordination or solvation interactions with dissolved Co^{2+} species, while the hydrophobic carbon chains contribute to phase separation from water. This interpretation is supported by the water solubility test (Figure 5d), which showed two immiscible phases, confirming the hydrophobic behavior of the NADES system (Makoś et al., 2020; Maduka et al., 2025). In the context of cobalt recovery, such amphiphilic structural features may help dissolve cobalt-containing species during leaching while enabling subsequent stripping and solvent regeneration.

Preparation and Leaching of LIB Cathode Using NADES

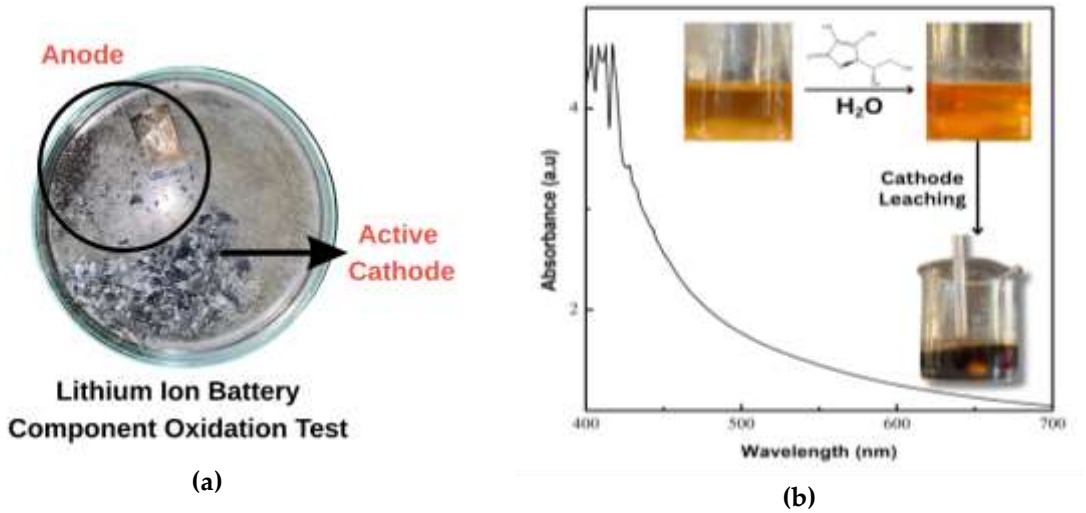

Figure 6. (a) Disassembled LIB components and cathode fraction; (b) UV-Vis spectrum of the leachate.

The spent lithium-ion batteries were first discharged in 5% NaCl solution and then dismantled to separate the anode and cathode components (Figure 6a). The anode appeared as black graphite powder adhered to copper foil, whereas the cathode fraction consisted of metal-oxide active material attached to aluminum foil. Voltage measurements before and after discharge showed a decrease from 2.28 V to 0.02 V, indicating that the batteries had reached a safer level for manual disassembly. This pretreatment is important because residual charge can pose

short-circuit, heat, or ignition risks during battery dismantling (Harper et al., 2019; BRIN, 2025).

UV-Vis analysis of the leaching phase (Figure 6b) showed spectral characteristics associated with cobalt-containing species in the NADES medium. In the presence of ascorbic acid, Co^{3+} in the cathode oxide can be reduced to Co^{2+}, which is more favorable for dissolution and complexation (Peeters et al., 2020; Hanada and Goto, 2022). The observed spectral response suggests the formation of soluble cobalt complexes stabilized by oxygen-containing groups from the NADES components. Similar DES-based leaching studies have reported that metal dissolution is governed by a combination of reduction, ligand coordination, hydrogen-bond disruption, and mass transfer within the viscous solvent phase (Tran et al., 2019; Wang et al., 2024). Therefore, the UV-Vis result supports the role of NADES as both a leaching medium and a stabilizing phase for dissolved cobalt species.

Recovery of Cobalt (Co) and Regeneration of NADES

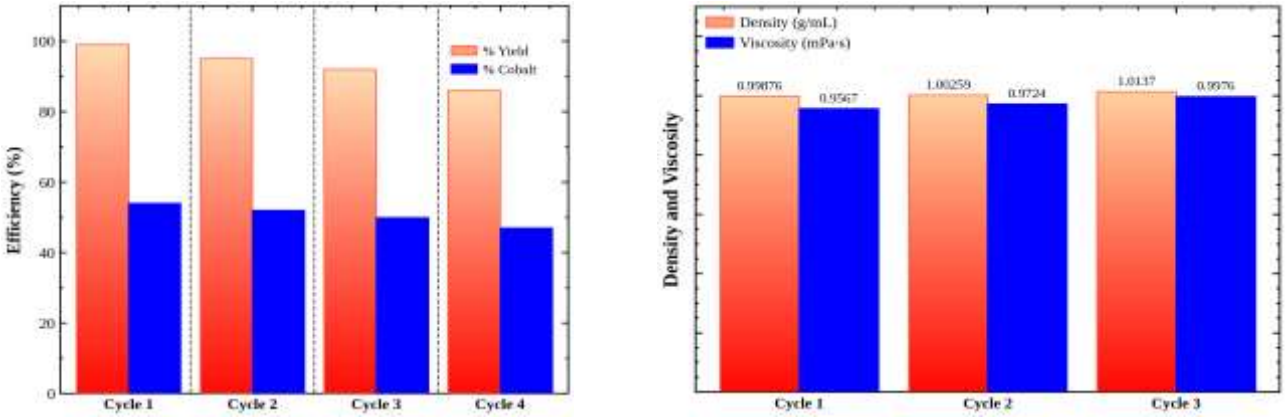

Figure 7. (a) Apparent cobalt-containing solid recovery and solid recovery yield; (b) physicochemical properties of regenerated NADES.

