Highly Efficient Lithium-ion Battery Cathode Materials Recycling Using Deep Eutectic Solvent Based Nanofluids

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Note 1: A typical separation experimental preparation procedure

Battery removal and pre-processing: The discarded cathode pieces used in the experiments were discharged and disassembled prior to the separation experiments. Specifically, after obtaining the discarded lithium-ion battery, it was discharged using the existing Xinwei discharge device in the laboratory with a discharge parameter of 10 V, 5 A and a discharge rate of 2 C. After the battery had been discharged at a high rate, a discharge rate of 0.5 C was continued to discharge all the residual power in the discarded battery. After the battery is discharged and cooled down, the battery is disassembled in a fume hood using a simple mechanical disassembly method to cut and separate the battery shell and internal components such as the electrode sheet and diaphragm using scissors and a cutter respectively. The electrolyte, which is moderately toxic, evaporates spontaneously during disassembly, and the final electrode sheet is separated only from the diaphragm for the next step. In addition, the obtained positive electrode material was rinsed and soaked in dimethyl carbonate (DMC), one of the main components of the electrolyte and non-toxic, for experimental safety and elimination of interference factors in the separation experiments.

Preparation of deep eutectic solvent nanofluids: The deep eutectic solvent nanofluids are prepared in a two-step process using deep eutectic solvent as the base solution. The first step is the preparation of the base solution, which is a glycol/choline chloride based deep eutectic solvent used in the separation experiments, and the corresponding deep eutectic solvents are prepared in molar ratios of 1:1, 2:1, 3:1, 4:1 and 5:1. To ensure the experimental method of orthogonal variables, the preparation conditions were uniformly set to 100 °C and 600 r/min. The second step was the addition of nanoparticles, and the nanofluids used in the present work were added according to the mass fraction, which is an experimental parameter. The mass fractions used in this experiment were 0.1%, 0.2%, 0.5%, 0.8%, and 1.0%. The nanoparticles were selected according to different criteria of six types namely, graphene, graphene oxide, boron nitride, silicon carbide, silicon dioxide, respectively. Therefore, the nanofluids prepared and used in this experiment are 30 in total according to the difference of type and mass fraction.

Lithium-ion battery separation experiment using deep eutectic solvent: The experimental procedure of lithium ion battery cathode active material separation experiment is relatively simple with low pollution. The treated waste lithium-ion battery cathode material was cut into square pieces of 1×1cm2 size and added into the prepared deep eutectic solvent nanofluid according to the solid-liquid ratio of 1g: 30ml. The experimental conditions were set at 60°C, 70°C, 80°C, 90°C and 100°C with a fixed rotational speed of 600 r/min. The time parameters were set at 8h, 12h, 16h, 20h and 24h. In order to reduce the error of the experiment, each experiment was repeated three times and more, and the error range of the separation rate was obtained. After the reaction, the post-processing of the experiments and the calculation of the separation rates were also relatively simple. Only the solid-liquid reaction system was filtered and cleaned at the end of the reaction to achieve the solid-liquid separation. Then the black lithium-ion battery cathode active material particles and the large aluminum foil were separated and dried separately. Finally, the aluminum foil was weighed and calculated according to the separation rate calculation formula in the text. Here, the whole separation experiment was completely finished.

Deep eutectic solvent recycle experiments: Deep eutectic solvents (DESs) form a new type of solvent with low toxicity and recyclability due to the formation of hydrogen bonds. The same experimental conditions for nanofluid preparation and separation as before were used to perform five separations of deep eutectic solvent cycles. After the filtration step of the previous separation experiment, the filtrate was used again as the base fluid to prepare 0.2 wt% (boron nitride) BN nanofluid to continue the next experiment until the reaction was treated to obtain the result that the aluminum foil was clearly no longer separated from the black cathode material particles. From the experimental phenomena, it can be concluded that after four separation experiments, the separation experiments with recycled deep eutectic solvents became less effective, so we finally conducted five separation cycles of experiments. The specific experimental results and data have been reflected in the text.



Fig. S1 Schematic diagram of experimental process.

