On Electric Conductivity of Greases

Akepati Bhaskar Reddy (✉ abreddy@kth.se)

KTH Royal Institute of Technology: Kungliga Tekniska Hogskolan <https://orcid.org/0000-0002-2716-8810>

Faiz Ullah Shah

Lulea University of Technology: Lulea Tekniska Universitet

Johan Leckner

Axel Christiernsson International AB

Mark Rutland

KTH Royal Institute of Technology Department of Chemistry: Kungliga Tekniska Hogskolan Institutionen for kemi

Sergei Glavatskih

KTH Royal Institute of Technology: Kungliga Tekniska Hogskolan

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On electric conductivity of greases

Akepati Bhaskar Reddy^{1*}, Faiz Ullah Shah², Johan Leckner^{3,1}, Mark W. Rutland^{4,5,6} and Sergei Glavatskih^{1,6,7}

^{1*}Department of Machine Design, KTH Royal Institute of Technology, Stockholm, 100 44, Sweden.

²Chemistry of Interfaces, Luleå University of Technology, Luleå, 97 187, Sweden.

³Axel Christiernsson International AB, Nol, 449 11, Sweden.

⁴Division of Surface Chemistry and Corrosion Science, Department of Chemistry, KTH Royal Institute of Technology, Stockholm, 100 44, Sweden.

⁵Laboratoire de Tribologie et de Dynamique des Systèmes, Ecole Centrale de Lyon, Ecully, 69134, France.

⁶School of Chemistry, University of New South Wales, Sydney, 2052, NSW Australia.

⁷Department of Electromechanical, Systems and Metal Engineering, Ghent University, Ghent, 9052, Belgium.

*Corresponding author(s). E-mail(s): abreddy@kth.se;

Abstract

The development of new lubricant systems compatible with modern e-drive conditions, including stray voltages and currents, has gained momentum in recent years. One of the strategies being widely evaluated to achieve these goals is the development of conductive lubricants. Greases, however, have a complex architecture which renders measurement and analysis of their electrical properties challenging; this has severely limited the implementation of the strategy for this class of materials. In this work, a measurement methodology implementing electrochemical impedance spectroscopy to evaluate the electrical properties of grease samples is introduced. Compared to the commonly used conductivity meters, this method, through its multi-frequency alternating current (AC) impedance measurement approach, is able to effectively distinguish the separate contributions of the bulk and the sample-electrode interface to the measured electrical response. Thus, the bulk conductivity of a range of greases with different electrical properties could be obtained independently of their interfacial behavior at the electrode. The temperature dependent bulk conductivity of lithium complex grease doped with different ionic liquids and non-ionic solids (copper and graphite) has accordingly been evaluated and the rather different conductivity mechanisms explained. Bulk ionic conductivity of neat ionic liquids has also been measured using a liquid sample specific electrochemical cell, which aids in the interpretation of the ionic liquid based greases.

Keywords: grease, ionic liquid, electric conductivity, impedance spectroscopy

1 Introduction

With increasing electric mobility, lubrication requirements are evolving beyond the basic

requirements of reducing friction and wear, heat removal, fatigue protection and corrosion prevention [1]. Extensive investigations have been reported for evaluating bearing damages due to electric current passage, circulating currents and electrostatic discharges [2–4]. Lubricating greases, which are generally formulated as semi-solid dispersion of a thickener in base oil with some additives, have been shown to undergo chemical changes and decomposition in the presence of electric fields leading to corrosion of steel bearing surface [5]. For the elimination or mitigation of these issues, several measures, such as insulating tribo-surfaces, grounding machine components, and using conductive lubricants, can be applied [4].

The use of conductive lubricants is expected to prevent voltage build-up across the lubricating film, thereby preventing electric discharge breakdowns [6, 7]. Thus, the development of bespoke conductive lubricants could ensure longevity and reliability of machine components exposed to electrical currents and voltages. Several approaches have been reported towards the development of conductive lubricating greases through the use of conductive materials either as grease matrix or additives; graphite or nano-carbon [8–10], carbon nano-tubes [11–14], and metal and metal oxides (copper, silver, antimony doped tin-oxide) [15]. Although these options provide conductive pathways, some of them, (e.g. addition of metal particles to greases [16]), result in deterioration of lubrication and wear resistant properties.

