Journal of The
Electrochemical Society

OPEN ACCESS

3-Methyl-2-oxazolidinone (JEFFSOL® MEOX) as a Substitute Solvent for NMP in Battery Manufacturing

To cite this article: Jean E. Marshall *et al* 2024 *J. Electrochem. Soc.* **171** 090523

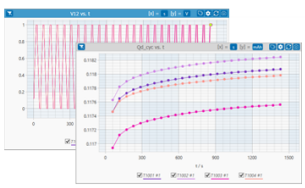
View the article online for updates and enhancements.

You may also like

- A study of ^{239}Pu production rate in a water cooled natural uranium blanket mock-up of a fusion–fission hybrid reactor
Song Feng, Rong Liu, Xinxin Lu et al.
- Approximate processing of the level–level interference in an R-matrix formalism
Elsayed K Elmaghraby
- Cross-section measurements of (*n*, 2*n*) and (*n*, *p*) reactions on ^{124,126,128,130,131,132}Xe in the 14 MeV region and theoretical calculations of their excitation functions
Junhua Luo, , Li Jiang et al.

PAT-Tester-x-8 Potentiostat:
Modular Solution for Electrochemical Testing!

EL-CELL®
electrochemical test equipment



✓ Flexible Setup with up to 8 Independent Test Channels!
Each with a fully equipped Potentiostat, Galvanostat and EIS!

✓ Perfect Choice for Small-Scale and Special Purpose Testing!
Suited for all 3-electrode, optical, dilatometry or force test cells from EL-CELL.

✓ Complete Solution with Extensive Software!
Plan, conduct and analyze experiments with EL-Software.

✓ Small Footprint, Easy to Setup and Operate!
Usable inside a glove box. Full multi-user, multi-device control via LAN.

Scan me!

Contact us:
+49 40 79012-734
sales@el-cell.com
www.el-cell.com

This content was downloaded from IP address 50.124.247.251 on 19/12/2024 at 17:40

Journal of The Electrochemical Society, 2024 **171** 090523

3-Methyl-2-oxazolidinone (JEFFSOL® MEOX) as a Substitute Solvent for NMP in Battery Manufacturing

Jean E. Marshall,[1,z] Victoria White,[2] Ke Zhang,[2] Philip Bellchambers,[1] Jerzy Gazda,[2] Mark Copley,[1] Louis F. J. Piper,[1] and Matthew J. Capener[1]

[1]*WMG, International Manufacturing Centre, Coventry CV4 7AL, United Kingdom*
[2]*Huntsman, The Woodlands, Texas 77381, United States of America*

This work presents 3-Methyl-2-oxazolidinone (JEFFSOL® MEOX) as a substitute solvent for N-methyl-2-pyrrolidone (NMP) in the manufacture of Li-ion batteries. NMP is a good solvent for polyvinylidene difluoride (PVDF, a common binder material), and also has a high boiling point (202 °C), allowing for gradual drying of electrode slurries to form homogeneous coatings. However, NMP has a reprotoxic effect and its use is attracting increasing legislative pressure; it would be advantageous to the battery-making industry to find a more benign alternative. Of the few other solvents that will readily dissolve PVDF, examples such as dimethylformamide are also significantly toxic, so further investigation is necessary to find a credible alternative solvent system. We show that JEFFSOL® MEOX (boiling point 225 °C) is capable of dissolving PVDF at accessible temperatures (40 °C–50 °C), and that at a similar ratio of active material:binder:solvent, both JEFFSOL® MEOX and NMP produce electrode slurries with a viscosity of 5–6 Pa.s at 25 °C and at a shear rate of 10 s^{-1}. Cells formed using JEFFSOL® MEOX-made and NMP-made cathode coatings showed comparable electrochemical performance.

© 2024 The Author(s). Published on behalf of The Electrochemical Society by IOP Publishing Limited. This is an open access article distributed under the terms of the Creative Commons Attribution 4.0 License (CC BY, http://creativecommons.org/licenses/by/4.0/), which permits unrestricted reuse of the work in any medium, provided the original work is properly cited. [DOI: 10.1149/1945-7111/ad77b1]

Manuscript submitted July 19, 2024; revised manuscript received August 28, 2024. Published September 18, 2024.

Supplementary material for this article is available online

Li-ion batteries are an increasingly popular choice for energy storage devices in a range of applications and industrial sectors.[1] As such, there is continued demand for better and more sustainable manufacturing processes in their production.[2,3] The solvent N-methyl-2-pyrrolidone (NMP) is commonly used in Li-ion battery manufacturing, but is associated with significant chemical hazards;[4–7] the focus of this article is on the possibility of replacing NMP with an alternative, 3-Methyl-2-oxazolidinone (JEFFSOL® MEOX).

Conventionally, Li-ion battery electrodes are manufactured by mixing the required chemical components (active material, conductive additive(s), and binder) with a solvent to form a homogeneous slurry. The slurry is then cast on to a thin metallic foil and the solvent evaporates forming the electrode film.[8,9]