Note 2: A typical leach experimental preparation procedure

The experiments in this work also contain leaching experiments on important metallic elements in commercial NMC111 batteries. to investigate the leaching ability of deep eutectic solvent nanofluid for metal elements. It mainly contains experiments on the leaching of metallic elements by temperature with deep eutectic solvent and deep eutectic solvent nanofluid, and data processing of LA-ICP-MS after the experiments. From the experimental data and results, we continue to evaluate the ability of deep eutectic solvent nanofluid to recover waste lithium-ion battery cathode materials. Specifically, these include.

Leaching experiments of waste lithium-ion battery cathode materials. Because it is desired to investigate the effect of increasing temperature on the leaching of lithium ion battery cathode material by deep eutectic solvent nanofluid. So the most important parameter is the effect of temperature on the leaching experiment. In the leaching experiments, we increased the temperature to 120°C versus 140°C, keeping other conditions constant, to obtain the results of the leaching ability of deep eutectic solvent and deep eutectic solvent nanofluid on nickel, cobalt, and manganese, the metal elements contained in commercial batteries.

Post leaching experiment treatment. After 20 hours of leaching reaction, the solidliquid mixture was filtered three times to obtain a clarified filtrate. Several filtrates were also subjected to LA-ICP-MS test pre-treatment. The filtrates were first extracted in small portions and the organic matter was carbonized at 200°C, and then slowly added with aqua regia at high temperature until the clarified liquid was obtained. To ensure the accuracy of the test, the liquid was diluted to 50 ml, and finally the sample was loaded and sent for testing.

LA-ICP-MS test and experimental results processing. LA-ICP-MS is a test device that can detect most of the elements in the periodic table of elements both qualitatively and quantitatively. Therefore, we used this test to perform the leaching rate of metal elements, and the test data were used to calculate the mass of metal elements. The calculated mass is compared with the mass of the metal in the original waste cell to derive the leaching rate of the metal from a mass perspective. It is worth mentioning that the reliability of the LA-ICP-MS test is determined by the internal standard recovery. The magnitude of the internal standard recovery can reflect the strength of matrix effects or anomalies in the feed process and can be used to correct for matrix effects. It is widely accepted that data obtained with internal standard recoveries in the range of 80% to 120% are reliable. And whether the internal standard recovery rate is within a reasonable range is also taken as an important indicator in our work. So it can be further verified that the leaching ability of deep eutectic solvent nanofluid for metal can be enhanced with the increase of temperature.

Note 3: LA-ICP-MS Leaching rate calculation

Due to the testing technique, the data from LA-ICP-MS tests are measured in ppb. Therefore, further calculation of the leaching rates of nickel, cobalt, and manganese elements in deep eutectic solvent nanofluids is required. We used to calculate the mass of metal elements in the tested filtrate from the mass point of view and compared it with the mass of the same metal elements calculated in the raw material to derive the actual leaching rate. The leaching rate in this work is calculated as follows: Calculation of metal element concentration :

$$C = \frac{C_s V_s}{m_s M}$$
 Eq S1

Calculation of leaching rate :

$$\eta_{leach} = \frac{CMV_l \rho_l}{m_{element}}$$
Eq S2

where C_s , V_s , and m_s are the concentration, volume, and mass of an element in the test sample, respectively, M is the molar mass of this element, C is the concentration of this element in the leachate, V_1 and ρl are the volume and density of the leachate, respectively, $m_{element}$ is the theoretical mass of this element in the waste cathode material, also after testing from LA-ICP-MS using these two equations were calculated.

Note 4: Infrared photography experimental equipment

In the text, we carried out infrared camera to obtain the temperature rise changes of different types of nanofluids. The specific experimental device cannot be displayed in the text due to length reasons. So, as shown in **Fig. S2**. We use the existing magnetic stirrer and 50ml small beaker to build a simple temperature visualization platform. Finally, through the infrared camera in a fixed position time range for shooting.



Fig. S2 Dynamic experimental device.

