

# Seaweed extractions as promising polymer electrolytes for lithium batteries

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**Abstract.** Polymer electrodes are vital components of lithium-ion batteries and have gained great interest in recent years. Currently, the majority of electrodes are petroleum-based that are toxic and non-biodegradable, which consists of serious environmental concerns. Therefore, exploration of natural polymer electrolytes, such as seaweed extracts, balances non-toxicity and ion conductivity as new research directions. In this review, we mainly focus on the progress of polymer electrolytes based on alginate, kappa carrageenan, and other natural polymers of seaweed extracts, including agar and iota carrageenan. We first give a brief overview of the fundamentals of polymer electrolytes. Then, we discuss the structures and performances of seaweed extracts derived polymer electrolytes. Finally, we compare the ion conductivity of natural polymer derived electrolytes and synthetic polymers, and insights on the future direction for improvement of this field.

## 1 Introduction

In the past several decades, the demand for batteries has increased significantly, owing to a sharp growth in the thirst for electronic devices [1]. A battery is a closed system consisting of an anode, a cathode, an electrolyte, and a separator (Fig. 1.) [2]. The anode and the cathode are charge-transfer media redox reactions occur to supply electrical energy. An electrolyte is the key determinant of battery conductivity and efficiency; it serves to conduct ions between the cathode and the anode and physically separate the positive and negative electrodes incorporated into most cell designs.

Recently, the significance of introducing new electrolytes has grown remarkably. Most batteries contain non-biodegradable polymers [3]. A notable drawback of those organic solutions is that most of them are flammable and poisonous [4]. Moreover, only a small proportion of waste can be recycled and reused, resulting in further environmental issues [5]. Therefore, the discovery of biodegradable and non-toxic materials as polymer electrolytes will be significant for an electric device to be wearable, portable, and safe [6]. Recently, solid polymer electrolytes based on natural polymers have been regarded as promising materials to act as a conductor and the substitution of liquid electrolytes because of their unique advantages, such as non-leakage, biodegradable, volumetric stability, solvent-free condition, easy handling, and wide electrochemical stability windows over liquid electrolytes [7]. Among various solid polymer electrolytes, electrolytes based on polysaccharide-based polymer obtained from seaweed have attracted great interest due to

their porous structure [8]. Therefore, it is vital to look into the resultant ion conductivity of one energy-sufficient, portable material consisting of non-biodegradable, costless, and non-toxic electrolytes with different salts.

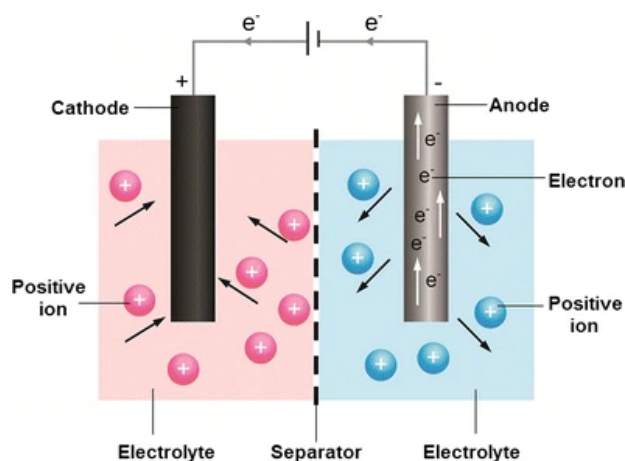


Fig. 1. The structure of a lithium-ion battery [2]

