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Synthesis and ionic conductivity of siloxane based polymer electrolytes with pendant propyl acetoacetate groups

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Abstract: Hydrosilylation reaction of 2.4.6.8-tetrahydro-2.4.6.8-tetramethylcyclotetrasiloxane (D_4^H) with allyl acetoacetate at 1:4.1 molar ratios of initial compounds in the presence of platinum hydrochloric acid (0.1 M solution in THF), Karstedt's catalyst ($Pt_2[(VinSiMe_2)_2O]_3$) and platinum on the carbon have been carried out and D_4^R type compounds obtained. Reaction order, rate constants and activation energies for hydrosilylation reactions in the presence of Karstedt's catalyst have been determined. Ring-opening polymerization reaction of D_4^R in the presence of catalytic amounts of powder-like potassium hydroxide has been carried out. A linear methylsiloxane oligomer with regular arrangement of propyl acetoacetate groups in the side chain has been obtained. The synthesized methylorganocyclotetrasiloxane and oligomers were studied by FTIR, ^1H , ^{13}C and ^{29}Si NMR spectroscopy. Comb-type oligomers were characterized by gel-permeation chromatography, wide-angle X-ray diffractometry and differential scanning calorimetry. Solid oligomer electrolyte membranes have been obtained via sol–gel processes involving lithium trifluoromethylsulfonate (triflate) or lithium bis(trifluoromethylsulfonyl)imide. The specific volumetric electrical conductivity of the membranes at room temperature covers a wide range, from 10^{-10} to $10^{-4} S \cdot cm^{-1}$, and depends on the structures of grafted anion receptors and the polymer backbones. Higher values of the specific conductivity are seen for the membranes containing triflate.

Keywords: conducting polymers; membranes; POLYCHAR-25; polymer electrolyte membranes; polysiloxanes.

Introduction

Solvent-free polymer electrolytes may be formed by the interaction of polar polymers with metal ions. Ion transport in polymer electrolytes has been extensively studied since the first report of high conductivity of a polyethylene oxide/KSCN complex by Wright and Co-workers [1] in 1973 and the unique idea of employ-

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ing these polymer electrolytes in battery applications by Armand et al. [2] afterwards. Transport mechanism models developed by Ratner and Shriver [3] indicate that polymers with low glass transition temperatures T_g have at higher temperatures high free volumes – which favor ion transport. Better results are obtained for polymers with highly flexible backbones, bearing oligo(ethylene glycol) (EO) side chains.

Interest in polysiloxane-based polymer electrolytes arose early in the 1980s. Poly(ethylene oxide) (PEO) – substituted polysiloxanes as ionically conductive polymer hosts have been investigated [4–11]. Their relatively high ionic conductivity was ascribed to the highly flexible inorganic backbone, which produced a fully amorphous polymer host. In recent years, improved battery performance has been observed for systems containing polymer electrolytes, with a Li^+ transference number close to unity [12]. Efforts have also been made to design and synthesize siloxane-based single-ion conductors [13, 14].

Polysiloxanes are promising components for comb polyelectrolytes precisely because they possess a flexible backbone that enhances the transports of ions. Their amorphous and highly flexible $[\text{Si}-\text{O}]_n$ backbone results in glass transition temperatures as low as $-100\text{ }^\circ\text{C}$; there is little or no crystallinity at room temperature. In addition, each monomer unit has two sites for cross-links or functional side chains through bond formation with silicon. Simulations indicate that comb polyelectrolytes should display higher conductivity values than their analogs to local motion of the bound anions in comb systems [15].

In order to improve ionic conductivity, various approaches may be employed. The main requirements for high conductivity are ability to solvate ions and low glass transition temperatures affording facile ion transport. The promotion of the anion-cation dissociation is desirable, as it should lead to enhancement of ionic conductivity via an increase in free ion concentration. In this situation, we have decided to develop solid polyelectrolyte membranes on the basis of new siloxane matrices.

Experimental

Materials

2,4,6,8-tetrahydro-2,4,6,8-tetramethylcyclotetrasiloxane = D_4^{H} , platinum hydrochloric acid, $(\text{Pt}_2[(\text{VinSiMe}_2)_2\text{O}]_3)$ Karstedt's catalyst or platinum (0)-1,3-divinyl-1,1,3,3-tetramethyldisiloxane complex (2 % solution in xylene), platinum hydrochloric acid, Pt/C (10 and allyl acetoacetate were purchased from Aldrich and used as received. Lithium trifluoromethylsulfonate (triflate = salt S_1) and lithium bis(trifluoromethylsulfonyl)imide (salt = S_2) were also purchased from Aldrich. Toluene was dried over and distilled from sodium under an atmosphere of dry nitrogen.