The active material is a source of Li^+ ions; for this work, NMC622 was selected as a suitable model material.[10,11] The conductive component is most often a carbon additive, and the binder is conventionally a polymer that will promote both cohesion within the film and adhesion between the film and the current collector foil. In addition to good chemical compatibility with the powders and foil, the binder must be chemically inert and thermally stable, thus capable of withstanding the relatively extreme conditions inside the battery without significant degradation. Fluoropolymers are desirable materials for this type of application because their strong internal carbon-fluorine bonds lend them a high level of durability and inertness; indeed, the fully fluorinated polymer polytetrafluoroethylene (PTFE) is not soluble in any commonly used organic solvent (though it may be soluble in some perfluorinated solvents at an elevated temperature and pressure). PTFE is therefore not a useful binder material for 'wet' electrode processing, whereas the partially fluorinated fluoropolymer polyvinylidene difluoride (PVDF) has some solubility in a few solvents,[12] and is now a common binder in Li-ion batteries. The industry standard solvent for PVDF is NMP, which will readily dissolve PVDF at room temperature to form solutions that are stable for some time (ideal for making slurries for battery electrodes). However, NMP presents considerable health hazards including reproductive toxicity and skin irritation.[13]

Since the use of NMP is likely to become more restricted by legislation in future,[14] it would be advantageous to replace the NMP/PVDF carrier system with a more benign alternative. Switching to aqueous systems would be favourable in terms of both lower expense and lower toxicity,[15,16] but this requires a different binder to be used (as PVDF is insoluble in water) and is also impractical for many cathode chemistries because the cathode active materials are water sensitive. A less-toxic organic solvent for PVDF would be highly convenient, since it could directly replace NMP in current manufacturing processes and would allow manufacturers to keep the desirable properties of PVDF as a binder (such as good durability and adhesion to the components of the electrode).[17] However, such a solvent is difficult to find due to the relative insolubility of PVDF in most other organic solvents. Possible alternative solvents such as dimethylformamide (DMF)[18] and dimethylacetamide (DMAc) also present hazards to health and therefore are not suitable benign substitutes.

Attempts have been made to rationalize the low solubility of PVDF in organic solvents. In 1988, Bottino et al. listed a series of solvents and non-solvents for PVDF, and generated Hansen solubility parameters for PVDF.[19] In that early study, of fifty solvents studied only eight showed affinity for PVDF, and these were all polar aprotic compounds. Of these, the least toxic examples were dimethylsulfoxane (DMSO) and triethylphosphate (TEP). Studies attempting to use these solvents in electrode manufacturing have been published in the last few years.

DMSO has been used previously as a PVDF solvent in the manufacture of PVDF membranes.[20,21] Though associated with some adverse effects in humans,[22] it is not considered hazardous to reproductive health in the same way as NMP and is therefore a potential candidate as a more benign replacement. A 2020 study investigated DMSO as an alternative solvent for electrode manufacture.[23] The authors found DMSO to be a promising candidate solvent, with the ability to dissolve PVDF at the required concentration and to form electrodes. PVDF was dissolved in DMSO at 70 °C for the purposes of this study but the resulting electrode slurries could be coated at room temperature. Some questions remain about the stability of DMSO at elevated temperatures and whether sulfur-containing compounds could be deleterious to the battery performance.

TEP has also been used as a "more benign" solvent to dissolve PVDF in membrane manufacture.[24–28] It carries no hazard labelling

[z]E-mail: Jean.Marshall@warwick.ac.uk

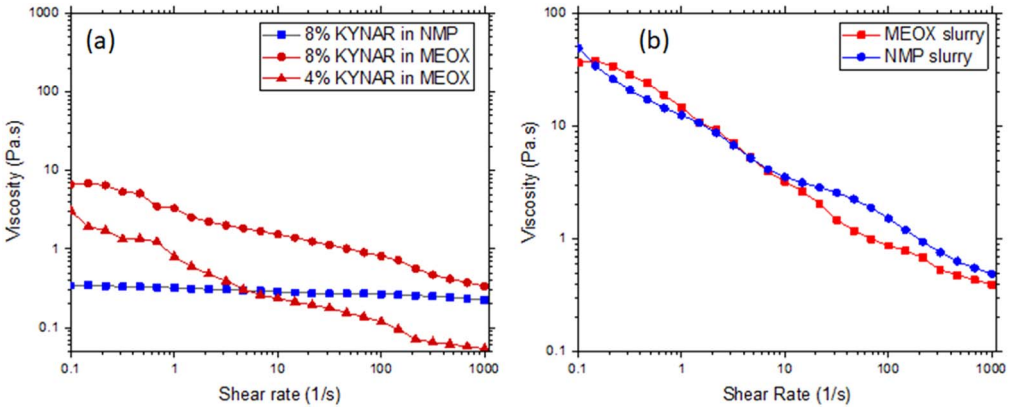


Figure 1. (a) Graph to show viscosity vs shear rate for 4 wt% and 8 wt% suspensions of KYNAR HSV900 in JEFFSOL® MEOX. The same graph for 8 wt% KYNAR in NMP is shown for comparison. (b) Graph to show viscosity vs shear rate for two cathode slurries, where the composition of the slurries is the same except for solvent type.