Conductive greases based on ionic liquids (ILs), generally defined as organic salts existing as liquid below 100°C [17], have recently attracted a lot of interest. Along with generally exhibiting lubricant friendly properties, like non-volatility, non-flammability, low melting point, ILs have been reported to enhance lubrication both as a neat lubricant or as an additive [18]. They are also expected to provide electrically conductive pathways through the presence of dissociated ions. In development of electrically conductive greases, ILs have been used as either the base oil [19, 20] or as additives [21–24]. Greases with IL additives have also shown to reduce friction and wear [21, 22, 24–28].

One of the complications on the path to designing new conductive lubricating greases is to characterize their conductive properties. Although

there are specific standards for measuring conductivity of liquid samples, DIN 55667 for paints and varnishes and ASTM D2624 for aviation and distillate fuels, to our knowledge there is no standardized method for measuring electric conductivity of greases. Most of the studies reported for conductive greases measure conductivity using conventional conductivity meters [9, 12–15, 19–24, 29]. These methods, while being fast, inexpensive, and non-destructive, require either instantaneous measurement or alternating current (AC) measurement to minimize errors due to ion depletion and polarization. These methods are therefore not suitable for samples that exhibit complex impedance (frequency-dependent) and/or contributions from interfacial structuring processes (like diffusion) at the electrode-sample interface. Electrochemical impedance spectroscopy (EIS), which is a multi-frequency AC electrochemical measurement technique, is an efficient method to evaluate the electrical characteristics of material-electrode systems [30]. The multi-frequency approach permits the separation of the contributions from the bulk matrix and the interfacial structuring (and dynamic processes) to the conductivity. The bulk properties and electrode effects are generally associated with the high end and low end of the frequency range respectively. The impedance data can be fitted to equivalent electric circuits which act as fingerprints of the sample and help extract valuable information about the electrical properties of the bulk material as well as of the electrode-sample interface [31, 32].

EIS has been successfully applied in various complex systems, e.g., cement and concrete [33–38]; ceramics and composites [39]; and polymeric electrolytes [40]. It has also been used to measure the conductivity and capacitance of lubricating oils with additives [41].

In this study we present the application of EIS to evaluate the electrical conductivity of greases doped with various ionic liquids and solid additives. The greases are based on lithium complex, one of the most commonly used type of thickener [42].

2 Methods and materials

2.1 Greases

A model lithium complex grease with a non-polar base oil (PAO-10) was produced using the method described elsewhere [43, 44]. A small amount of adipate ester (6.3%) was added to the base oil to ensure rapid and complete saponification reaction through increased base oil polarity. Table 1 gives an overview of the grease properties. The grease was produced in a pilot scale reactor as previously described [43].

Table 1: Base grease properties

Property	LiX-10
Thickener type	Lithium complex
Thickener content [%]	20.6%
Base oil composition [wt%]	93.7 PAO-10 + 6.3 Adipate ester
Base oil viscosity [mm ² s ⁻¹] @ 40°C	61.4
@ 100°C	9.4
NLGI grade	2

Five ionic liquids (ILs) were selected for this study. Four of these ILs were based on phosphonium cation decorated with long alkyl chains while the remaining one was based on an imidazolium cation. The molecular structures, designations and the respective sources of the ILs are given in Table 2. Promising tribological gains offered by P-BOB, P-BMB and P-DCA when used as additives to greases, as reported by Ploss et al. [26], formed the basis for their selection. P-BEHP was selected because of its high popularity in tribological research [47]. In addition to these, EMIM-TFSI was studied as comparison because of its reported conductivity [48] being multiple orders of magnitude higher than that reported for the orthoborate ILs, P-BOB and P-BMB [45, 46]. This choice of ILs provides a big range of additive conductivity to evaluate the proposed methodology.