Tetrahydrofuran (THF) was dried over and distilled from K–Na alloy under an atmosphere of dry nitrogen. 0.1 M solution of platinum hydrochloric acid in THF was prepared and kept under nitrogen at a low temperature.

Characterization

FTIR spectra were recorded on a Nicolet Nexus 470 machine with MCTB detector. ^1H , ^{13}C NMR and ^{29}Si NMR spectra were recorded on a Varian Mercury 300VX NMR spectrometer, using DMSO (dimethyl sulfoxide, $(\text{CH}_3)_2\text{SO}$), and CCl_4 as the solvent and an internal standard, respectively. Differential scanning calorimetric investigation (DSC) was performed on a Netzsch DSC 200 F3 Maia apparatus. Thermal transitions including glass transition temperatures T_g were taken as the maxima of the DSC peaks. The heating and cooling scanning rates were 10 K/min.

Gel-permeation chromatography (GPC) was carried out with a Waters Model 6000A chromatograph with an R 401 differential refractometer detector. The column set comprised 10^3 and 10^4 Å ultrastaygel columns. Sample concentration was approximately 3 % by weight in toluene; typical injection volume for the siloxane

was 5 μl , flow rate about 1.0 ml/min. Standardization of the GPC was accomplished by the use of styrene or polydimethylsiloxane standards with the known molecular weight. Determination of $\equiv\text{Si-H}$ content was calculated according to the method described in [16].

Hydrosilylation reaction of D_4^{H} with allyl acetoacetate

D_4^{H} (2.00 g, 0.00832 mol) were transferred into a 100 ml flask under nitrogen using standard Schlenk techniques. High vacuum was applied to the flask for half an hour before the addition of allyl acetoacetate (4.85 g, 0.0341 mol). The mixture was then dissolved in 7 ml of toluene; 0.1 M solution of platinum hydrochloric acid in tetrahydrofuran (from $5 \cdot 10^{-5}$ to $9 \cdot 10^{-5}$ per 1.0 g of starting substance) was introduced. The homogeneous mixture was degassed and placed into an oil bath, which was previously set to 50 °C and the reaction continued at 50 °C. The reaction was controlled by decrease of intensity of active $\equiv\text{Si-H}$ groups. Then 0.1 % activated carbon was added and refluxed for 12 h for deactivation of the catalysts.

All volatile products were removed by rotary evaporation and the compound was precipitated at least three times into pentane to remove side products. Finally, all volatiles were removed under vacuum and further evacuated under high vacuum for 24 h to isolate 6.49 g (96.5 %) of colorless viscous compound I-2.4.6.8-tetramethyl-2.4.6.8-tetra(propyl acetoacetate)cyclotetrasiloxane (D_4^{R}).

The hydrosilylation reactions in the presence of other catalysts were carried out according to the same method.

Ring-opening polymerization reaction of D_4^{R}

The 1.137 g (1.40 mmol) of compound D_4^{R} was transferred into a 50 ml flask under nitrogen. High vacuum was applied to the flask for half an hour. Then the compound was dissolved in 1.8 ml dry toluene and 0.01 % of total mass powder-like potassium hydroxide was added. The mixture was degassed, placed in an oil bath that was previously set to 60 °C, and polymerized under nitrogen for 25 h. Then 7 ml of toluene were added to the reaction mixture and the product washed with water. The crude product was stirred with MgSO_4 for 6 h, filtered and evaporated; the oligomer was precipitated at least three times into pentane to remove side products. Finally, all volatiles were removed under vacuum to isolate 1.06 g (93 %) colorless viscous oligomer (II).

Ring-opening polymerization reaction of compound I at various temperatures has been carried out in the same manner.