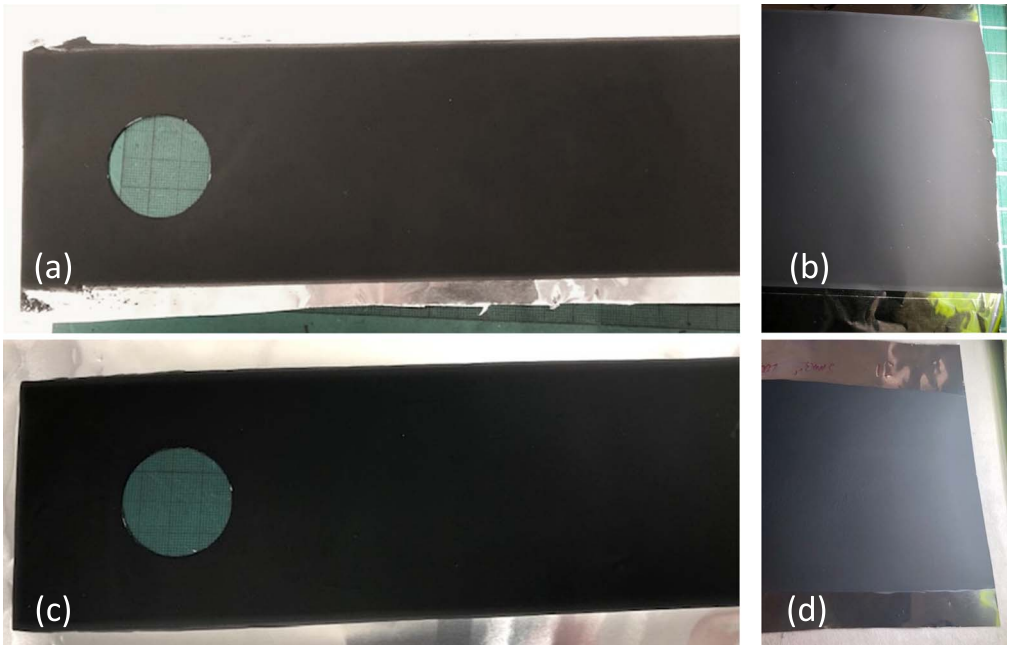


Figure 2. Photographs to show coatings made from the JEFFSOL® MEOX and NMP based slurries. (a) A 200 GSM uncalendered coating formed from a JEFFSOL® MEOX slurry. (b) A JEFFSOL® MEOX-formed coating photographed under illumination, to show that no visible aggregates are present. (c) A 200 GSM uncalendered coating formed from an NMP slurry. (d) An NMP-formed coating photographed under illumination to show that no visible aggregates are present.

related to reproductive health risks. TEP cannot necessarily be regarded as a "green" (i.e. sustainable) solvent, since global phosphorus supplies are limited, but it is much less hazardous to human health than NMP. TEP is a weaker solvent for PVDF than NMP, and articles dealing with PVDF-TEP systems show that, as with DMSO, TEP must be heated to dissolve PVDF. Nonetheless, a recent study investigated TEP as an alternative to NMP in cathode manufacture;[29] the authors dissolved PVDF in TEP at 60 °C and were able to mix and coat electrode slurries at room temperature.

Some polar-aprotic compounds have become of interest as "greener" PVDF solvents much more recently than DMSO and TEP. Notable candidates include cyrene™ (dihydrolevoglucosenone) and γ -valerolactone. These solvents show much lower toxicity than PVDF, and bio-based synthesis routes are available for them.[30–32] However, these solvents require considerable heating to dissolve PVDF and may require the PVDF mix to be kept at elevated temperatures for slurries/coatings. This has implications for adjustments that would need to be made to manufacturing processes in order to accommodate these solvents. A study on electrode fabrication using cyrene™[33] showed that the PVDF-cyrene™ mixes require temperatures of 80 °C for mixing, and coatings made using these mixes showed poor adhesion to the metal current-collector. A similar study using γ-valerolactone as the solvent[34] also showed concerns about adhesion; binder dissolution and slurry making were carried out at 60 °C before coating at room temperature.

In this work, we present 3-Methyl-2-oxazolidinone (JEFFSOL® MEOX) as a suitable solvent with which to replace NMP. While NMP has GHS classification for 1B for reprotoxicity, MEOX was found to be not mutagenic according to the Ames test by OCED testing. JEFFSOL® MEOX is currently available from Huntsman for a cost approximately 20%–30% higher than the cost of NMP, though its price is expected to reduce in the future. We show herein that JEFFSOL® MEOX can dissolve battery-grade PVDF at accessible temperatures in the range of 40 °C–50 °C and be used to form slurries and coatings in a similar manner to NMP.