Note 5: Geometry of the Multi-phase flow mass transfer simulation

In this work, a series of experiments have been made around the separation and leaching of waste lithium-ion battery cathode materials in deep eutectic solvent nanofluids, but none of them have addressed the mechanistic aspects. On the one hand, there are still many unexplained imaginations in the recovery of waste lithium-ion battery cathode materials, and on the other hand, the mechanistic investigation has not been deeply explored. Therefore, we chose to numerically simulate the mass transfer process of waste lithium-ion battery cathode material particles in a deep eutectic solvent in a stirred tank from the perspective of heat and mass transfer in our discipline using the general-purpose Ansys Fluent software. In the multiphase flow related part of this work, due to textual limitations, many details cannot be shown specifically in the text. Therefore, they will be explained in this section with additional information.

The model of the stirrer is referenced from previous related articles. It is our impeller model derived from a synthesis of the relevant literature. Our geometric model is compared with the previous model of the agitator impeller as shown in Fig.S3. In previous work, it has undergone progressive improvements with single-blade impellers, two-blade impellers, three-blade impellers and four-blade impellers. The three-blade impeller was found to have unique advantages. For example, de Lamotte et al.¹ studied the hydrodynamic field of a hybrid tank with two impellers. Ansys Fluent software and an implementable k-ɛ-turbulence model were used to develop an implementable k-ɛturbulence model and a volumetric method of fluid for multiphase flow. Two approaches to simulate impeller motion were investigated using a moving reference system and a sliding mesh approach. A qualitative and quantitative comparison of the velocity fields obtained by numerical simulations and PIV measurements was performed and a good overlap was obtained. It is clear from his series of work on stirrers that the three-blade impeller has a stronger influence on the fluid flow and solute dispersion in the stirrer relative to the number of other blades. To better simulate and compare the degree of homogeneous dispersion of solid particles in the liquid phase, we used a three-bladed impeller in our modeling to simulate stirring in a real stirred tank. In addition, to better simulate the flow in the stirrer and reduce the resistance, we chamfered the impeller blades to reduce the resistance to flow and vortex, which is also critical for the chemical reactions occurring in the reactor.

The final geometric model of the stirred tank is shown in **Fig. S3e**. The overall size of the mixing tank is 120*120*200 mm, including two parts: rotating domain and stationary domain, where the rotating domain is the small cylindrical shaped area including the stirrer, and the other areas are stationary domains. The size of the stationary domain is 116*116*200 mm, and the rotating domain is the inner area of the rotating domain 60*60*25 mm. The material in the mixing tank is mixed by a three-blade impeller of 54*54*16 mm size.



Fig. S3 : a)², b)³, c)⁴, d)⁵ Common agitator impeller geometry model; e)our impeller geometry mode.

Note 7: Mesh of the Multi-phase flow mass transfer simulation

To understand the mechanisms associated with solid-liquid mass transfer in a stirred reactor, the flow field was simulated for different configurations using Ansys Fluent 2020, and the mesh was depicted using Ansys Fluent Meshing 2020. The entire fluid computational domain consists of two parts, namely the rotational domain and the stationary domain. To obtain a high-quality mesh, the rotational domain is divided into 3 blocks: the small cylindrical region where the impeller is located, the rotational axis and the stationary domain. In this case, the rotational and stationary domains transfer material and data through an interface. Therefore, the external surface grid of the small cylinder and the grid of the impeller are divided into a more precise hexahedral grid. The other regions are divided into tetrahedral meshes. In addition, we also adjusted the mesh size to 0.1^{-2} mm and 2^{-4} mm for the rotating and stationary domains, respectively. The total number of meshes outlined is about more than 2.3 million, and the meshes on both sides of the boundary are encrypted by more than 5 layers at each boundary, and the details of the mesh structure are shown in Fig. S4. Where Fig.S4a shows the grid encryption and details of the x-axis profile and Fig.S4b shows the grid encryption and details of the y-axis profile. Since the main simulation focus of this work is on the diffusion of the cathode material particles in the stirred reactor, the main attention is paid to the two parameters of the flow field of the solid-liquid system and the solid volume fraction distribution. The parameters related to the other experiments were unified in the specific settings, and the number of nanoparticles added in the simulation process was all set to 0.2 wt%. The nanoparticles contained in the nanofluid itself are neglected and the nanofluid is considered as a special single-phase flow. Therefore, the simulations were performed using the Eulerian multiphase flow mixing model and the multi-reference system (MRF) method assuming a constant bubble diameter of 11.2 μm. The gas inlet and gas outlet are set as velocity inlet and pressure outlet boundary conditions, respectively. For the wall case, a no-slip boundary condition is specified. The discretization equations are solved by the SIMPLE discretization algorithm using the default model constants. The second-order windward and second-order implicit transient formulations of the momentum are utilized. The appropriate time step is set to 0.01 seconds. The convergence of the calculation was determined by testing whether the residuals of the iterations were less than 10^{-4} and whether the torque was constant.