To evaluate the viability of the methodology for non-ionic additives as well, two grease samples with copper powder (Rogal Copper GK 0/50, Schlenk Metallic Pigments GmbH) and graphite powder (ENSACO 250G, Imerys Graphite & Carbon Switzerland Ltd.) as additives were also tested.

Ploss et al. demonstrated that the tribological benefits from ILs were much pronounced when IL concentration was high (10 wt%) [26]. Therefore, the ILs and solid additives were blended into the grease, in 10% wt concentration, using a DAC600

SpeedMixer. The mixing was carried out in three steps where grease was manually mixed using a spatula between steps. This was done to ensure that heavy components, like the copper powder, would blend in properly into the grease matrix.

2.2 Impedance spectroscopy

The conductivity of the greases was evaluated in a two electrode cell assembly using Metrohm Autolab (PGSTAT302N) electrochemical workstation with FRA32M module for impedance spectroscopy. A TSC battery cell (RHD instruments, Germany, Figure 1) was used to obtain the impedance spectra of grease samples. The grease sample was sandwiched between two stainless steel disc electrodes with a contact area of 8 mm in diameter within the battery cell. The size of the sample was controlled by using a PEEK spacer that had an internal diameter of 8 mm and an external diameter of 12 mm. The cell was packed with a screw cap and the contact pressure was adjusted with a gold plated spring (2.3 N.mm⁻¹ spring constant). The screw cap was tightened by two turns, resulting in a force of 4.6 N acting on an area of 1.13 cm². The total contact pressure applied on the sample was about 40 kPa. The cell constant for the battery cell, ratio of the spacer thickness and the exposed area of the electrode, was calculated to be 0.015915494 cm⁻¹. Impedance measurements for grease samples were performed in the frequency range of 0.1 Hz to 1 MHz with an AC amplitude of 10 mV_{rms}.

The cell was mounted on a Microcell HC cell stand (RHD instruments, Germany) coupled with a temperature controller unit. In the Microcell HC setup, the temperature was controlled by a Peltier element and experiments were performed in the temperature range of 0 to 100 °C. Prior to each experiment, the electrodes were polished with a Kemet diamond paste (0.25 µm). Impedance responses of four temperature sweeps, alternating between heating and cooling (hereinafter referred to as 1-Heating, 2-Cooling, 3-Heating and 4-Cooling sweeps), were recorded. The cells were thermally equilibrated for at least 10 min before each measurement. The measured impedance data were recorded using the NOVA 2.1 software.

The obtained impedance spectra were visualized and analyzed using RelaxIS3 software from RHD Instruments. The characteristics of the

Table 2: Molecular structures of the ionic liquids

Cation	Anion	Designation	Source
trihexyl(tetradecyl)phosphonium	bis(oxalato)borate	P-BOB	[45]
bis(mandelato)borate	P-BMB	[46]
	dicyanamide	P-DCA	<i>Cytec</i>
	bis(2-ethylhexyl)phosphate	P-BEHP	<i>Iolitec</i>
1-Ethyl-3-methylimidazolium	bis(trifluoromethylsulfonyl)imide	EMIM-TFSI	<i>Iolitec</i>

Nyquist impedance plots (real part of impedance plotted against the imaginary part at a series of frequencies) were evaluated to establish relevant equivalent circuits which would reflect the bulk as well as the interfacial electrical properties. The equivalent circuits were composed of electrical circuit elements like resistors, constant phase elements (CPEs) and Warburg elements. CPEs are general circuit elements with a constant phase angle (0° for a resistor, -90° for a capacitor, or $+90^\circ$ for an inductor) [49]. Here, CPEs are used to represent imperfect bulk capacitance and double layer capacitance at the electrode interface. The non-ideal behavior can be attributed to various factors such as surface roughness, non-uniform current and potential distribution, electrode porosity and deviations due to oxide layer [50]. The impedance of a CPE can be expressed as: $Z_{CPE} = \frac{1}{(i\omega)^\alpha Q}$; where Q is the admittance value and α is an