General procedure for preparation of cross-linked polymer electrolytes

In a typical preparation, 0.75 g of oligomer II was dissolved in 4 ml of dry THF and thoroughly mixed for half an hour before the addition of a catalytic amount of acid (one drop of 0.1 N HCl solution in ethyl alcohol) to initiate the cross-linking process. After stirring for another 3 h, a required amount of lithium triflate (salt to be called S_1) from the previously prepared stock solution in THF was added to the mixture and stirring continued for 1 h more. The mixture was then poured onto a Teflon mold with a diameter of 4 cm and solvent was allowed to evaporate slowly overnight. Finally, the membrane was dried in an oven at 70 °C for 3 days and at 100 °C for 1 h. Homogeneous and transparent films with average thickness of 200 μm were thus obtained. These films were insoluble in all solvents, only swollen in THF.

AC impedance measurements

The total ionic conductivity of samples was determined by placing an electrolyte disk between two 10 mm diameter brass electrodes. The electrode/electrolyte assembly was secured in a suitable constant volume

support which allowed highly reproducible measurements of conductivity to be performed between repeated heating-cooling cycles. The cell support was located in an oven and the sample temperature was measured by a thermocouple positioned close to the electrolyte disk. The bulk conductivities of electrolytes were obtained during a heating cycle using the impedance technique (an impedance meter BM 507 – TESLA for frequencies 50 Hz–500 kHz) over a temperature range between 20 and 100 °C.

Results and discussion

Hydrosilylation reaction of D_4^H with allyl acetoacetate

Preparation of new siloxane matrices with pendant propyl acetoacetate groups was carried out in two stages. In the first stage, hydrosilylation reaction of D_4^H with allyl acetoacetate at 1:4.1 molar ratios of initial compounds in melt condition and in dilute solution of dry toluene or THF (50 %) at 50 °C in the presence of platinum catalysts was carried out.

Separately, preliminary heating of initial compounds in the temperature range of 50–60 °C in the presence of catalysts showed that in these conditions polymerization of D_4^H , or allyl acetoacetate and scission of siloxane backbone does not take place. No changes in the NMR and FTIR spectra of initial compounds were found.

As it was established earlier by some of us [17] in the melt condition, the hydrosilylation reaction proceeds vigorously with initiation of side reactions; therefore, for obtaining the addition product, kinetic parameters the hydrosilylation reactions have been studied in dry solution of toluene. The initial compounds are insoluble systems; in spite of the fact that the reaction is carried out in toluene solution, the homogeneity of the solution is not achieved. The same problem has been observed when using tetrahydrofuran. Therefore, the investigation of hydrosilylation reaction have been extended in dry toluene solution at $C = 0.127$ mol/l concentrations. Accordingly, hydrosilylation reaction of D_4^H with allyl acetoacetate proceeds according to the following Scheme 1. Here Karstedt's catalyst at 50 °C = I, at 40 °C = I¹, at 30 °C = I², platinum hydrochloric acid at 30 °C = I³, Pt/C (5 %) at 60 °C = I⁴.

The structure and composition of so obtained compound I = D_4^H were determined, namely the molecular mass, FTIR, ¹H, ¹³C and ²⁹Si NMR spectra.

In the FTIR spectra of compound I the absorption band characteristic for $\equiv\text{Si-H}$ at 2160–2170 cm^{-1} region disappeared; instead, one observes absorption bands at 1079 and 1149 cm^{-1} , characteristic for asymmetric valence oscillation of $\equiv\text{Si-O-Si}\equiv$ and $\equiv\text{CO-O-C}\equiv$ bonds. Also one observes absorption bands at 1265 and in the 2800–3000 cm^{-1} region, characteristic for $\equiv\text{Si-C}\equiv$ bonds and for deformation oscillation of $\equiv\text{C-H}$ bonds. The absorption band at 1720 cm^{-1} corresponds to carbonyl groups in the ester fragment. Some characteristics of alkonotetrasiloxanes is provided in Table 1.

Scheme 1: Hydrosilylation reaction of D_4^H with allyl acetoacetate.

Table 1: Some physical chemical properties of organocyclotetrasiloxanes.

#	Reaction Temp., °C	Yield, %	n_D^{20}	d_4^{20}	M_{RD}^a		M^b	
					Calcul.	Found	Calcul.	Found
I	50	93	1.462	1.1789	186.5	188.7	809	800
I ¹	40	91	1.462	1.1789	186.5	—	809	—
I ²	30	90	1.462	1.1789	186.5	—	809	—
I ³	30	90	1.4623	1.1789	186.5	187.3	809	795
I ⁴	60	87	1.462	1.1789	—	—	809	—

^a M_{RD} is molecular refraction, ^b M are molecular masses determined by ebulliometry.