## 2 Results and Discussion

### 2.1. Fundamentals of polymer electrolytes and ion conductivity

#### 2.1.1 Mechanism of polymer electrolytes

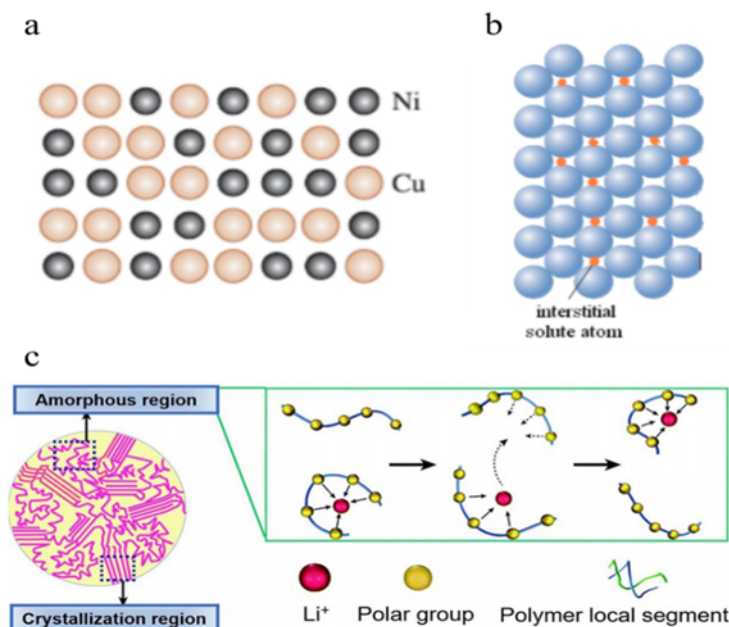
Even with the use of highly amorphous polymers (high content of amorphous phase can facilitate the migration of ions [9]), the minimum ionic conductivity requirement is

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needed for practical applications (which is normally  $\sim 1.00 \text{ mS cm}^{-1}$  [10]) cannot be met. Consequently, the solution of introducing an ionic salt into polymer hosts to further enhance the ion conductivity was proposed by Meyer et al. [11]. They suggested that polymer electrolytes are solid solutions with alkali metal salts being doped in polymers. There are two major types of solid solutions: substitutional and interstitial. The former is made by substituting the solvent particles with solute atoms, while the latter is made by inserting the solute atoms into the gaps between solvent particles [12]. These are two main means of doping alkali metal salts in polymers, and Fig. 2a and Fig. 2b illustrate

these two ways of doping [12].

In a lithium-ion battery, lithium ions can coordinate with the lone pair of oxygen of the polymer electrolyte, such as polyethylene oxide (PEO), via Coulombic interactions, resulting in the dissociation of cations and anions of lithium salt [13]. Under the electrical field, lithium ions can move from one coordination point to another along the polymer [13], and Fig. 2c shows how a specific polymer called poly (ethylene oxide) can act as a host for Li salts [14].



**Fig. 2.** (a) The solute particles substitute the solvent particles (way 1 of doping salts into solid solution) [12] (b) The solute particles insert into the solution (way 2 of doping salts into solid solution) [12] (c) The diffusion of  $\text{Li}^+$  in poly (ethylene oxide)-based SPEs [14].

### 2.1.2 Conditions needed for a high ion conductivity

Under certain conditions, the ion conductivity can be calculated using the following equation:

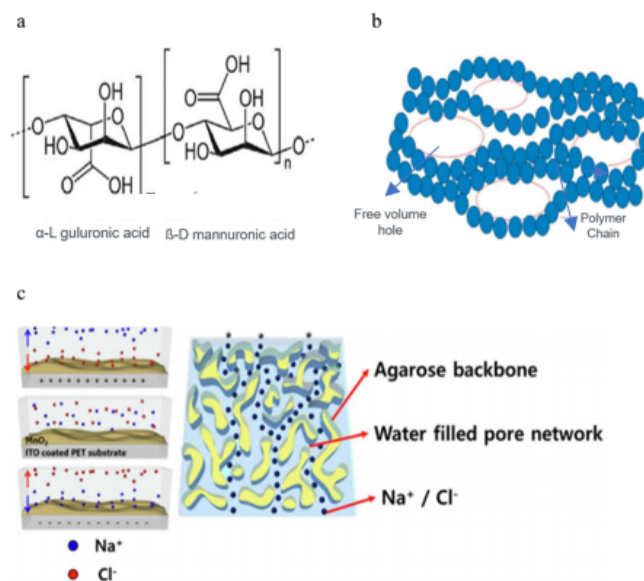
$$\sigma = F \sum n_i q_i \mu_i \quad (1)$$

Where  $F$  is the Faraday constant,  $n_i$  represents the number of free ions,  $q_i$  represents the number of charges, and  $\mu_i$  represents the mobility [13].

The ion conductivity of an electrolyte highly depends on the dissociation of ions in it [15]. Therefore, to increase the ion conductivity of the electrolyte, a variety of conditions should be applied on both electrolytes and salts doped with the electrolytes. For salts, they should have low lattice energy to facilitate their dissociation [11]. For the host polymer electrolytes, requirements include high dielectric constants [14], the high molecular weight of the polymer matrix [14], high degree of amorphous phase [16], low glass transition temperatures [11], and high backbone flexibility [13].