exponent generally between 0.9 and 1 ($\alpha = 1$ for ideal capacitor). A Warburg element is commonly used to describe semi-infinite linear diffusion, i.e., unrestricted diffusion to a large planar electrode [51]. The impedance of a Warburg element can be expressed as: $Z_W = \frac{A_W}{\sqrt{\omega}} - i \frac{A_W}{\sqrt{\omega}}$; where A_W is the Warburg-coefficient. The impedance of the resistor is equal to its resistance, and is independent of the AC frequency (ω). The elements could be combined in various configurations, combination of series and parallel arrangements, to correspond to the various features of the spectra. For example, an offset from zero could be represented by a resistor, an inclined straight line could be described by a double-layer capacitance (CPE) or diffusion (Warburg element), while a semi-circular feature could be represented by a RC circuit (resistor and capacitor in parallel), or by a RP circuit (resistor and CPE in parallel) in case of a "depressed" semicircle.

Fig. 1: Schematics of TSC battery cell. The cell is in a two electrode configuration with the sample sandwiched between two parallel stainless steel electrode plates, with a constant separation maintained through a PEEK spacer, packed together with a screw cap and spring mechanism.

The impedance data were fitted to the equivalent electrical circuits to obtain values for the different circuit elements. The bulk conductivity was extracted from the fitted value of the resistor of the part of circuit representing the bulk properties. The conductivity, σ $\mu\text{S cm}^{-1}$, was calculated as: $\sigma = \frac{k}{R} \times 10^6$; where k is the cell constant in cm^{-1} and R is the fitted resistance in Ω .

For evaluation of ionic conductivity of neat ILs, a TSC 70 cell (RHD instruments, Germany) was employed. The cell was set-up in a two-electrode configuration, with a 0.25 mm platinum wire as a working electrode and a 70 μL platinum cup as a sample container and as a counter electrode (Figure 2). Impedance measurements were performed in the frequency range from 1 Hz to 1 MHz with a 10 mV_{rms} AC voltage amplitude. The cell constant of the liquid cell, $K_{Lcell} = 18.5396 \text{ cm}^{-1}$, was determined by using a 100 $\mu\text{S cm}^{-1}$ KCl standard solution from Metrohm.

3 Results and Discussion

Nyquist plots for the grease samples at 60 °C during the 3-Heating temperature sweep are shown

Fig. 2: Schematics of TSC 70 battery cell. The cell uses a two-electrode configuration with a 0.25 mm platinum wire working electrode (a); 70 μL platinum cup (b) acting as a counter electrode while also holding the liquid sample (c).

as examples in Figure 3. In Appendix A, the equivalent electrical circuits selected for fitting the various impedance spectra are listed (Figure A1). The physical significance of all the circuits used in this study are also explained in Table A1.

Almost all of the grease samples exhibited the semi-circular spectral feature (originating from complex impedance of the bulk), leading to a selection of RP circuit component to represent the bulk electrical properties. Only for the grease with EMIM-TFSI, an offset from zero was observed on the real axis, motivating the use of only a resistor element to represent the bulk electrical properties. This shows that for the grease with EMIM-TFSI the bulk capacitance is negligible.

The nature of the equivalent circuit representing the bulk remains stable over time and temperature for all the samples, though of course the values of the circuit components change. The bulk conductivity is extracted from the appropriate value of the bulk resistivity component (an example of which is labeled as R_B in Figure 3). The other circuit components (detailed in Figure A1) are characteristic of interfacial processes, which while important for the grease function are not the focus of this work. Changes were observed,

Fig. 3: Impedance spectra Nyquist plots for the grease samples at 60 °C during 3-Heating temperature sweep, and the respective equivalent electrical circuits.