²⁹Si NMR spectra of compound I show resonance signal with the center of chemical shift at $\delta = -20$ ppm that can be assigned to the RR'SiO (D) units in the cyclic tetramer [18, 19], thus a structure containing siloxane D units only.

We now provide ²⁹Si NMR spectra of compound I in Fig. 1 and ¹H NMR spectra of that compound in Fig. 2.

One can observe above signals with the center of the chemical shift $\delta = 0.14, 0.95$ and 2.95 ppm, characteristic for methyl protons in fragments $\equiv Si-Me$, $=CH-CH_3$ and $-OCH_2CH_3$, respectively. Multiplet signals with the center of the chemical shift $\delta = 0.6, 1.60, 3.50$ and 4.1 ppm are characteristic for methylene protons in $\equiv SiCH_2$, $-CH_2CH_2CH_2-$, $-COCH_2CO-$ and $-OCH_2-$ fragments, respectively. The multiplet signal with the center of the chemical shift $\delta = 2.50$ ppm corresponds to the methine group $=CH-$ (α addition).

We show ¹³C NMR spectra of compound I in Fig. 3. One can observe signals for the carbon cores with chemical shifts: $-2.1, 13.2, 21.8, 29.8, 67.6, 167$ and $166.2-200$ ppm characteristic for carbon in: $\equiv Si-Me$, $\equiv Si-CH_2$, $-CH_2-CH_2-CH_2-$, $CO-CH_2$, $CO-CH_2-CO$, CH_2-O and $C=O$ group, respectively.

We have obtained also ¹³C NMR spectra and we have found that they agree well with ¹H NMR spectra. We show in Fig. 3 the changes of concentration of active $\equiv Si-H$ groups with time during hydrosilylation reaction of D_4^H with allylacetate in the presence of Karstedt's catalyst. Evidently the hydrosilylation reaction proceeds vigorously for the first 10 min, then it slows down.

In Fig. 4 we present the changes of concentration of active $\equiv Si-H$ groups during hydrosilylation reaction of D_4^H with allyl acetate in the presence platinum hydrochloric acid; it proceeds in the same way as in the presence of Karstedt's catalyst. As regards platinum on the carbon, that is Pt/C, the hydrosilylation reaction proceeds less vigorously at $60^\circ C$ than at low temperatures.

Fig. 1: ²⁹Si NMR spectra of compound I.

Fig. 2: ^1H NMR spectra of compound I.

Fig. 3: ^{13}C NMR spectra of compound I.

The reaction rate constants of hydride addition reactions of D_4^{H} with allyl acetoacetate at various temperatures were determined: $k_{30^\circ\text{C}} = 0.933$, $k_{40^\circ\text{C}} = 1.235$ and $k_{50^\circ\text{C}} = 1.664 \text{ mol} \cdot \text{l}^{-1} \cdot \text{s}^{-1}$, and the activation energy of hydrosilylation reaction has been calculated as $E = 31.1 \text{ kJ/mole}$.

Ring-opening polymerization reaction of D_4^{R}

We have obtained linear polysiloxanes with pendant acetoacetate groups by ring-opening polymerization reactions of compound I in the presence of (0.01–0.005 % of total mass) alkali fluorides (KF, LiF and CaF_2) as catalysts and tetra(methyl ammonium) hydroxide. The reactions were carried out in the inert atmosphere, in melt condition and in toluene solution between 50 and 110°C . Gas-liquid chromatography (GLC) shows that alkali fluorides at these conditions do not promote polymerization; only 25–30 % of initial compounds participate in the ring opening polymerization in the case of KF, a very low yield. Catalytic activity of the fluorides decreases in the order: $\text{CaF}_2 < \text{LiF} < \text{KF}$.

Polymerization reaction of compound I also have been studied in inert atmosphere in toluene solution ($C = 0.669 \text{ mol/l}$) in the temperature range $60\text{--}70^\circ\text{C}$ in the presence of powder-like anhydrous potassium hydroxide (0.05–0.01 % of total mass). In these conditions the ring-opening polymerization takes 28–35 h.