Fig. S4 x-axis and y-axis grid encryption detail diagrams: a),b)

Note 8: Related formulas of the Multi-phase flow mass transfer simulation

Conservation of mass equation :

$$\nabla \cdot (\rho v) = 0$$
 Eq S3

Conservation of momentum equation. :

$$(v \cdot \nabla)\rho v = -\nabla p + \mu \nabla^2 v$$
 Eq S4

Energy conservation equation. :

$$v \cdot \nabla T = \frac{\lambda}{\rho c_p} \nabla^2 T$$
 Eq S5

Realizable k-ɛ turbulence model⁶ :

Note 9: Internal standard recovery rate of LA-ICP-MS

In LA-ICP-MS testing, the most important parameter to evaluate the reliability of the data is the internal standard recovery. Therefore, the internal standard recovery curves related to the leaching experiments in our work are all presented in **Fig. S5**. As can be seen in **Fig. S5**, the internal standard recoveries we obtained were all within the confidence range of 80%-120%. Therefore, our leaching experimental data are very reliable.



Fig. S5 Internal standard recovery graph of LA-ICP-MS.

Note 10: Information about the cathode active materials used in the experiments In this work, the disassembled and used batteries were all discarded commercial NMC111 batteries purchased from the manufacturer. And the specific experimental compositions we refer to the composition and weight of the batteries with the same cathode active material tested by LA-ICP-MS for the relevant separation and leaching rate calculations. The specific compositional data and ratios are reflected in **Table S1** and **Fig. S6**.

 Table S1 Content of major metal elements in waste lithium-ion battery cathode

 materials.

Chemical element	Quality	Proportion	Total weight
Li	0.0493	4.93%	
Al	0.0802	8.02%	0.5263
Mn	0.1047	10.47%	
Со	0.1441	14.41%	
Ni	0.1480	14.80%	



Fig. S6 Content of major metal elements in waste lithium-ion batteries.

Note 11: Materials and Methods of the Multi-phase flow mass transfer simulation To simulate solid suspensions, modern researchers usually use two models: the multiphase Eulerian model and the mixed Eulerian model^{3, 6-8}. In the multiphase Eulerian model, the equilibrium equations of continuity and momentum for each phase are solved. The hybrid Eulerian model is a simplified version of the multiphase flow model and is only applicable to sparse suspensions and a small number of Stokes particles. For this model, the equilibrium equations of continuity and momentum of the mixed phases are solved. where the hydrodynamic parameters are calculated by averaging the properties of each phase over the mass. The hybrid Eulerian model incorporates only the volume fraction transfer equation to solve for the transfer of the dispersed phase to the continuous phase. It consists of four different interphase forces: the lift force, the Bassett force, the virtual mass force and the drag force. Since the lift, Bassett and virtual mass forces have little effect on the modulated solid content profile, only drag forces can be considered in the interfacial momentum exchange terms^{9, 10}. In previous studies using both models, it was shown that the multiphase Eulerian model is more accurate than the mixed Eulerian model. Therefore, in our work, the Eulerian multiphase model was used to simulate the process. to achieve a clearer representation of the mass transfer process. In this work, we chose a multiphase Eulerian model to describe the flow and diffusion behavior of nanofluid and solid-phase cathode material particles considered as a single liquid phase. The multiphase Eulerian model is the most complex model, which solves a set of momentum equations for each phase. The results obtained for each phase will be more accurate, but will also have a larger computational demand in terms of volume. To more clearly represent the dispersion in turbulence, we chose the implementable k- ε model. When using a particle model, the scattering turbulence model is the appropriate model. And the cathode material particles were set to 11.2 µm. Finally in this study good network quality is ensured so that the skewness of the mesh quality is less than 0.96. We defined two fluid regions: a rotating inner cylinder centered on the rotor and another volume containing the rest of the reactor. The second volume contains all the rest of the rotating domain at the reactor. The interface is located at the same distance from the rotor blades and the inner edge of the