### 2.2 Alginate-based electrolytes

Alginate-based electrolytes have gained interest since 2004 because of their biocompatibility and non-toxicity [18]. Alginate is a polysaccharide found in brown algae (Phaeophyceae), with 1-4 linked mannuronic acid and 1-guluronate being linked linearly [19]. Alginate has attracted many researchers from diverse areas for applications such as tissue engineering, biomedicine, and delivery vehicles for drugs and food packaging [20]. Some important merits allow the industrial applications of alginates, such as their ability to form gels in the presence of bipolar ions (such as calcium ions) [21]. In alginates, their polymer chains generate accessible spaces and form free volumes (volumes of the total mass that are not occupied by polymer chains [22]), which facilitate the migration of ions and increase ion conductivity [23]. Fig. 3a [24] and Fig. 3b [22] show the structure of alginate and the free volume in a polymer, respectively.



**Fig. 3.** (a) Structure of alginate [24] (b) The free volume in a polymer [22] (c) Polymer backbone with salts [27].

The intrinsic proton conductivity of alginates is low because of the absence of a constant transfer pathway and the weak conductivity of the sole polymer [25]. In 2016, Masoud et al. demonstrated an effective way of improving mechanical stability and optimizing water absorption by introducing an inorganic additive [26] (Fig. 3c [27]), which spiked wide research interests in this field.

Concerning the effects of doping  $\text{NH}_4$  salts with alginate-based electrolytes, Rasali et al. investigated the ion conductivity of alginate doped with different amounts of ammonium nitrate ( $\text{NH}_4\text{NO}_3$ ) found that the maximum ion conductivity could be obtained by using 25 wt. %  $\text{NH}_4\text{NO}_3$  at 303K, with its figure of  $5.56 \times 10^{-5} \text{ S cm}^{-1}$  [15].

The maximum ion conductivity was obtained when Fuzlin et al. doped  $\text{NH}_4\text{Br}$  of the various amount, being  $4.41 \times 10^{-5} \text{ S cm}^{-1}$  for a composition of 20 wt. % [28]. Khan et al. also investigate alginate-based electrolyte doped with  $\text{NH}_4\text{Cl}$ , and the maximum ion conductivity was  $3.18 \times 10^{-7} \text{ S cm}^{-1}$  with 8 wt.% of  $\text{NH}_4\text{Cl}$  [29].

As for the effects of doping Li salts with alginate-based electrolytes, Fuzlin et al. and Fuzlin et al. investigated alginate-based electrolytes doped with  $\text{LiNO}_3$  [23] and  $\text{LiBr}$  [30], and the results revealed that conductivity for composition for 15 wt.% of  $\text{LiNO}_3$  overtook that for 15 wt.%  $\text{LiBr}$ , with  $1.14 \times 10^{-4} \text{ S cm}^{-1}$  for the former and  $7.46 \times 10^{-5} \text{ S cm}^{-1}$  for the latter.

**Table 1.** Ion conductivity of alginate doped with ammonium and lithium salts at 303 K.

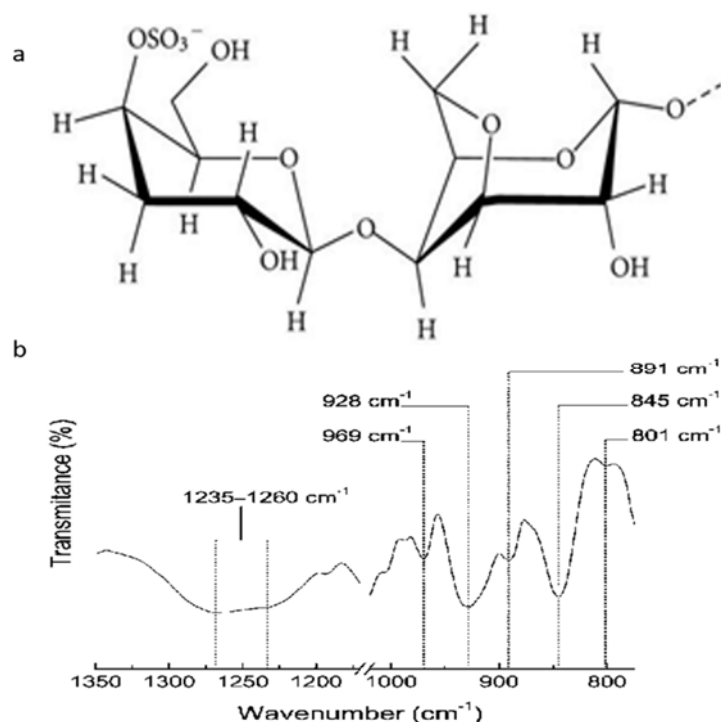
Salts	Ion conductivity/ $\text{S cm}^{-1}$	Ref.
25wt.% $\text{NH}_4\text{NO}_3$	$5.56 \times 10^{-5}$	[15]
20wt.% $\text{NH}_4\text{Br}$	$4.41 \times 10^{-5}$	[28]
8wt.% $\text{NH}_4\text{Cl}$	$3.18 \times 10^{-7}$	[29]
15wt.% $\text{LiNO}_3$	$1.14 \times 10^{-4}$	[21]
15 wt.% $\text{LiBr}$	$7.46 \times 10^{-5}$	[30]