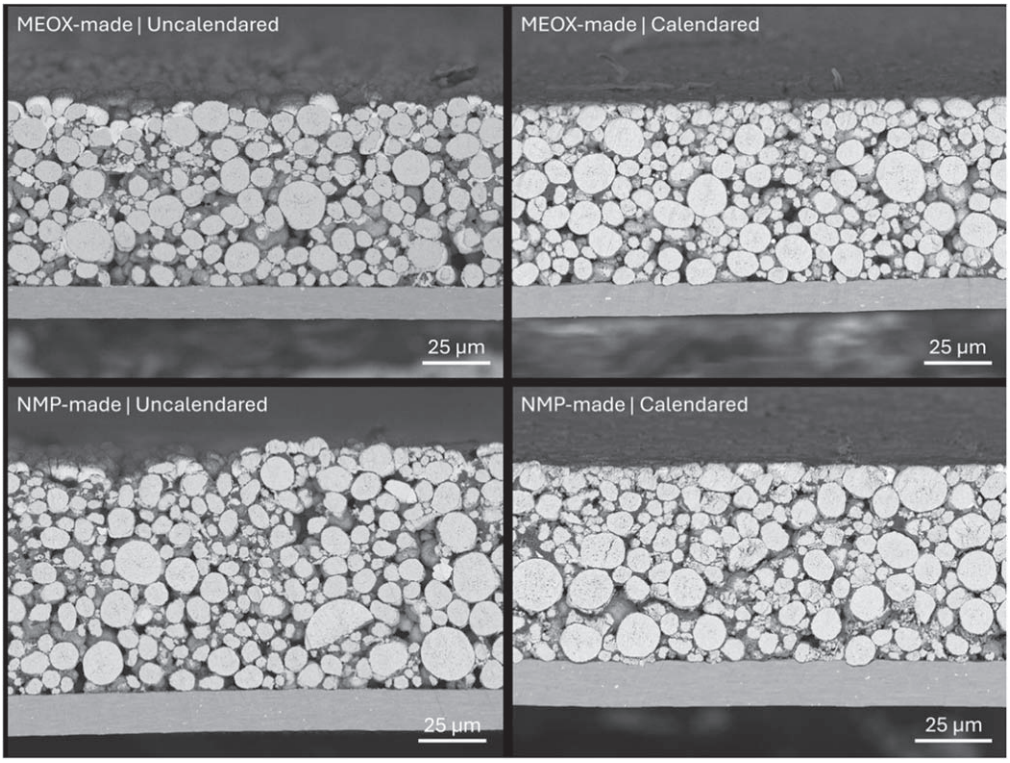


Figure 3. Cross-sectional SEM imaging of uncalendered electrodes (200 GSM) and calendered electrodes (200 GSM, density >3.2 g/cm^3) made with JEFFSOL® MEOX and NMP.

Table I. Characterisation data of cells made using JEFFSOL® MEOX-based and NMP-based coatings at the cathode. For each solvent coating-type and coatweight, 3 cells are summarized; the average coatweight and density is given, along with average charge, discharge and first-cycle efficiency (FCE) for the formation cycle.

				Density		Charge		Discharge		FCE	
		Coat Weight (GSM)		(g/cm^3)		(mAh/g)		(mAh/g)		(%)	
Solvent	No. cells	Avg.	Std Dev.	Avg.	Std Dev.	Avg.	Std Dev.	Avg.	Std Dev.	Avg.	Std Dev.
JEFFSOL® MEOX	3	208.1	6.1	3.1	0.2	195.6	1.0	180.6	1.2	92.3	0.3
JEFFSOL® MEOX	3	96.3	1.3	3.3	0.3	197.1	2.6	183.4	0.8	92.4	0.8
NMP	3	208.8	3.4	3.2	0.0	196.4	2.1	181.2	1.4	91.9	1.0
NMP	3	97.7	1.1	3.6	0.1	196.4	2.3	182.5	2.1	92.6	0.3

Experimental

Materials.—NMC622 (BASF), Kynar HSV 900 PVDF (Cambridge Energy Solutions), C65 Carbon black (Imerys), JEFFSOL® MEOX (Huntsman), NMP (Merck), 1 M LiPF6 EC:EMC (3:7 vol%) +1 wt% VC electrolyte (Solvionic) and current collector (15 µm Al, Avocet Precision Metals) were used as received.

Electrode and cell fabrication.—An 8 wt% mixture of PVDF in the required solvent (JEFFSOL® MEOX or NMP) was obtained using an overhead mixer (Precimix 2.5, Buhler). In the case of NMP, PVDF can dissolve in the solvent at room temperature, whereas the JEFFSOL® MEOX mixture had to be heated to 40 °C–50 °C in order to achieve good dissolution. Mixtures were degassed under vacuum at room temperature.

In the subsequent stage, a 250 mL vessel was charged with NMC622 and C65, which were then homogenised by a ARE-250 (Thinky) planetary-centrifugal-mixer. PVDF solution (in either NMP or JEFFSOL® MEOX) was then added such that the final ratio (by mass) of NMC, C65 and PVDF in the mixture would be 96:2:2, followed by a subsequent mixing stage. In subsequent mixing stages, solvent was added until the final solid content of the slurry was 70%.

The slurry was cast on to aluminium foil using an automated drawdown coater with vacuum bed and fixed gap applicators (COATMASTER 510,Erichsen) and subsequently dried on a hot plate, held at 80 °C for NMP-based slurries and 120 °C for JEFFSOL® MEOX-based slurries. Samples of each coating were measured to check coatweight. Coatings were then calendered at 85 °C (Innovative Machine Corp). Cathode electrodes (14.8 mm diameter) were cut from each coating using a manual punch (El-cell) and assembled into half-coin cells (Hoshen) with lithium (China Energy Lithium) at the anode, using Celgard® H1609 as the separator and 80 µl of electrolyte. Cell assembly was performed in an Argon glovebox (0.1 ppm H_2O, 0.1ppm O_2).

Electrochemical testing was carried out in a 25 °C Binder using BioLogic BCS-805 channels. A test program was run with a formation cycle at CC-CV C/20, I < C/50, 4.3–2.6 V, followed by 5 conditioning cycles at C/5. Rate performance was then evaluated at a series of C rates between C/5 and 5 C, before the cell was subjected to 30 cycling steps at C/2 to evaluate long-term performance.