baffle. A finer grid is used around the turbine that has a larger spatial velocity gradient. Fig. S4 shows that a finer grid is used around rotors that are expected to have a higher spatial velocity gradient. It is reasonable to keep the same grid size between the rotating and stationary interfaces. Keeping the same grid size between the rotating and stationary interfaces ensures a good exchange of hydrodynamic quantities between the two regions during the calculation. A good exchange of hydrodynamic quantities between the two regions during the calculation. The "no-slip" condition was applied to the walls of all geometries." The "no-slip" condition was applied to the walls of all geometries^{3, 5, 8, 11, 12}. There are two working materials, two materials corresponding to two phases: a liquid with density $\rho_l = 1107.4 \text{ kg} / \text{m}^3$, and a particle size $d_p = 11.2 \text{ }\mu\text{m}$. Other material parameters are shown in Table S2. In addition to the multiphase Eulerian model, we continue to use the turbulent flow model. It is difficult to reduce to a laminar flow model because the motion of the fluid in the stirred reactor exhibits a long and noncontinuous period. However, the limitations of the turbulence model in predicting the dissipation rate and power number are objective¹¹. And the numerical results also depend on the limitations of the model used to simulate the impeller rotation. Our analysis focuses only on predicting the flow field and particle dispersion in the reactor to evaluate particle mass transfer in different types of nanofluids. For this purpose, the accuracy of using turbulence models can also be achieved with good results. While in the specific setup, we use a sliding grid approach where the resolution of the flow is unstable. At each time step, the position of the rotating zone relative to the stationary zone is recalculated and the mesh interface of the rotating zone slides along the interface of the stationary zone through the mesh interface of the rotating zone. This leads to greater demands on computing power such as computers, which means that the computations are time consuming. For all simulations, the time step is set as a function of the impeller speed, i.e. 600 r/min. The time step is set to 0.01 sec. At the end of each time step, after a maximum of 20 iterations, the convergence criteria for continuity, momentum and turbulence reach 10-4. The simulations are performed in double precision using an isolated implicit solver in double precision. The transient formulation is second order and the spatial discretization scheme of the pressure is

PRESTO! Momentum and turbulence are quickly. The SIMPLE algorithm has been applied to the pressure-velocity coupling¹³⁻¹⁶. The results were analyzed after a total flow time of 1 s because the diffusion caused by a stirring speed of 600 r/min is very fast and brings a large amount of calculations.

Material	mass fraction	Density	Viscosity	Diameter
	(wt%)	(kg / m^3)	(kg / (m*s))	(mm)
Deep eutectic solvent	-	1107.4	0.01614	-
Graphene nanofluid	0.2	1141.2	0.02618	-
SiC nanofluid	0.2	1136.2	0.02693	-
graphene oxide nanofluid	0.2	1100.2	0.02092	-
SiO ₂ nanofluid	0.2	1118.2	0.02362	-
carbon black nanofluid	0.2	1123.1	0.02520	-
BN nanofluid	0.2	1122.5	0.02420	-
Lithium-ion battery	-	-	-	0.0112
cathode material particles				

Table S2 Material and simulation parameters (the testing temperature is 60 °C).

