Supporting Information

Preparation of LAGP/P(VDF-HFP) polymer electrolytes for Li-ion batteries

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Characterization

The BET N_2 absorption/desorption characterization was performed using the surface area and pore size analyzer (Quantachrome, Quadrasorb *SI*). The energy dispersive spectrometer (EDS) of the polymer electrolytes were measured through SEM (Hatchi S-4800). Element analysis was calculated from inductively coupled plasma atomic emission spectrometry (ICP-AES) (Thermo Fisher, i CAP6300). FTIR measurements were carried out on the infrared spectrometer (Thermo Fisher, Nicolet 6700).

Supplementary figures

The BET N_2 absorption/desorption data was shown in Table S1 and S2. The BET surface area of P(VDF-HFP) is 0.253 m^2 g^{-1}, and is smaller than 0.274 m^2 g^{-1} of LAGP/P(VDF-HFP). The result demonstrated that the composite membranes with LAGP show more enormous number of micropores compared with pure P(VDF-HFP), which helps with entrapping or storing a large amount of liquid solution (salt and plasticizer mixtures). The composite GPEs with large surface area enlarge the contact area between the polymer and liquid electrolyte which ensures that the electrolyte is well retained in the polymer membranes. This is favorable to improve the ionic conductivity and other electrochemical performance.

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Table S1 The BET N_2 absorption/desorption data of P(VDF-HFP)

Relative Pressure P/P_0	Volume (mL g^{-1})	$1/[W((P_0/P)-1)]$
1.07120e-02	0.0279	3.1005e+02
2.34114e-02	0.0466	4.1119e+02
2.81824e-02	0.0508	4.5650e+02
4.27926e-02	0.0411	8.7130e+02
5.23878e-02	0.0467	9.4653e+02
7.76769e-02	0.0596	1.1306e+03
BET summary Slope = 13611.018 Intercept = 1.542e+02 Correlation coefficient, r = 0.962893 C constant= 89.256 Surface Area = 0.253 m^2 g^{-1}		

Table S2 The BET N_2 absorption/desorption data of LAGP/P(VDF-HFP)

Relative Pressure P/P_0	Volume (mL g^{-1})	$1/[W((P_0/P)-1)]$
4.28044e-02	0.0170	2.1089e+03
5.27335e-02	0.0226	1.9684e+03
7.79852e-02	0.0327	2.0676e+03
1.03186e-01	0.0272	3.3819e+03
1.52712e-01	0.0372	3.8747e+03
2.02829e-01	0.0486	4.1851e+03
2.52973e-01	0.0691	3.9233e+03
BET summary Slope = 11021.385 Intercept = 1.679e+03 Correlation coefficient, r = 0.885918 C constant= 7.564 Surface Area = 0.274 m^2 g^{-1}		

To further prove the existence of LAGP, we made the EDS characterization as shown in Fig. S1. The result shows that the elementary composition of the hybrid consisting of C, O, F, Al, Ge and P. The existence of Al, Ge, P and O elements prove the existence of LAGP.
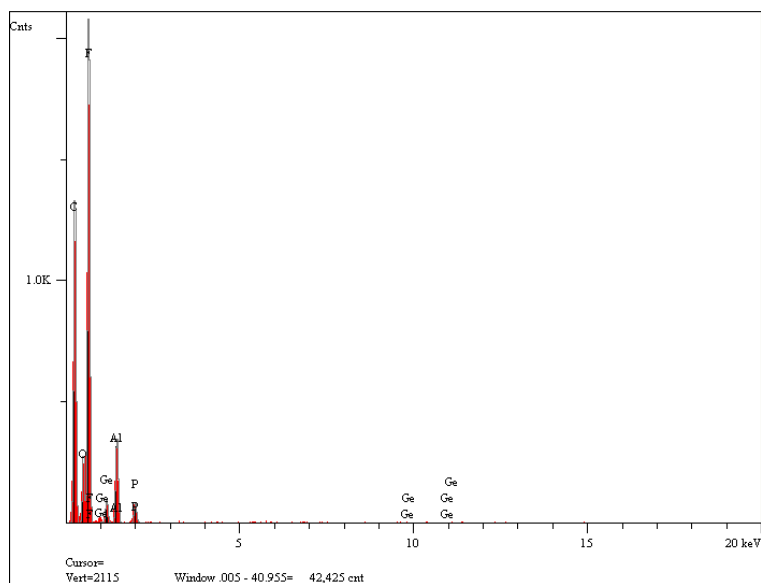
Figure S1 EDS of LAGP/P(VDF-HFP) composite polymer membranes

Table S3 The content of the lithium element in LAGP

Analysis line	Li6707
Weight of Sample/g	0.35
Volume/mL	250
Dilution multiple	1
Standard quantity /ppm	20
Reading of the standard 1	12.03
Reading of the sample	0.963
Element content/%	0.114357
Reading of the standard 2	12.03
Element content/%	0.114357

In the preparation of LAGP/P(VDF-HFP) electrolytes, the LAGP content in the composite was kept at 5 wt.% with respect to P(VDF-HFP). The weight percentage of LAGP in the composite is about 4.76%. To calculate the percentage of LAGP, element analysis was carried out by figuring up the content of lithium on ICP-AES. As is seen from Table S3, the content of the lithium element in the sample is 0.114357%. Because the weight percent of lithium in LAGP ($Li_{1.5}Al_{0.5}Ge_{1.5}(PO_4)_3$) can be calculated by the following equation $(7×1.5)/(7×1.5+27×0.5+72.6×1.5+95×3)=10.5/417.9=2.5\%$, the percentage of LAGP in the

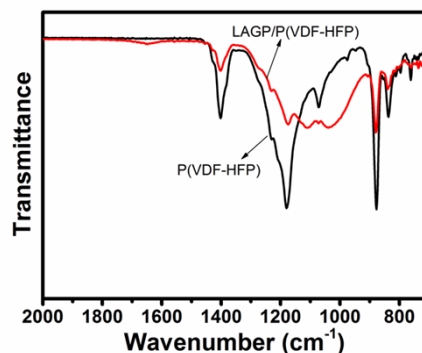sample is 0.114357%/2.5% =4.57% (close to 4.76%).

Fig. S2 FTIR of LAGP/ P(VDF-HFP) and P(VDF-HFP) polymer membranes

To investigate the interaction between LAGP and polymer matrix P(VDF-HFP), the FTIR spectra was employed to confirm the structure of the polymer electrolytes with LAGP. It is obviously observed from Fig. S2, the peaks at 1176 and 1403 cm^{-1} are assigned to the symmetrical stretching of $-CF_2$ and $-CH_2$ groups, respectively. The peak at 879 cm^{-1} is assigned to the vinylidene group of the polymer (Ref.41). It is worthy to note that the absorption bands around 1176 cm^{-1} get weaker and shift to lower wavenumber after the addition of LAGP, indicating that intermolecular forces exist between LAGP and P(VDF-HFP) matrix. Thus, the interactions increase the compatibility of the units in P(VDF-HFP)