over time and temperature, in the nature of the circuit representing the sample-electrode interface, reflecting the complexity and dynamics of this region. All the grease samples, other than the base grease itself, show an interfacial spectrum which can be well represented by a double layer capacitance and a charge transfer resistor in parallel. The interfacial spectral features for greases with solid additives become evident only after some time into the 1-Heating temperature sweep. The greases with ILs, other than P-BEHP, exhibit an additional spectral feature which can be represented by a diffusion component (either Warburg element or a CPE) in the equivalent circuit. For the grease with copper and P-BOB at lower temperatures, the charge transfer resistor associated with the double layer capacitance appears to become too high to be captured within the measured frequency range, leading to the spectra being better fitted using only a CPE element. It must be noted that the spectral features corresponding to the interfacial structuring (low frequency) observed within the tested frequency range are applied solely to provide a good fit to this data and allow the higher frequency features (bulk conductivity) to be accurately captured. Therefore, the choice of equivalent circuit for the interfacial region is only indicative and not intended to give detailed information on the structuring at the electrode interface. More in-depth

studies would be needed to reveal the interfacial structuring and its electrical characteristics.

The conductivity values obtained for all the grease samples, during the four temperature sweeps, are shown in Appendix B (Figure B4). It can be observed that, there is a certain dependence on the sweep number, particularly in the case of the particulate additives, which is likely related to a conditioning process. The base grease (without any additives) exhibits only a very low conductivity despite its ionic nature (Figure B4a). The increasing conductivity with increasing temperature can be attributed to the thermally induced increase in mobility of the charge carriers (possibly dissociated lithium ions from the thickener) as well as the decrease in rheological resistance, with increase in temperature, of the grease matrix.

Figure 4 presents a comparison of the temperature dependent conductivity of the LiX greases with solid additives, during the 3-Heating and 4-Cooling temperature sweeps. It can be seen that the addition of solid additives to the grease increases the conductivity by several orders of magnitude. Grease containing a graphite additive shows an unusual inverse conductivity dependency on temperature. Similar observations have been reported for pure graphite [52] as well as for semiconductor graphite/epoxy composites [53]. This indicates that the conductivity mechanism is dominated by the conduction through a network of graphite chains. This network appears to form during the 1-Heating temperature sweep since the conductivity during that sweep is much lower than the following temperature sweeps (Figure B4c). The network formation is presumably facilitated by reduced viscosity at the higher temperature end of the first heating sweep, allowing connections to be made between the dispersed particles. The reduction in conductivity is thus related to the known poorer conductivity of graphite at elevated temperatures [52]. In the case of grease with copper powder, the conductivity is lower than for the graphite case, and the strong relation between conductivity and temperature is inconsistent with the intrinsic electrical conductivity of copper metal. This shows that the grease sample with copper follows an ionic conductivity pathway, or possibly hybrid. Adsorption of anionic grease thickener component to the metal surface may lead to the liberation of more dissociated lithium

Fig. 4: Bulk conductivity of the grease samples with solid additives, i.e., copper and graphite, as a function of temperature during 3-Heating (a), and 4-Cooling (b) temperature sweeps.

ions, resulting in an increased conductivity compared to the base grease. Released copper ions may also contribute. As in the case of graphite additives, the conductivity during the 1-Heating sweep is quite low (Figure B4b), suggesting that the association is thermally assisted. It is also possible that the particles are able to more easily align, with the applied field, or partially aggregate leading to the hybrid conduction mechanism alluded to above – that is to say a polarization in the metallic particles with correspondingly facilitated ion separation.

Figure 5 shows the comparison of temperature dependent conductivity for LiX greases with IL additives, during the 3-Heating and 4-Cooling temperature sweeps. Addition of ILs to the grease also results in an increase in conductivity by several orders of magnitude. The direct relation between temperature and conductivity confirms that the ionic conductivity pathway is followed. The increase in conductivity compared to the neat grease can be seen to be highly dependent on the choice of IL. The EMIM-TFSI additive results in the highest conductivity, up to three orders of magnitude higher than the next most conductive grease sample (LiX + P-DCA) which in turn is over an order of magnitude more conductive than the least conductive grease sample (LiX + P-BMB). The EMIM-TFSI grease conductivity is also greater than that displayed by the grease with graphite additive, which is otherwise the most

conductive sample. Its high conductivity presumably reflects a greater degree of dissociation of the ions (lower ion-pairing). By the same token, the larger proportional increase in conductivity of the other ionic liquids with temperature suggests that increased temperature leads not only to reduced rheological resistance, but also to an increase in ion dissociation.