Table S3 Specific parameters of the simulation

Temperature	Rotate speed	Time step	Calculating	Initial volume fraction
			time	of cathode material
(K)	(r / min)	(s)		(%)
			(s)	
333.2	600	0.01	1.00	0.35

Note 12: Velocity distribution of the Multi-phase flow mass transfer simulation

In this work, two important parameters are of main interest: velocity and volume fraction. Because the velocity represents the flow state in the stirred reactor and the volume fraction of the cathode material represents the diffusion effect in the stirred reactor. They are both important indicators to assess the mass transfer effect of the cathode material in a deep eutectic solvent. The results of the flow and diffusion in the seven solvents have been plotted in the main paper. The specific values and increases and decreases of the two parameters in each solvent are not shown due to space limitation. Therefore, they are added here.

The velocity distribution is the parameter that most visually reflects the flow state and is the most commonly used parameter in simulations. In this work, the velocity distribution also reflects the stirring effect of the fluid in the same working time from the side. This is because a higher average velocity in the x-axis and y-axis profiles indicates less resistance to the stirring process, which means that the mass transfer process is more favorable. **Table S4** details the velocity growth rates in the two important profiles. It can be seen that the other nanofluids show negative velocity growth in the x-axis profile and only in the BN nanofluid shows a good growth rate of 30.90%. On the other hand, the y-axis profile presents essentially the same velocity variation because it is right in the center of the rotational domain. This also indicates that the simulation has been carried out around a speed of 600 r/min in the center of the stirring.

Solvent	x-axis	Amplification	y-axis	Amplification
	(m/s)	(%)	(m/s)	(%)
Deep eutectic solvent	0.29	/	0.38	/
Graphene nanofluid	0.22	-22.21	0.42	0.11
SiC nanofluid	0.23	-20.81	0.42	0.11
graphene oxide	0.24	-16.95	0.22	-0.42
nanofluid				
SiO ₂ nanofluid	0.23	-20.39	0.43	0.12
carbon black	0.25	-11.48	0.43	0.12
nanofluid				
BN nanofluid	0.37	30.90	0.30	-0.22

 Table S4 Average velocity table of cathode material.

Note 13: Volume fraction distribution of the Multi-phase flow mass transfer simulation.

The volume fraction of the cathode material for Li-ion batteries is the most important and intuitive parameter in this work. The distribution of the volume fraction directly reflects the diffusion of the solicited material. Since the initialization distributes all the cathode material with 35% volume fraction directly below the stirrer. The lower the average volume fraction, the more uniform the diffusion is indicated. The specific simulation growth rate data are shown in **Table S5**. the average volume of the X-axis profile of BN nanofluid is only 0.16%, which is a 9-10 times increase in diffusion uniformity compared to other nanofluids. Its volume fraction growth rate also decreases nearly twice as much as DES, at 86.41%. Moreover, the same as the velocity distribution, the growth rate of the diffusion effect in the y-axis due to the position is not significant. The volume fraction growth rate of the anode material is reduced by 96% compared to DES. This result strongly reflects that the mass transfer effect of BN nanofluid is much higher than that of other nanofluids. It also corroborates with the separation experiments in the main text.

Solvent	x-axis	Amplification	y-axis	Amplification
	(%)	(%)	(%)	(%)
Deep eutectic solvent	1.17	/	1.54	/
Graphene nanofluid	1.05	-10.47	0.92	-0.40
SiC nanofluid	1.02	-13.03	0.97	-0.37
graphene oxide				
nanofluid	1.24	6.14	1.92	0.25
SiO ₂ nanofluid	0.97	-17.16	0.95	-0.38
carbon black				
nanofluid	0.96	-17.63	0.94	-0.39
BN nanofluid	0.16	-86.41	0.06	-0.96

Table S5 Volume fraction of cathode material distribution

Note 14: Nomenclature

- C particle concentration in specified region, m-3
- d_p particle diameter, m
- ρ_1 density of the liquid phase, kg/m3
- ρ_s density of the solid phase, kg/m3
- n rotational speed, r/min
- x radial co-ordinate direction, m
- ChCl choline chloride
- k thermal conductivity, W/(m*K)
- MRF multi-reference frame
- LIBs lithium-ion batteries
- DESs deep eutectic solvents
- DES deep eutectic solvent
- DMC dimethyl carbonate
- GO graphene oxide
- EG ethylene glycol

Note 15: Raw data from FTIR

The FTIR data plots in the main text are smoothed curves, and the original processed data images are presented in Fig.S7 and Fig.S8:



Fig. S7 Raw FTIR of waste cathode material.



