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## **Supplementary Information for**

# The effect of SiO<sub>2</sub> additives on solid hydroxide ion-conducting polymer electrolytes: A Raman microscopy study

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### Comparison of SiO<sub>2</sub> powders

SiO <sub>2</sub>	Particle size	Density (g mL <sup>-1</sup> )	Surface Area (m <sup>2</sup> g <sup>-1</sup> )
Micro-sized	44 μm	2.2	300-350
Nano-sized	7 nm	2.3	370-420

Table S1: Physical properties of mSiO<sub>2</sub> and nSiO<sub>2</sub>.



Figure S1: XPS spectra of (A) Si2p and (B) O1s of mSiO<sub>2</sub> and nSiO<sub>2</sub>.



Figure S2: (A) Raman spectra and (B) IR spectra of mSiO<sub>2</sub> and nSiO<sub>2</sub> powders.

Table S1 shows the physical properties of the two  $SiO_2$  powders provided by the supplier. The main difference between the two is the particle size. XPS measurements were performed on the

SiO<sub>2</sub> powders to study their surface chemistries as shown in Fig. S1. The values of the binding energies still fall within the range of SiO<sub>2</sub> as reported in the literature.<sup>1–3</sup> Fig. S2 shows the Raman and FTIR spectra of mSiO2 and nSiO2 powders. No chemical stretches were observed from the Raman measurements (Fig. S2A), indicating no unexpected functionalities in the powders. Additionally, the presence of -OH functionality was excluded as the FTIR spectra of both powders revealed no signature in the vibrational frequency region for O-H stretching (Fig. S2B).



#### Raman spectra fitting for liquid and solid TEAOH

Figure S3: Raman spectra peak fitting of  $TEAOH_{(aq)}$  and  $TEAOH_{(s)}$  and their respective content of conformations.

Fig. S3 shows Raman spectra of liquid and solid TEAOH in the region between 600 and 750 cm<sup>-1</sup>. This peak belongs to  $C_4N$  stretch of the TEA cation. The peak split of 672 and 682 cm<sup>-1</sup> corresponds to the *trans-gauche* (tg.tg) and all-*trans* (tt.tt) conformation of TEA, respectively. Here we see that the area ratio gives an indication of how much of the conformation is present in each system.

## Optical images of electrolytes during crystallization



Figure S4: Optical images of the dendrite like crystal growth in (A) TEAOH-PAM, (B) TEAOH-PAM-mSiO<sub>2</sub>, and (C) TEAOH-PAM-nSiO<sub>2</sub>

## References

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