As a comparison, the conductivity of neat ILs during 3-Heating temperature sweep is shown in Figure 6. The selection of equivalent electrical circuits for the neat IL impedance spectra are shown in Appendix A (Figure A2). The ionic conductivity showed negligible variance between the four temperature sweeps, reflecting the much simpler liquid structure. EMIM-TFSI once again exhibits the highest conductivity of all ILs, with the results congruent with values reported by Widegren et al.[48], measured using a commercial conductivity cell based on AC impedance bridge technique. The high conductivity of EMIM-TFSI can be attributed to the less bulky ions compared to the phosphonium ILs, therefore allowing for better ionic mobility, combined with a higher degree of ion dissociation. The conductivity values for P-BOB and P-BMB are also in good agreement with previously reported data [45, 46].

Here it can be seen that the order of conductivity of neat ILs is not the same as the order of conductivity when the same ILs are added to the grease. As a neat IL, P-BEHP has much

Fig. 5: Bulk conductivity of the grease samples with ILs as a function of temperature during 3-Heating (a), and 4-Cooling (b) temperature sweeps.

Fig. 6: The conductivity values obtained for different neat ILs during the 3-Heating temperature sweep. Negligible variation was observed between the different temperature sweeps.

lower conductivity than that of P-BOB (Figure 6), whereas the conductivity of greases with P-BEHP higher than that for grease with P-BOB (Figure 5). This is likely a result of P-BEHP being the only IL miscible in the base oil, allowing conductivity through the continuous oil phase rather than solely through polar regions associated with the LiX thickeners. Supporting this hypothesis is the fact that hysteresis in the temperature

dependence of conductivity can be observed for all grease samples other than the grease with P-BEHP. The conduction mechanisms thus do not depend on the liquid properties of the ILs, but rather upon how and where they are distributed in the grease matrix. Therefore, the conductivity trend for greases with IL additives can not be directly equated to the conductivity of neat ILs. Along with the degree of ion dissociation, the self assembly properties of the ions with the grease matrix or the base oil must also be considered while evaluating the conductivity mechanism of greases doped with ILs.

The impedance measurements described and their ensuing analysis demonstrate that the functionality of EIS can be extended to conductive greases based on both ionic and non-ionic additives. In comparison to the commonly used resistive or capacitive sensing techniques, through evaluation of both the real and imaginary part of the impedance, EIS allows for much more detail to be extracted about the electrical properties of the bulk grease sample and the interfacial structuring on the electrode surface. This method is particularly useful for complex systems where the impedance of the bulk is frequency dependent. It allows for the accurate measurement of temperature and time dependent conductivity by isolating the contributions of the interfacial features.

4 Conclusion

A methodology to measure the bulk conductivity of greases doped with ionic (ionic liquids) and non-ionic (copper and graphite) additives, using electrochemical impedance spectroscopy (EIS) has been proposed in this paper. In contrast to traditional measurement techniques, study of the multi-frequency impedance spectra (real and imaginary) and analysis of the suitable equivalent electrical circuits is an effective means for extracting the bulk electrical properties, by isolating them from the electrical contributions from the electrode-sample interfacial features. Future work will also be directed at the interfacial responses of these systems which are of course of great interest for understanding the boundary layer response. LiX greases are complex colloidal systems, but the conductivity mechanisms can nonetheless be understood in terms of the additive properties. The approach can also permit understanding of how mechanisms vary with temperature and also how heating conditioning of the grease can be used to tune the electrical properties. In the case of graphite additives, temperature cycling allowed the conductivity to become reduced with increasing temperature, due to the network formation across the grease facilitated by reduced viscous forces. This opens avenues for controlling the temperature dependence of the grease conductivity. Addition of IL to grease in 10% wt. concentration resulted in increases in conductivity by three to six orders of magnitude, indicating both the large differences in conductivities of the pure ILs themselves, but also large differences of the ability to interact with the grease materials themselves – for example as an ionic solute in the oil or in association with the polar regions of the matrix. Thus, not only the ion dissociation is important, but also the self-assembly properties of ions with the grease matrix. The results allow for the systematic control and study of the electrical properties of greases over many orders of magnitude, and the control of both temperature and temporal response.