One remarkable advantage of alginate is its relatively low glass transition temperature ( $T_g$ ), mainly due to the semi-rigidity of the carbohydrate backbone chains [31]. However, owing to monovalent cations, alginate is soluble in water, increasing water permeation and leading to the swelling problem. Consequently, it will threaten the ion flow and ion conductivity [32].

In summary, the maximum ion conductivity of  $1.14 \times 10^{-4} \text{ S cm}^{-1}$  in alginate-based electrolytes can be achieved when 15 wt.% of  $\text{LiNO}_3$  is doped with the electrolyte, a fact which renders  $\text{LiNO}_3$  the most suitable salt for an alginate-based electrolyte.

### 2.3 kappa carrageenan-based electrolytes

Extracted from several species of red seaweed from the family Rhodophyceae, carrageenan (CGN) is a polysaccharide with various types [33]. This section will mainly focus on  $\kappa$ -carrageenan, a type of polysaccharide which has an ester sulfate content of about 25 to 30% and a 3,6-AG (amino guanidine) content of about 28 to 35% [34] and a snapshot of kappa carrageenan is shown in Fig. 4a [35].



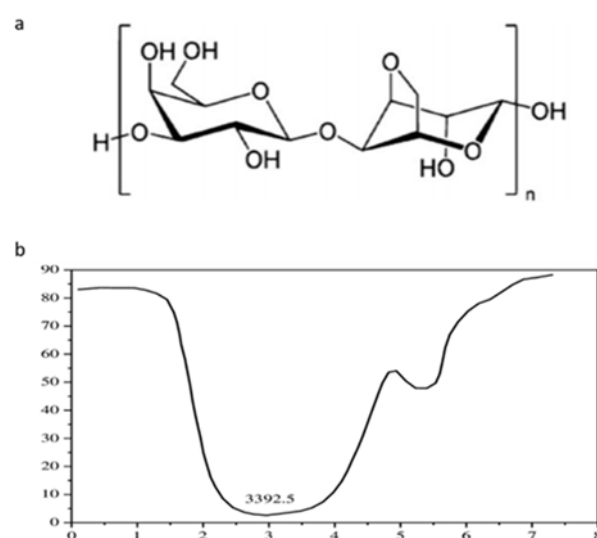
**Fig. 4.** (a) Structure of  $\kappa$ -carrageenan [35] (b) The FTIR spectrum of  $\kappa$ -carrageenan [36]

Owing to the presence of hydroxyl, ether, and sulfate groups in  $\kappa$ -carrageenan that can effectively bind lithium ions,  $\kappa$ -carrageenan becomes a potential candidate for being the electrolyte in lithium-ion batteries [37]. In 1968, Anderson et al. investigated the relationship between the gelling ability of kappa-carrageenan and electrolytes [38], and it was first used as an electrolyte by Fujishima et al. in 2008, [39]. In 2016, the first use of lithium salt as an additive to kappa carrageenan-based electrolyte was demonstrated [40], while that of ammonium salt as an additive was reported by Rani et al. in 2015[41]. So far, all investigations about ion conductivity of kappa carrageenan with ammonium and lithium salts as additive used the traditional solution-casting technique (which involves dissolving the host polymer in hot distilled water and incorporating salts with various concentrations with the polymer solution after magnetically stirring it for an hour [42]).