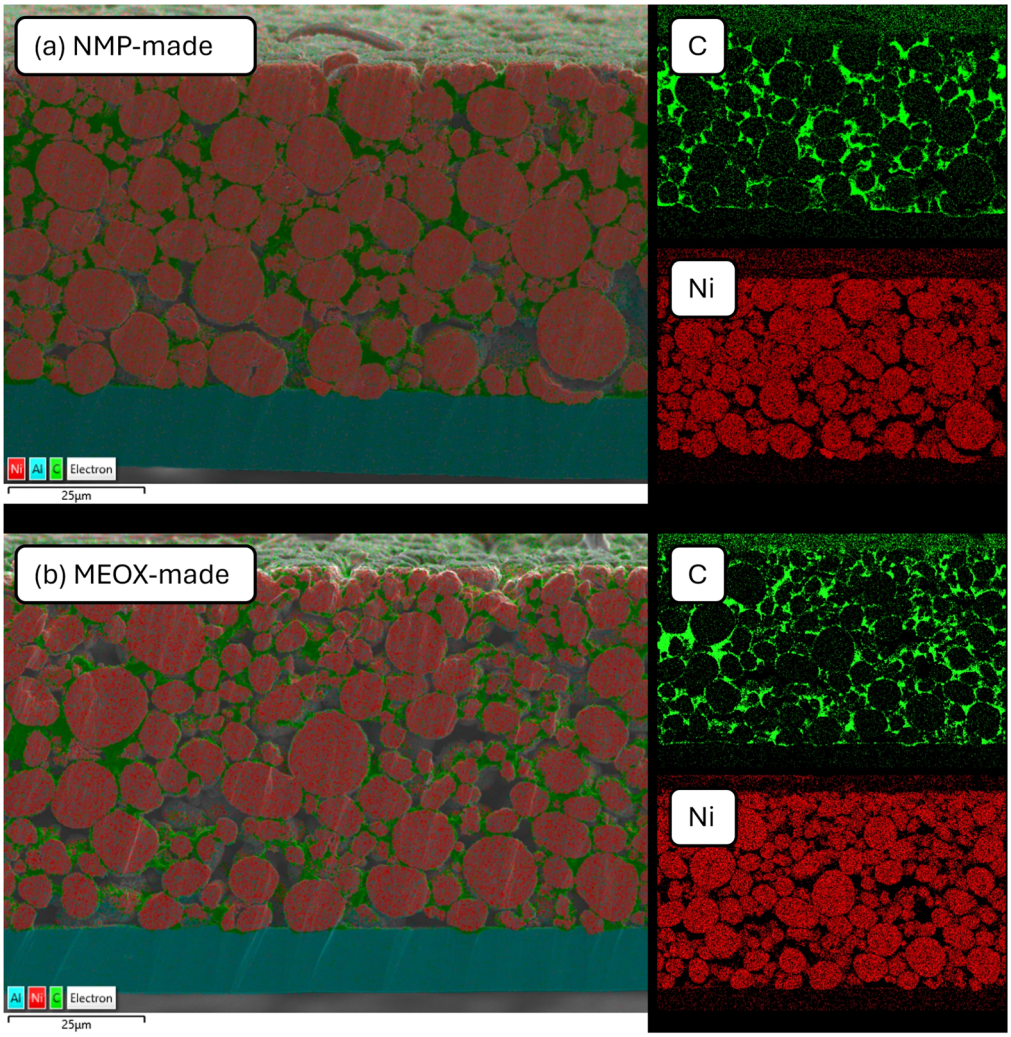


Figure 4. SEM-EDX maps of cross sections of calendered samples for (a) NMP-made and (b) JEFFSOL® MEOX-made cathodes.(200 GSM, density >3.2 g cm^{-3}).

Characterisation.— *SEM.*—Cross-sections of electrodes before and after calendering were prepared using a Hitachi IM4000plus broad beam (Ar^{+}) ion miller. Milling was performed at 6 kV for 100 min with 40° stage rocking. The sections were stored in an inert glove box but transferred in air to a Tescan Clara scanning electron microscope (SEM) equipped with a EDX detector (Oxford Instruments). Back Scattered Electron (BSE) images were acquired at 5 kV using a dedicated scintillator-type detector which detects the wide-angle back-scattered electrons and gives both topographical and elemental contrast. EDX mapping was performed using the Aztec software package and a pixel dwell time of 100 µs (3 frames).

Adhesion testing was carried out using a the Z005 universal testing machine and Z-direction fixture (Zwick Roell), following the method described by Haselrieder et al.[35] Resistivity measurements were carried out on the electrodes using a HIOKI RM2610. Rheology measurements were carried out using a 40 mm grit-blasted parallel plate with a 500 µm gap at 25 °C, using a RH20 rheometer (TA instruments) from 0.1 to 1000 s^{-1}.

Results and Discussion

KYNAR HSV 900 was selected as a high molecular-weight, battery grade PVDF-based binder. As with other grades of PVDF, it is readily soluble in NMP at a concentration of 8 wt%. Using an overhead mixer, it was shown not to dissolve in JEFFSOL® MEOX at room temperature, but to dissolve readily if the JEFFSOL® MEOX is heated to 40 °C–45 °C. This mixture remained liquid and pourable over the course of several hours but would gel if left to stand overnight. This gelation was not thermoreversible. JEFFSOL® MEOX mixtures must therefore be used within their window of operation, but we must note that this behaviour is in contrast with previous published work by other authors showing that, for example, Cyrene™ solutions of PVDF must be heated to 80 °C to achieve dissolution, and electrode slurries based on Cyrene™ must be kept at elevated temperatures to achieve coating.[33] JEFFSOL® MEOX therefore offers a significant potential benefit in being able to be processed at a lower temperature.

Figure 1a shows how the viscosity of 8 wt% and 4 wt% solutions of PVDF/JEFFSOL® MEOX vary with shear rate. For comparison, data is also shown for an 8 wt% solution of PVDF in NMP. From these data, it is clear that the JEFFSOL® MEOX solutions show increased shear-thinning behaviour compared to the NMP solution, and overall higher viscosity for the same shear rate. However, even at 8 wt% binder loading the viscosity of the JEFFSOL® MEOX solution is still processable.