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Appendix A Equivalent electrical circuits

Figure A1, A2, A3 and Table A1.

Appendix B Conductivity of grease samples

Figure B4.

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Sample	1-Heating		2-Cooling		3-Heating		4-Cooling	
	0 °C →	100 °C	100 °C →	0 °C	0 °C →	100 °C	100 °C →	0 °C
LiX-10								
LiX-10 + 10% Cu		60 °C		40 °C		30 °C		40 °C
LiX-10 + 10% Graphite		90 °C						
LiX-10 + 10% P-BOB		90 °C		30 °C		20 °C		30 °C
LiX-10 + 10% P-BMB		20 °C		20 °C		10 °C		20 °C
LiX-10 + 10% P-DCA		40 °C						
LiX-10 + 10% P-BEHP								
LiX-10 + 10% EMIM-TFSI								

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Fig. A1: Equivalent electrical circuit selection for different impedance spectra of grease compositions.

Sample	1-Heating		2-Cooling		3-Heating		4-Cooling	
	0 °C →	100 °C	100 °C →	0 °C	0 °C →	100 °C	100 °C →	0 °C
P-BOB	* 30 °C		50 °C		30 °C		40 °C	*
P-BMB								
P-DCA		70 °C		80 °C				
P-BEHP		60 °C		60 °C		60 °C		60 °C
EMIM-TFSI	§ 30 °C							

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Fig. A2: Equivalent electrical circuit selection for different impedance spectra of neat ILs

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Fig. B4: Conductivity as a function of temperature for all the grease samples during the four temperature cycles.

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Declarations

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Table A1: Explanation of physical relevance of the equivalent electrical circuits

Equivalent circuit	Explanation
<p>This circuit assumes the absence of interfacial layers. Resistor 1 represents bulk electrolyte resistance while CPE 1 represents bulk capacitance.</p>
<p>Resistor 1 represents the bulk resistance while CPE 1 represents the bulk capacitance. CPE 2 corresponds to the double layer capacitance at the interface. It might be possible that the double layer capacitance is supported by a parallel charge transfer resistance which is too large to be captured in the spectra.</p>
<p>Simplified Randles circuit: Resistor 1 represents ionic resistance (electrolyte conductivity). The bulk has very low capacitance which is not captured by the spectra. CPE 1 represents double layer capacitor (charging of double layer) while Resistor 2 in parallel represents charge transfer resistance.</p>
<p>Resistor 1 and CPE 1 represent the bulk resistance and capacitance respectively. CPE 2 represents the double layer capacitance, supported by a parallel charge transfer resistance, Resistor 2.</p>
<p>Randles circuit: Resistor 1 represents ionic resistance (electrolyte conductivity). CPE 1 represents double layer capacitor while Warburg element 1 and Resistor 2 in series represent the rate of the faradaic reaction controlled by the diffusion of the reactants to the electrode surface.</p>
<p>Modified Randles circuit: This circuit is similar to the Randles circuit, but the bulk is represented by a resistive element (Resistor 1) and a capacitive element (CPE 1).</p>
<p>This circuit is similar to the Randles circuit, but with non-ideal diffusion. Therefore, the Warburg element in Randles circuit is replaced by CPE 1.</p>
<p>This circuit is similar to the modified Randles circuit, but with non-ideal diffusion. Therefore, the Warburg element in the modified Randles circuit is replaced by CPE 2.</p>