In terms of the ion conductivity of  $\kappa$ -carrageenan after doping  $\text{NH}_4$  salts, Nithya et al. obtained and maximum ion conductivity of  $2.80 \times 10^{-3} \text{ S cm}^{-1}$  with 30 wt. %  $\text{NH}_4\text{Br}$  [43]. Perumal et al. investigated the ion conductivity of  $\kappa$ -carrageenan doped with different amounts of  $\text{NH}_4\text{COOH}$ , and the maximum result was obtained when 0.4 wt. %  $\text{NH}_4\text{COOH}$  was added, being  $8.54 \times 10^{-4} \text{ S cm}^{-1}$  [44], which is higher than that of  $6.83 \times 10^{-4} \text{ S cm}^{-1}$  (with 0.5% of  $\text{NH}_4\text{SCN}$ ) [45],  $3.89 \times 10^{-4} \text{ S cm}^{-1}$  (with 20 wt.%  $\text{NH}_4\text{Br}$ ) [46], and  $2.99 \times 10^{-4} \text{ S cm}^{-1}$  (with 20 wt.%  $\text{NH}_4\text{Cl}$ ) [47]. When it comes to the effects of doping  $\kappa$ -carrageenan with Li salts, Mary et al. investigated the ion conductivity of  $\kappa$ -carrageenan- based electrolyte doped with LiBr [48] and  $\text{LiNO}_3$  [49], with the maximum ion conductivity being achieved when 0.5 wt.% of LiBr was doped ( $3.43 \times 10^{-3} \text{ S cm}^{-1}$ ) and when 0.65 wt.% of  $\text{LiNO}_3$  was doped ( $1.89 \times 10^{-3} \text{ S cm}^{-1}$ ) at room temperature.

However, doping different salts with kappa

carrageenan-based electrolytes has certain limitations. For example, the open-circuit voltage of a fabricated battery with dopant  $\text{NH}_4\text{Br}$  is relatively high (4.29 V)[43], yet this value decreases to 1.57 V when LiBr is used [46]. However, it shows the highest ion conductivity. In addition, the electrolyte doped with  $\text{NH}_4\text{Cl}$  is advantageous in terms of  $T_g$ , with 29.16 °C for its  $T_g$  [46], but with low ion conductivity. Finally, it is worth mentioning that some literature did not perform the electrochemical stability window test, leading to doubts on the reliability of using kappa carrageenan. All results are summarized in Table 2.



**Fig. 5.** (a) The structure of agarose [51] (b) The FTIR spectrum of agar [54].

**Table 2.** Ion conductivity of kappa carrageenan doped with ammonium and lithium salts at 303 K.

Salts	Ion conductivity/ S cm <sup>-1</sup>	Ref.
0.5 wt.% LiBr	$3.43 \times 10^{-3}$	[48]
30 wt. % NH <sub>4</sub> Br	$2.80 \times 10^{-3}$	[43]
0.65 wt.% LiNO <sub>3</sub>	$1.89 \times 10^{-3}$	[49]
0.4 wt. % NH <sub>4</sub> COOH	$8.54 \times 10^{-4}$	[44]
0.5% of NH <sub>4</sub> SCN	$6.83 \times 10^{-4}$	[45]
20 wt.% NH <sub>4</sub> Cl	$2.99 \times 10^{-4}$	[47]

Concerning the new approach of using kappa carrageenan as the host polymers in electrolytes, Nuneset al. developed an electrolyte composed of kappa carrageenan, 1-butyl-3-methyl-1H-imidazolium chloride ([Bmim] Cl) ionic liquid, and glycerol [37]. The addition of 50 wt.% [Bmin]Cl to kappa carrageenan-based electrolyte increases the ion conductivity from  $5.3 \times 10^{-7}$  S cm<sup>-1</sup> (ion conductivity of pure kappa carrageenan) to  $1.4 \times 10^{-3}$  S cm<sup>-1</sup> at 303 K. The main factor that is attributed to the enhancement in ion conductivity is the greater dissociation of [Bmin]<sup>+</sup> and Cl<sup>-</sup>. As hydrogen bonds between the H atoms of the imidazolium ring and the anion are weakened, large blueshifts (the weakening effect on H-anion bond and the strengthening effect on C-H bond) are observed in ionic liquids, and the distance between [Bmin]<sup>+</sup> and Cl<sup>-</sup> is greater, leading to a complete dissociation. However, the proton transporting mechanism in kappa carrageenan-based electrolyte in acidic conditions and the species of charge carriers in it are still unclear, a fact which limits the potential of using it as a host polymer in an electrolyte.