Following preparation of the binder/solvent mixtures, slurries were created such that the ratio (by mass) of the solid materials in the slurry would be 96:2:2 NMC622:C65:PVDF. The solid content of each slurry was adjusted to 70%. As shown in Fig. 1b, an NMC/solvent slurry containing a solid content of approx. 70% gives similar rheological behaviour whether the solvent used is NMP or JEFFSOL® MEOX. We therefore assert that if the JEFFSOL®

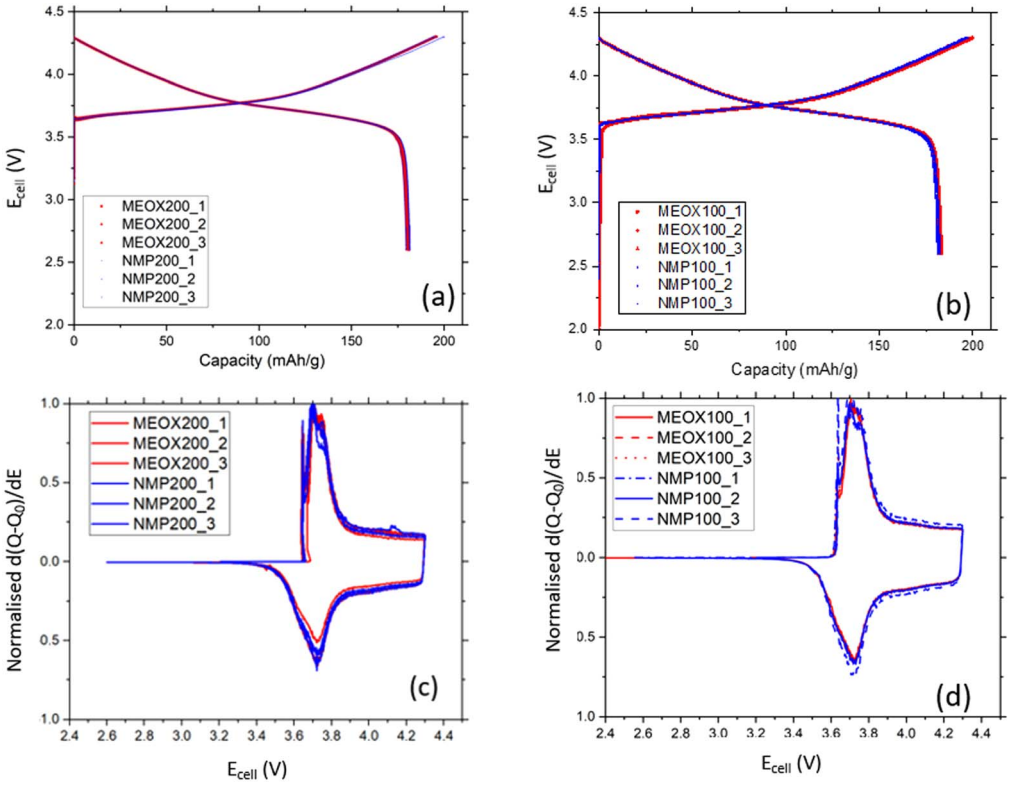


Figure 5. Ecell vs capacity during the formation cycle of cells containing cathodes with (a) higher coatweight (200 GSM) and (b) lower coatweight. (100 GSM). dQ/dE data for the formation cycle of cells with (c) higher coatweight and (d) lower coatweight. dQ/dE data are normalized to the highest value. Data from cells containing cathode made with JEFFSOL® MEOX are coloured in red, while those made with NMP are coloured in blue. The formation cycles are notably rather similar for both JEFFSOL® MEOX-made and NMP-made cells.

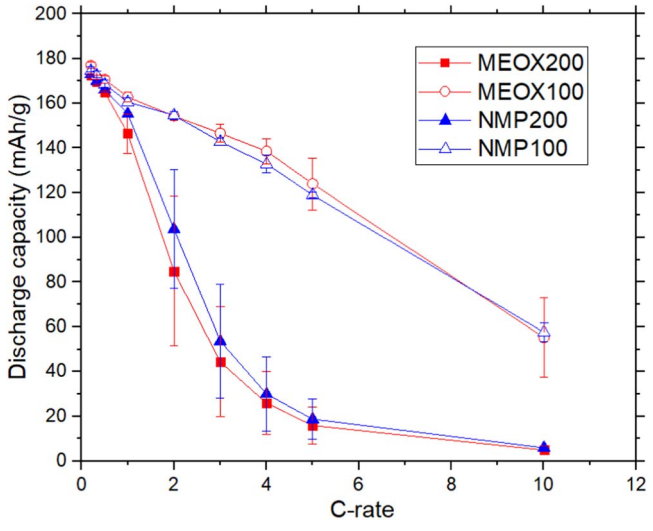


Figure 6. Variation in discharge capacity with C-rate, for cells made with JEFFSOL® MEOX and NMP, and at 2 different coatweights for each solvent. As expected for this cell chemistry, capacity retention at high C-rates is higher for the lower coatweights; however, there is little difference in performance between the JEFFSOL® MEOX- and NMP-prepared cathodes.

MEOX solution is used within the first few hours after it is made, then its rheological behaviour is similar to that of NMP.

Following mixing of the slurries, each slurry was coated on to an aluminium foil to give homogenous coatings, as shown in Fig. 2. The figure shows photographs of coatings formed using JEFFSOL® MEOX and using NMP; the photographs taken under illumination show difference in the coating quality before or after drying with no visible aggregates and inhomogeneities for either JEFFSOL® MEOX or NMP based coatings. Coatings were prepared at two different coatweights (approx. 100 and 200 GSM (grams/m^2)); these loadings were selected to reflect industry standards for cathode coatings) in order to compare the electrochemical performance of thicker and thinner cathode coatings.