Fig. 4: Changes of concentration of active $\equiv\text{Si-H}$ groups with time during hydrosilylation reactions of D_4^{H} with allyl acetoacetate in the presence of Pt/C (5 %) catalyst at 60 °C (1), platinum hydrochloric acid at 30 °C (2), Karstedt's catalyst at 30 °C (3), 40 °C (4) and 50 °C (5).

Low molecular mass oligomers are obtained at higher temperatures. Better results are obtained in the temperature range 50–60 °C.

Preliminary heating of initial organocyclotetrasiloxane in the temperature range 60–80 °C without catalyst showed that under these conditions neither polymerization, nor elimination of methane, nor scission of siloxane backbone, nor any other side processes take place. No changes in the FTIR spectra of the initial compound were found. GLC has confirmed that those processes do not proceed under these conditions.

Polymerization reactions of compound I in solution, in the presence of powder-like potassium hydroxide, at various temperatures have been studied and decrease of concentration of compound I in the time was determined via GLC.

Polymerization reaction of D_4^{R} proceeds according to the following Scheme 2. Here: 40 °C = II², 50 °C = I¹ and 60 °C = II.

The synthesized oligomers are vitreous, viscous products, well soluble in ordinary organic solvents with specific viscosity $\eta_{\text{sp}} \approx 0.14\text{--}0.20$. The structure and composition of oligomers were determined by elemental analysis, molecular masses by GPC, also FTIR and NMR spectra were obtained. The yields and some physical-chemical properties of oligomers are presented in Table 2.

We find that the polymerization of compound I proceeds more vigorously compared with 2,4,6,8-tetramethyl-2,4,6,8-tetrapropylbutyratecyclotetrasiloxane; the latter takes more than 48 h [20]. The reaction rate constants

Scheme 2: Polymerization reaction of compound I.

Table 2: Some physical-chemical parameters of polymethylpropylacetoacetatesiloxane.

No	Yield, %	Polymerization temperature T, °C	The time of polymerization, h	η_{sp}	T _g , °C	$\bar{M}_n^a$ $\bar{M}_w$, D
II	93	60	25	0.14	−95	$7.23 \cdot 10^3$ $2.86 \cdot 10^4$ 3.96
II ¹	94	50	27	0.20	−98	—
II ²	92	40	30	0.18	—	—

^aMolecular masses were determined by GPC; polydispersity index $D = \bar{M}_w / \bar{M}_n$.

Fig. 5: ¹H NMR spectra of oligomer II.

of polymerization reaction at various temperatures were determined: $k_{40^\circ\text{C}} = 1.147 \cdot 10^{-2}$, $k_{50^\circ\text{C}} = 0.894 \cdot 10^{-2}$ and $k_{60^\circ\text{C}} = 0.723 \cdot 10^{-2} \text{ l mol}^{-1} \cdot \text{s}^{-1}$; the activation energy is $E_{\text{act}} = 155 \text{ kJ/mol}$.

The compositions and structures of new oligomers were determined by elementary analyses, determination of molecular masses, FTIR and NMR spectra.

For oligomer II, ²⁹Si NMR spectra show only one signal with chemical shift $\delta = -20.0 \text{ ppm}$, this indicates that the oligomer chains contain D units only.

We show in Fig. 5 the ¹H NMR spectrum of oligomer II. One observes in that figure signals with centers of chemical shifts $\delta = 0.14$, 0.95 and 2.95 ppm , characteristic for methyl protons in the fragments $\equiv\text{Si}-\text{Me}$, $=\text{CH}-\text{CH}_3$ and $-\text{COCH}_3$, respectively. Multiplet signals with centers of chemical shifts $\delta = 0.6$, 1.60 , 3.50 and 4.1 ppm are characteristic for methylene protons in $\equiv\text{SiCH}_2$, $-\text{CH}_2\text{CH}_2\text{CH}_2-$, $-\text{COCH}_2\text{CO}-$ and $-\text{OCH}_2-$ fragments, respectively. Multiplet signal with the center of chemical shift $\delta = 2.50 \text{ ppm}$ corresponds to the methine group (α addition). ¹³C NMR spectra of oligomers are in accordance with ¹H NMR spectra data.

Wide angle X-ray scattering (WAXS) was determined. Our oligomers are single phase amorphous systems. The diffractograms show two diffractions maxima at $2\theta \approx 11.75\text{--}12.00^\circ$ and $2\theta \approx 20.50 \div 20.75^\circ$. The first maximum corresponds to the interchain distances d_1 in the range $d_1 = 7.37\text{--}7.53 \text{ \AA}$ and the second one to $d_2 = 4.28\text{--}4.33 \text{ \AA}$, corresponding to both intra- and interchain interactions.