In conclusion, LiBr is the most suitable salt for κ-carrageenan-based electrolyte, taking the highest ion conductivity of doping it with the electrolyte into account.

## 2.4 Electrolytes based on other seaweed extract polymer

### 2.4.1 Agar-based electrolytes

Agar is the polysaccharide extracted from certain species of red algae [50] (Fig. 5a [51]). Agar is a linear polysaccharide which bonded with 1,3-linked β-d-galactose and 1,4-linked 3–6-anhydro-l-galactose units [52]. Agarose belongs to the gelling part of agar. It can be applied to various fields, including solid, quasi-solid synthesis of gel and agar electrolyte [53].

Selvalakshmi et al. [55-57] studied the performance of agar-based electrolytes with ammonium salts. The ammonium salts are reported to improve ion conductivity [58]. For agar-NH<sub>4</sub>SN polymer electrolyte under 40:60 ratio, at 70°C, the highest ion conductivity approaches 0.86 mS cm<sup>-1</sup>. As for the agar-NH<sub>4</sub>I (70°C) and agar-NH<sub>4</sub>Br (100°C) under the same 50:50 ratio, the conductivity is 0.24 mS cm<sup>-1</sup> and 1.84 mS cm<sup>-1</sup>, respectively. The high ion conductivity of NH<sub>4</sub>Br is

mainly attributed to the 0.3% ethylene carbonate additives, which are used as a cosolvent in the solution [59]. In terms of agar-NH<sub>4</sub>NO<sub>3</sub> electrolyte under 70°C, it has an ion conductivity of 1.09 mS cm<sup>-1</sup> [60]. In all the four cases listed above, the measurements of the highest ion conductivity are taken under the most appropriate temperature due to Arrhenius behaviour. In 2011, Wang et al. investigated the conductivity of the agar-LiI electrolyte for 20 wt.% LiI, under 30°C, the experiment obtained the maximum ion conductivity at  $3.98 \times 10^{-3}$  S cm<sup>-1</sup> [61]. In other evaluations, including 15wt.% agar-2M Li<sub>2</sub>SO<sub>4</sub>, the conductivity can achieve 43 mS cm<sup>-1</sup> and 118 mS cm<sup>-1</sup> respectively [62]. All results are summarized in Table 3.

**Table 3.** Ion conductivity of agar doped with ammonium and lithium salts at various temperatures

Salt	Ion conductivity/ S cm <sup>-1</sup>	Ref.
60 wt.% NH <sub>4</sub> SN	$0.86 \times 10^{-3}$ <sup>a</sup>	[59]
50 wt.% NH <sub>4</sub> I	$0.24 \times 10^{-3}$ <sup>a</sup>	[59]
50 wt.% NH <sub>4</sub> Br <sup>c</sup>	$1.84 \times 10^{-3}$ <sup>b</sup>	[59]
60 wt.% NH <sub>4</sub> NO <sub>3</sub>	$1.09 \times 10^{-3}$ <sup>a</sup>	[59]
20 wt.& LiI	$3.98 \times 10^{-3}$ <sup>d</sup>	[61]
15 wt.% Li <sub>2</sub> SO <sub>4</sub> <sup>d</sup>	$4.3 \times 10^{-2}$	[62]
15 wt.% Li <sub>2</sub> SO <sub>4</sub> <sup>d</sup>	$1.18 \times 10^{-1}$	[62]

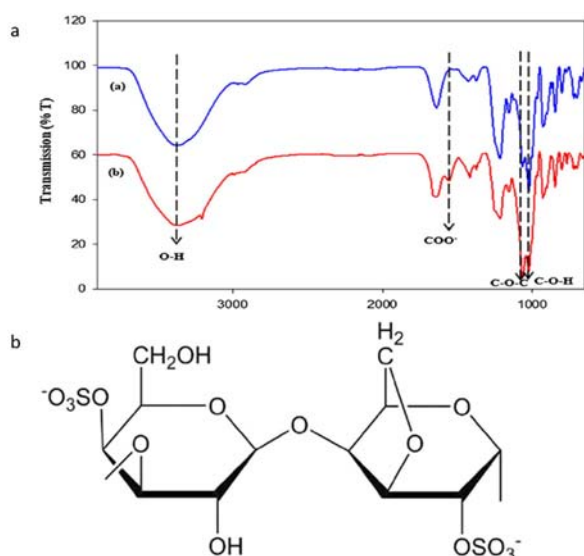
<sup>a</sup> at 70 °C <sup>b</sup> at 100 °C <sup>c</sup> with 0.3% ethylene carbonate <sup>d</sup> at 30 °C