Fig. S8 FTIR after separation experiment.

Note 16: TEM of cathode materials for lithium-ion batteries

To present the morphological changes of the cathode material of Li-ion battery before and after the experiment better. We conducted TEM tests on the raw material and the material after the experiment. The specific image is shown in **Fig. S9**. It can be seen from this that the adhesive can be clearly seen in the original material. However, the material after the peeling experiment shows almost no adhesive residue and presents a smoother edge.



Fig. S9 TEM of (a) before and (b) after treatment with DES-based nanofluids.

Note 17: XPS all-element assay data

In this work, XPS (X-ray Photoelectron Spectroscopy) tests were performed on the cathode material particles before and after the separation experiments. In this work, XPS (X-ray Photoelectron Spectroscopy) was performed on the cathode material particles before and after the separation experiments to obtain the elemental species and relative content in the cathode material before and after the separation experiments. The composition and valence of Ni, Co and Mn elements have been analyzed in the text, and the XPS full elemental spectra of the two materials are shown and supplemented in **Fig. S10**:



Fig. S10 XPS spectra of a) original cathode material, b) cathode material after separation experiment.

Note 18: NMR spectra of DESs at different recycle round

Pristine DES: ¹H NMR (400 MHz, DMSO) $\delta = 5.49$ ppm (dt, J = 11.8, 5.0 Hz, 1H, OH in ChCl), 4.58 ppm (s, 6H, OH in EG), 3.81 ppm (s, 2H, CH₂ in ChCl), 3.46 – 3.40 ppm (m, 2H, CH₂ in ChCl), 3.40 – 3.32 ppm (m, 12H, CH₂ in EG), 3.13 ppm (s, 9H, CH₃ in ChCl). ¹³C NMR (400 MHz, DMSO) $\delta = 67.03$ ppm (CH₂ in ChCl), 62.92 (CH₂ in EG), 55.22 ppm (CH₂ in ChCl), 53.58 ppm, 53.24 ppm, 53.20 ppm (CH₃ in ChCl).



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First round: ¹H NMR (400 MHz, DMSO) $\delta = 5.49$ ppm (dt, J = 11.8, 5.0 Hz, 1H, OH in ChCl), 4.58 ppm (s, 6H, OH in EG), 3.81 ppm (s, 2H, CH₂ in ChCl), 3.46 – 3.40 ppm (m, 2H, CH₂ in ChCl), 3.40 – 3.32 ppm (m, 12H, CH₂ in EG), 3.13 ppm (s, 9H, CH₃ in ChCl). ¹³C NMR (400 MHz, DMSO) $\delta = 67.03$ ppm (CH₂ in ChCl), 62.90 (CH₂ in EG), 55.23 ppm (CH₂ in ChCl), 53.28 ppm, 53.25 ppm, 53.21 ppm (CH₃ in ChCl).



Second round: ¹H NMR (400 MHz, DMSO) $\delta = 5.50$ ppm (s, 1H, OH in ChCl), 4.58 ppm (s, 6H, OH in EG), 3.81 ppm (s, 2H, CH₂ in ChCl), 3.42 ppm (s, 2H, CH₂ in ChCl), 3.37 ppm (s, 12H, CH₂ in EG), 3.13 ppm (s, 9H, CH₃ in ChCl). ¹³C NMR (400 MHz, DMSO) $\delta = 66.98$ ppm (CH₂ in ChCl), 62.87 (CH₂ in EG), 55.18 ppm (CH₂ in ChCl), 53.24 ppm, 53.20 ppm, 53.17 ppm (CH₃ in ChCl).