Preparation of solid polymer electrolyte membranes

Polymeric membranes preparation started with dissolving of oligomer in dry THF in glassware, adding 0.1 M solution of lithium salt $[\text{CF}_3\text{SO}_3\text{Li}=\text{S}_1 \text{ or } \text{CF}_3(\text{SO}_2)\text{N}^-(\text{Li}^+)(\text{SO}_2)\text{CF}_3]=\text{S}_2$ in THF and 3–5 drop tetraethoxysilane

Scheme 3: Cross-linking reaction.

for sol–gel reaction and then applying a magnetic stirrer for 30 min. At this point one drop of 0.1 M solution of HCl in methyl alcohol was added for the initiation of sol–gel processes.

Polymer electrolyte (PE) membranes formation from oligomers obtaining via polymerization reaction of D_4^{R} proceeds according to the following Scheme 3:

Dependence of the electrolytes conductivity on concentration of the salts is presented in Fig. 6.

We see in Fig. 6 maxima on both curves. An explanation of Zhang and Fang [21] might be applicable: formation of ion pairs is taking place. The ion pairs move around together, as particles with a double mass of a single ion, with corresponding reduction of the charge mobility and the conductivity. Wang and Schlenoff [22] as well as Lappan and Scheler [23] discuss also a possibility of pair formation by dissimilar ions such that the total charge is equal to zero; consequently such tandems do not contribute to the electric conductivity at all. We note a small difference in the locations of the maxima. The maximum on the curve for PS_2 (relatively higher molecular mass) appears at a lower salt concentration than for PS_1 electrolyte.

We have also created traditional Svante Arrhenius plots of logarithmic conductivity vs. reciprocal temperature, obtaining approximately straight lines. We do not include these plots for brevity. As expected given the differences in masses, S_1 containing systems have higher conductivity. Values of the conductivities of the PEs at initial and final experimental temperatures are listed in Table 3.

The voltammograms of PE membranes are presented in Fig. 7 and show the effects of varying salt concentrations.

Fig. 6: Dependence of the electrical conductivity of polyelectrolytes PS_1 (1) and PS_2 (2) on the salt concentrations.

Table 3: The conductivities of PEs based on polymer P and salts S_1 and S_2 at temperatures 30 and 90 °C.

Salt	S_1					S_2				
Concentration, wt. %	5	10	15	20	25	5	10	15	20	25
S (30 °C), S/cm	$3.4 \cdot 10^{-9}$	$2.2 \cdot 10^{-6}$	$5.0 \cdot 10^{-5}$	$7.0 \cdot 10^{-5}$	$9.0 \cdot 10^{-5}$	$1.0 \cdot 10^{-10}$	$3.2 \cdot 10^{-7}$	$2.0 \cdot 10^{-5}$	$5.0 \cdot 10^{-5}$	$7.2 \cdot 10^{-6}$
S (90 °C), S/cm	$1.2 \cdot 10^{-7}$	$5.2 \cdot 10^{-5}$	$2.0 \cdot 10^{-4}$	$5.0 \cdot 10^{-4}$	$6.4 \cdot 10^{-4}$	$1.1 \cdot 10^{-9}$	$2.3 \cdot 10^{-5}$	$6.2 \cdot 10^{-5}$	$1.3 \cdot 10^{-4}$	$2.6 \cdot 10^{-5}$

Fig. 7: Voltammograms of PEs based on polymer P containing salt S_1 with concentrations of ions (in wt.%) as follows: 5 (G), 10 (F), 20 (D) and S_2 with concentrations (in wt.%) 5 (C), 10 (E), 20 (B).

The results presented in Fig. 7 are affected by increasing charge – phonon scattering because of increasing acceleration of charged particles in the constant electrical field. The current saturation levels are higher for compounds with the salt S_1 than for the compounds with salt S_2 because of higher mobility of the salt S_1 ions than that of salt S_2 .

Concluding remarks

As noted in the Introduction, polymer electrolytes have the potential to improve battery performance. Sustainability is and will be an increasingly important consideration [24]. Improvement of battery performance is significant also from this broader point of view.

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