One of the major drawbacks of agar-based electrolyte is its low conductivity at ambient temperature ( $10^{-4}$ – $10^{-5}$  mS/cm). It must be heated to achieve the highest ion conductivity, which may cause inconvenience in practical applications [63].

In summary, we compared the maximum conductivity of each electrolyte and found that the combination of 15 wt% agar - 2M LiClO<sub>4</sub> electrolyte showed the best ion conductivity of 118 mS cm<sup>-1</sup>.

### 2.4.2 Iota carrageenan-based electrolytes

Iota carrageenan, a type of carrageenan that has 28 to 30% ester sulfate content and 25 to 30% 3,6-AG content [34], is famous for its non-toxicity and bio-activity [42] and was first used as a solid electrolyte in 2015 by Jumaah et al. [64]. Its structure is shown in Fig. 6a [34]. All summarized experiments about the ion conductivity of iota carrageenan used the traditional solution-casting technique to prepare membranes.



**Fig. 6.** (a) The ATR-FTIR spectrum of iota carrageenan and the purified iota carrageenan powder [65] (b) The structure of iota carrageenan [42]

Concerning the ion conductivity of iota carrageenan-based electrolyte, the highest value of ion conductivity of the electrolyte can be obtained by using 0.3 wt.% LiCl [66], at  $5.33 \times 10^{-3} \text{ S cm}^{-1}$ , a figure which is followed by  $3.56 \times 10^{-3} \text{ S cm}^{-1}$  obtained by using 0.3 wt.%  $\text{NH}_4\text{SCN}$  [67] and  $3.33 \times 10^{-3} \text{ S cm}^{-1}$  obtained by using 0.5 wt.%  $\text{LiClO}_4$  [42]. The ion conductivities of the rest electrolytes are all below  $2 \times 10^{-3} \text{ S cm}^{-1}$ , with  $1.46 \times 10^{-3} \text{ S cm}^{-1}$  for 0.4 wt.%  $\text{NH}_4\text{NO}_3$  [68],  $1.27 \times 10^{-3} \text{ S cm}^{-1}$  for 0.4 wt.%  $\text{LiCF}_3\text{O}_3$  [69],  $1.11 \times 10^{-3} \text{ S cm}^{-1}$  for 0.4 wt.%  $\text{NH}_4\text{HCO}_2$  [70] and  $1.08 \times 10^{-3} \text{ S cm}^{-1}$  for 0.4 wt.%  $\text{NH}_4\text{Br}$  [71].

It is worth noting that in the work of Chitra et al. (2020) [42], due to its effect on increasing ionic dissociation and ionic mobility, the use of 0.3 wt.% Succinonitrile (SN) plastic crystal additive boosted the ion conductivity sharply given the fact that without using the additive, the ion conductivity can only reach  $3.57 \times 10^{-4} \text{ S cm}^{-1}$ . Also, the use of SN can increase the electrochemical stability window, with the figure increasing from 2.36 V to 3.10 V.

The use of iota carrageenan as the host polymer for

**Table 5.** Ion conductivity of natural algal-based electrolytes and synthetics-based electrolytes doped with ammonium and lithium salts at 303 K

Polymer	Salt	Ion conductivity/ $\text{S cm}^{-1}$	Ref.
Iota carrageenan	0.3wt.% LiCl	$5.33 \times 10^{-3}$	[66]
Agar	20 wt.% LiI	$3.98 \times 10^{-3}$	[62]
Kappa carrageenan	0.5wt.% LiBr	$3.43 \times 10^{-3}$	[48]
Poly(vinyl alcohol)	30wt.% $\text{NH}_4\text{Br}$	$(7.68 \pm 1.24) \times 10^{-4}$	[72]
EDTA/PVP/PVDF	50wt.% LiTFSI	$7.19 \times 10^{-4}$	[73]
GGG/CMC/EC <sup>a</sup>	25 wt.% LiI	$2.10 \times 10^{-4}$	[74]
Alginate	15wt.% $\text{LiNO}_3$	$1.14 \times 10^{-4}$	[23]