Cobalt dissolved in the NADES phase was recovered using 0.5 M oxalic acid as a stripping agent. Oxalic acid promotes the transfer of cobalt from the organic NADES phase and precipitates cobalt as cobalt oxalate, enabling partial solvent regeneration (Peeters et al., 2020; Hanada and Goto, 2022). Based on Figure 7a, the first three cycles showed solid recovery yields of 99.82%, 95.52%, and 92.15%, respectively. The apparent cobalt-containing solid recovery values were 53.41%, 51.11%, and 49.30% for the first, second, and third cycles, respectively, and decreased to 46.33% in the fourth cycle. Since the recovered solid was confirmed qualitatively using the cobalt thiocyanate test rather than fully quantified by ICP-OES or AAS, these values should be interpreted as indicative recovery performance of cobalt-containing precipitates rather than absolute cobalt recovery. The decline in recovery performance is associated with changes in the physicochemical properties of the regenerated NADES, particularly increased density and viscosity (Figure 7b), which can limit mass transfer and slow the diffusion of metal ions in the liquid phase (Zante and Boltoeva, 2020; Liu et al., 2023).

The recovery trend is consistent with previous studies showing that reusable hydrophobic eutectic systems can facilitate cobalt transfer and precipitation through leaching–stripping cycles, although the recovery performance strongly depends on solvent composition, viscosity, water content, and metal coordination environment (Hanada and Goto, 2022; Wang et al., 2024). The qualitative cobalt confirmation test produced a color change from pink cobalt oxalate (CoC_2O_4) to blue after reaction with HCl and NH_4SCN. This observation supports the presence of cobalt-containing species in the recovered solid, consistent with the formation of tetrathiocyanatocobaltate(II), $[Co(SCN)_4]^{2-}$, as described in classical qualitative inorganic analysis (Svehla, 1979). However, because the cobalt content in the recovered solid was not quantitatively determined, future studies should include ICP-OES, AAS, or calibrated UV-Vis analysis to establish a complete cobalt mass balance and determine true cobalt recovery efficiency.

CONCLUSION

This study successfully synthesized a biomass-derived natural deep eutectic solvent (NADES) from oil palm empty fruit bunch (EFB/TKKS) ash and used cooking oil. The 1:2 HBA:HBD molar ratio produced the most stable homogeneous solvent, with a density of 0.8895 g/mL and viscosity of 55.57 mPa·s. FTIR and GC-MS analyses confirmed the presence of hydroxyl-bearing HBD components and hydrogen-bond interactions between the K_2CO_3-rich HBA and polyol-rich HBD fractions, while the water solubility test confirmed the hydrophobic behavior of the NADES system.

The synthesized NADES was able to leach cobalt-containing species from spent lithium-ion battery cathode materials and allowed the recovery of cobalt-containing precipitates through oxalic acid stripping. The first three regeneration cycles produced apparent cobalt-containing solid recovery values of 53.41%, 51.11%, and 49.30%, with solid recovery yields of 99.82%, 95.52%, and 92.15%, respectively. The decline observed in the fourth cycle was associated with increasing density and viscosity, which likely reduced mass transfer during leaching and stripping. These findings show that solvent stability and diffusional properties are key factors controlling the recovery performance of the waste-derived NADES system.

The novelty of this research is the integration of two locally abundant waste materials EFB/TKKS ash and used cooking oil as HBA and HBD precursors for preparing a hydrophobic NADES for LIB cathode recovery. Practically, this approach offers a preliminary waste to solvent platform that connects palm-oil biomass valorization, used-oil management, and critical metal recycling. Its potential impact lies in reducing reliance on hazardous inorganic solvents and supporting circular economy practices in Indonesia. Future optimization is necessary to improve cobalt recovery above the current moderate level, extend solvent lifetime beyond repeated cycles, and validate the process using more diverse spent LIB cathode compositions.

RECOMMENDATION

Future research should focus on optimizing the NADES molar ratio, water content, reductant concentration, leaching temperature, and contact time to enhance cobalt recovery efficiency. Further work should also include quantitative determination of cobalt and other metals using AAS or ICP-OES, mass balance analysis, toxicity and biodegradability assessment of the regenerated solvent, and comparison with other biomass-derived HBA/HBD combinations. Expanding this approach to NMC and NCA cathode materials would also provide a more comprehensive evaluation of its potential for practical lithium-ion battery recycling.

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