Following removal of the solvent, coatings were calendered to a density of >3.2 g cm^{-3}. Resistivity measurements were taken from calendered and uncalendered coatings (see supplementary information). The interface resistance is particularly important for these coatings and this was shown to decrease to $< 0.2\ \Omega$ cm^2 after calendering, for both sets of coatings. This result is in line with previous studies on the resistivity of calendered and uncalendered coatings.[36]

Cross-sections were made of each sample, to allow characterization by SEM. Figure 3 contains SEM images of cross sections of JEFFSOL® MEOX-made and NMP-made electrodes. It is clear from these images that the electrodes appear rather similar, in spite of the two different solvents used in their manufacture. This point is supported further by the EDX maps in Fig. 4, which demonstrate an even distribution of carbon across the electrode for both JEFFSOL® MEOX-made and NMP-made electrodes, suggesting that both solvents are equally capable holding the PVDF binder in solution during the time period required for electrode coating. The JEFFSOL® MEOX coatings will require a higher drying temperature for a comparable drying time, as indicated in the evaporation profiles for both solvents (see supplementary information).

Adhesion tests also indicated similar performance for NMP-made and JEFFSOL® MEOX-made coatings. A peel-test for each type of coating gave a maximum tensile force of 1132 ± 33 kPa for the JEFFSOL® MEOX-made coatings and 1037 ± 70 kPa for the NMP-made coatings, with highly similar test profiles for each type of coating (see supplementary information). These tensile forces are consistent with literature values for this type of coating, which vary from 500–1200 kPa depending on formulation, density, and thickness.[35,37]

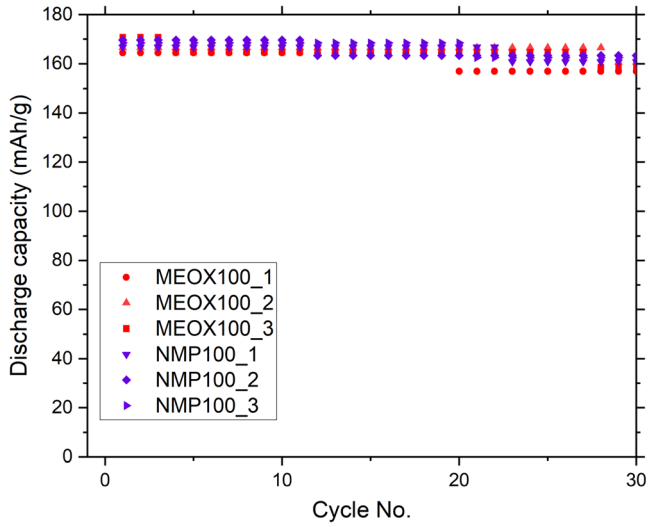


Figure 7. Cycling behaviour for 6 cells, all with coatweights around 100 GSM. 3 of these cells were made using the JEFFSOL® MEOX-made cathode, and the other 3 with the NMP-made cathode. Cycling was carried out at 0.5 C–0.5 C.

Electrochemical characterization.—Electrodes were cut manually from the coatings and assembled into half-cells with Li at the anode. For each coating type (JEFFSOL® MEOX-formed and NMP-formed) and coatweight, data was averaged from 3 cells; Table I tabulates the average coatweight and density for each cell type, along with the charge/discharge and efficiency for the formation cycle (rate 0.05 C). All 4 groups of cell give similar formation cycle data, with first-cycle efficiencies (FCE) around 92%. As shown in Fig. 5, the shape of the Ecell vs capacity curves, and the dQ/dE data, also have a similar appearance for both JEFFSOL® MEOX-made and NMP-made cathodes during the formation cycle with no additional peaks. These results indicate that the use of JEFFSOL® MEOX allows the formation of a good electrode coating and has no negative impact on the electrochemistry of the cathode.

Figure 6 compares the rate performance of JEFFSOL® MEOX- and NMP-processed electrodes, at both the higher and lower coatweights. It is clear from these data that cells made with the lower coatweight cathode have higher capacity retention at the higher C-rates; also that there is little difference between the JEFFSOL® MEOX-processed and NMP-processed cathodes. This conclusion is also borne out by the cycling data in Fig. 7. This shows the cycling of the cells after the rate testing steps shown in Fig. 6. We can draw no conclusions about cycling data at higher numbers of cycling steps; this partly because degradation processes at the lithium anode itself tend to limit cycle life in half-cells,[38–40] and partly because by the time this cycling stage started the cells had already undergone several cycling stages for conditioning and rate testing. However, from these data we can conclude that the electrochemical behaviour is similar for both the NMP-made and MEOX-made cells. To draw further conclusions about the cycling behaviour, testing in full cells would be required.

Conclusions

This study demonstrates that JEFFSOL® MEOX can dissolve PVDF at accessible temperatures (40 °C–50 °C) and that this solution can remain liquid and usable at room temperature for several hours after initial mixing. We have demonstrated the creation of electrode slurries using PVDF binder dissolved in both JEFFSOL® MEOX and NMP. Half-cells constructed from JEFFSOL® MEOX-made and NMP-made electrodes showed similar electrochemical behaviour. These results show that it can be feasible to make cells using JEFFSOL® MEOX as an NMP replacement, with no loss of battery performance (at half-cell coin cell level). However, there are still challenges to be overcome in using this process; firstly, the tendency of JEFFSOL® MEOX/PVDF solutions to gel on a timescale of 12–18 h, meaning that coating has to be completed within a processing window, and secondly the higher boiling point of JEFFSOL® MEOX such that JEFFSOL® MEOX-made coatings must be dried at a higher temperature than NMP-made coatings.