<sup>a</sup> guar gum succinate with a concentration of 0.5:0.5:0.6

electrolytes attracts criticism for several reasons. Chief among them is that, unlike kappa carrageenan-based electrolytes, iota carrageenan-based electrolytes are not all fabricated with a battery, thus casting doubt on their effectiveness. Another major factor is the Tg. Despite the constantly high ion conductivity of the electrolytes, their high Tg is the main demerit of using the electrolyte. All collected data are shown in Table 4.

**Table 4.** Ion conductivity of iota carrageenan doped with ammonium and lithium salts at 303 K

Salt	Ion conductivity/ $\text{S cm}^{-1}$	Ref.
0.3 wt.% LiCl	$5.33 \times 10^{-3}$	[66]
0.3 wt.% $\text{NH}_4\text{SCN}$	$3.56 \times 10^{-3}$	[67]
0.5 wt.% $\text{LiClO}_4$ <sup>a</sup>	$3.33 \times 10^{-3}$	[42]
0.4 wt.% $\text{NH}_4\text{NO}_3$	$1.46 \times 10^{-3}$	[68]
0.4 wt.% $\text{LiCF}_3\text{O}_3$	$1.27 \times 10^{-3}$	[69]
0.4wt.% $\text{NH}_4\text{HCO}_2$	$1.11 \times 10^{-3}$	[70]
20 wt.% $\text{NH}_4\text{Br}$	$1.08 \times 10^{-3}$	[71]

<sup>a</sup> with 0.3 wt.% SN

To conclude, the most suitable salt additive for iota carrageenan is LiCl due to the highest ion conductivity of the electrolyte doped with it. Although most iota carrageenan-based electrolytes have a relatively high ion conductivity, the use of the electrolyte has limitations, such as high Tg.

## 2.5 Comparison between electrolytes based on natural algal polysaccharide and electrolytes based on synthetic materials

To demonstrate the effectiveness of using algal as the host polymer of electrolytes to enhance the ion conductivity, we summarize the ion conductivity of some synthetic-based electrolytes and that of algal-based electrolytes doped with ammonium and lithium salts in Table 5.

It can be seen that the use of iota carrageenan as

electrolytes gives the highest ion conductivity, followed

by that of kappa carrageenan, agar, synthetics, and alginate. Therefore, the use of iota carrageenan as the host polymer of an electrolyte seems to be the most successful.

### 3 Conclusion

In conclusion, we reviewed the history and recent progress of seaweed extractions, including alginate, kappa carrageenan, and other seaweed extracts (agar and iota carrageenan), as a new class of polymer electrolytes for lithium-ion batteries. For alginate-based electrolyte, its main drawback is the relatively low ion conductivity while it gains competitiveness as its  $T_g$  is relatively low. The most suitable salt for the electrolyte is  $\text{LiNO}_3$ . For kappa carrageenan-based electrolyte, its high ion conductivity renders it a more potential electrolyte while it has a relatively high  $T_g$ . The most suitable salt for the electrolyte is  $\text{LiBr}$ . When it comes to agar-based electrolytes and iota carrageenan-based electrolytes, the former is less competitive due to its low ion conductivity. At the same time, the latter has the vital merit of high ion conductivity and a demerit of high  $T_g$ . The most suitable salts are  $\text{NH}_4\text{Br}$  and  $\text{LiCl}$ , respectively.

Despite progress made in enhancing the ion conductivity of electrolytes, much work lays ahead. One of the main problems is the limited number of measured indicators of ion conductivity. The ion conductivity of a host polymer is determined by many factors, such as dielectric constants, molecular weights, degrees of amorphous phase, glass transition temperatures and backbone flexibility. However, backbone flexibility is merely reported in most literature, and a proportion of them did not report  $T_g$  and dielectric constants. Therefore, more efforts are needed to further investigate these parameters to find correlations between these variables and develop methods to optimize them to achieve higher ion conductivities. It is also worthy of mentioning that algal-based electrolytes with high ionic conductivity normally have a high  $T_g$ . Hence, algal-based electrolytes will be more potential if  $T_g$  can be lowered without compromising the ion conductivity and other advantageous physical properties.

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