Third round: ¹H NMR (400 MHz, DMSO) $\delta = 5.48$ ppm (s, 1H, O*H* in ChCl), 4.60 ppm (s, 6H, O*H* in EG), 3.80 ppm (s, 2H, C*H*₂ in ChC), 3.59 ppm (s, 2H), 3.42 ppm (s, 2H, C*H*₂ in ChC), 3.37 ppm (s, 12H, C*H*₂ in EG), 3.13 ppm (s, 9H, C*H*₃ in ChCl).¹³C NMR (400 MHz, DMSO) $\delta = 67.08$ ppm (CH₂ in ChCl), 62.94 (CH₂ in EG), 55.28 ppm (CH₂ in ChCl), 53.34 ppm, 53.31 ppm, 53.27 ppm (CH₃ in ChCl).



Fourth round: ¹H NMR (400 MHz, DMSO) $\delta = 5.49$ ppm (s, 1H, OH in ChCl), 4.59 ppm (s, 6H, OH in EG), 3.81 ppm (s, 2H, CH₂ in ChC), 3.69-3.47 ppm (m, 3H), 3.42 ppm (s, 2H, CH₂ in ChC), 3.37 ppm (s, 12H, CH₂ in EG), 3.13 ppm (s, 9H, CH₃ in ChCl).¹³C NMR (400 MHz, DMSO) $\delta = 67.01$ ppm (CH₂ in ChCl), 62.86 (CH₂ in EG), 55.21 ppm (CH₂ in ChCl), 53.28 ppm, 53.24 ppm, 53.21 ppm (CH₃ in ChCl).



Fifth round: ¹H NMR (400 MHz, DMSO) $\delta = 5.48$ ppm (s, 1H, O*H* in ChCl), 4.62 ppm (s, 6H, O*H* in EG), 3.81 ppm (s, 2H, C*H*₂ in ChC), 3.73-3.50 ppm (m, 6H), 3.42 ppm (s, 2H, C*H*₂ in ChC), 3.37 ppm (s, 12H, C*H*₂ in EG), 3.12 ppm (s, 9H, C*H*₃ in ChCl).¹³C NMR (400 MHz, DMSO) $\delta = 67.17$ ppm (CH₂ in ChCl), 63.00 (CH₂ in EG), 55.63 ppm, 55.38 ppm (CH₂ in ChCl), 53.45 ppm, 53.42 ppm, 53.38 ppm (CH₃ in ChCl).



Note 19 NMR spetra of PVDF





Note 20: Half-cell battery liberation battery¹⁷

NMC111/Li half-cells were prepared using a cathode slurry consisting of 70% NMC111 (LiNiMnCoO2, 99.8% purity; Canrd), 20% conductive carbon black (≥99%; Carnoss-tech) and 10% polyvinylidene difluoride (PVDF) binder. A mixture of 100 mg of these three starting materials was dissolved by adding 3 mL of N-methylpyrrolidone (NMP, anhydrous grade, 99.5% purity; Aladdin) dropwise. The mixture was then stirred continuously in a magnetic stirrer (800 r/min for 12 h) to obtain the electrode slurry. The electrode slurry is then uniformly coated on the collector fluid. Afterwards, the electrodes were placed in a vacuum drying oven and dried at 120 °C for 24 h. The dried sheets were intercepted in an MTI MSK-T10 button cell slicer with a 6 mm radius as the positive electrode sheet. NMC111/Li button cells were fabricated from lithium sheets (Zhongneng Lithium Co., Ltd., 0.6 mm thick) in an argon-filled glove box (Mikrouna). The battery was charged and discharged at constant potential on a Neware cycler with 3.4 V as cutoff voltage for 100, 200 and 300 cycles at a rate of 1C. After the last discharge, the spent cells were disassembled and the whole harvested NMC111 electrodes were stirred (600 r/min) in BN nanofluid at 100°C for 24 h, and then dried after extraction to obtain electrode sheets filled with metallic luster.



Fig. S11 (a) Three half-cells, (b) images of half-cell components before assembling which taken in a glovebox under Ar atmosphere, (c) cathode materials recycle using DESs nanofluid, (d) images of Al foil after cathode materials liberation.

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