Acknowledgments

We would like to acknowledge Robert Duncan and Jaswinder Hothi from WMG, for assistance with adhesion test measurements; also Shaun Dixon and the rest of the technical support team from the Energy Innovation Centre in Warwick, for valuable assistance in the lab. This study was funded by Huntsman.

ORCID

Jean E. Marshall https://orcid.org/0000-0001-7617-4101
Philip Bellchambers https://orcid.org/0000-0002-3250-5682
Mark Copley https://orcid.org/0000-0002-6435-8680
Louis F. J. Piper https://orcid.org/0000-0002-3421-3210
Matthew J. Capener https://orcid.org/0000-0003-3370-9561

References

1. N. Nitta, F. Wu, J. T. Lee, and G. Yushin, *Mater. Today*, **18**, 252 (2015).
2. Y. Yang et al., *Energy Storage Mater.*, **36**, 186 (2021).
3. A. Örüm Aydin et al., *Batteries*, **9**, 555 (2023).
4. N. Ulrich et al., *Int. Arch. Occup. Environ. Health*, **91**, 1073 (2018).
5. M. Bader, R. Wrbitzky, M. Blaszkewicz, and C. Van Thriel, *Arch. Toxicol.*, **81**, 335 (2007).
6. http://ec.europa.eu/health/scientific_committees/consumer_safety/index_en.htm Accessed on 13/09/2024.
7. G. M. Solomon, E. P. Morse, M. J. Garbo, and D. K. Milton, *J. Occup. Environ. Med.*, **38**, 705 (1996).
8. W. B. Hawley and J. Li, *J. Energy Storage*, **25**, 100862 (2019).
9. H. Zheng, R. Yang, G. Liu, X. Song, and V. S. Battaglia, *J. Phys. Chem. C*, **116**, 4875 (2012).
10. N. Mohamed and N. K. Allam, *RSC Adv.*, **10**, 21662 (2020).
11. A. Manthiram, *Nat. Commun.*, **11**, 1 (2020).
12. J. E. Marshall et al., *Polymers (Basel)*, **13**, 1354 (2021).
13. https://www.merckmillipore.com/GB/en/product/msds/MDA_CHEM-806072?Origin=PDP (accessed on 13/09/2024).
14. J. Sherwood, T. J. Farmer, and J. H. Clark, *Chem*, **4**, 2010 (2018).
15. J. Li et al., *iScience*, **23**, 101081 (2020).
16. R. Sahore et al., *ACS Sustain Chem. Eng.*, **8**, 3162 (2020).
17. X. Zhong et al., *Appl. Surf. Sci.*, **553**, 149564 (2021).
18. R. Sliz, T. Hu, J. Valikangas, U. Lassi, and P. Vilmi, *IEEE 16th Nanotechnology Materials and Devices Conference (NMDC)*, **16**, 1–5 (2021).
19. A. Bottino, G. Capannelli, S. Munari, and A. Turturro, *J. Polym. Sci. B Polym. Phys.*, **26**, 785 (1988).
20. F. Russo et al., *Membranes (Basel)*, **10**, 36 (2020).
21. A. Figoli et al., *Green Chem.*, **16**, 4034 (2014).
22. B. K. Madsen, M. Hilscher, D. Zetner, and J. Rosenberg, *F1000Res*, **7**, 1746 (2019).
23. M. Wang, X. Dong, I. C. Escobar, and Y.-T. Cheng, *ACS Sustain Chem. Eng.*, **8**, 11046 (2020).
24. T. Marino, S. Blefari, E. D. Nicolò, and A. Figoli, *Green Processing and Synthesis*, **6**, 295 (2017).
25. J. Chang, J. Zuo, L. Zhang, G. S. O'Brien, and T. S. Chung, *J. Memb. Sci.*, **539**, 295 (2017).
26. H. R. Yang, Y. H. Huang, C. F. Wang, and T. S. Chung, *Desalination*, **566**, 116934 (2023).
27. N. Arahman, S. Mulyati, and A. Fahrina, *Mater. Res. Express*, **6**, 066419 (2019).
28. T. Marino, F. Russo, and A. Figoli, *Membranes (Basel)*, **8**, 71 (2018).
29. C. Chen, V. Reddy Tatagari, H. Lin, and L. Shaw, *Journal of Energy Chemistry*, **78**, 240 (2023).
30. I. T. Horváth, H. Mehdi, V. Fábos, L. Boda, and L. T. Mika, *Green Chem.*, **10**, 238 (2008).
31. J. E. Camp, *ChemSusChem*, **11**, 3048 (2018).
32. N. A. Stini, P. L. Gkizis, and C. G. Kokotos, *Green Chem.*, **24**, 6435 (2022).
33. H. Zhou, B. Pei, Q. Fan, F. Xin, and M. S. Whittingham, *J. Electrochem. Soc.*, **168**, 040536 (2021).
34. V. R. Ravikumar et al., *ACS Appl. Energy Mater.*, **4**, 696 (2021).
35. W. Haselrieder et al., *Int. J. Adhes. Adhes.*, **60**, 1 (2015).
36. C. Seidl, S. Thieme, M. Frey, K. Nikolowski, and A. Michaelis, *Batteries*, **10**, 105 (2024).
37. C. Schilcher, C. Meyer, and A. Kwade, *Energy Technology*, **4**, 1604 (2016).
38. R. C. McNulty et al., *J. Mater. Chem. A Mater.*, **11**, 18302 (2023).
39. B. Rowden and N. Garcia-Araez, *Energy Reports*, **7**, 97 (2021).
40. C. Heubner et al., *Batter Supercaps*, **4**, 1310 (2021).