Broadband dielectric behavior of MIL-100 metal-organic framework as a function of structural amorphization

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1. Powder X-ray diffraction (XRD)

Figure S1: The XRD patterns for: (a) Basolite F300 and (b) MIL-100-MG pellets, both normalized with respect to the highest data point. (c) XRD patterns in absolute intensities for

MIL-100-MG pellets. Inset of table shows the pellet crystallinity (%), estimated from the area ratio of the crystalline peaks to the total area found under the XRD pattern.



2. Fourier-transform infrared spectroscopy (ATR-FTIR)



Figure S2: (a) The pressure-dependent ATR-FTIR spectra for Basolite F300 pellets, (b) Derivative of peak shift over pelleting pressure.



3. Thermogravimetric analysis (TGA)

Figure S3: The pressure-dependent thermal stability measurement (TGA) for Basolite F300 pellets.

4. Dielectric properties

4.1 Basolite F300

4.1.1 Real Part of Dielectric Constant (ε')







Figure S4: The real part of dielectric constant for Basolite F300 pellets prepared under a compression load of: (a) 0.5-ton, (b) 1-ton, (c) 2-ton, (d) 3-ton (e) 5-ton (f) 7-ton, and (g) 10-ton, corresponding to the pressure of 36.96, 73.92, 147.84, 221.76, 369.6, 517.44 and 739.20 MPa, respectively.



4.1.2 Imaginary Part of Dielectric Constant (ε'')





Figure S5: The imaginary part of dielectric constant for Basolite F300 pellets prepared under a compression load of: (a) 0.5-ton, (b) 1-ton, (c) 2-ton, (d) 3-ton (e) 5-ton (f) 7-ton, and (g) 10-ton, respectively.

4.1.3 Dielectric Loss (tan δ)





Figure S6: The dielectric loss for Basolite F300 pellets prepared under a compression load of: (a) 0.5-ton, (b) 1-ton, (c) 2-ton, (d) 3-ton (e) 5-ton (f) 7-ton, and (g) 10-ton, respectively.

4.2 MIL-100-MG

4.2.1 Real Part of Dielectric Constant

Figure S7: The real part of dielectric constant for MIL-100-MG pellets prepared under a compression load of: (a) 0.5-ton, (b) 1-ton, (c) 2-ton, (d) 3-ton (e) 5-ton (f) 7-ton, and (g) 10-ton, corresponding to the pressure of 36.96, 73.92, 147.84, 221.76, 369.6, 517.44 and 739.20 MPa, respectively.

4.2.2 Imaginary Part of Dielectric Constant

Figure S8: The imaginary part of dielectric constant for MIL-100-MG pellets prepared under a compression load of: (a) 0.5-ton, (b) 1-ton, (c) 2-ton, (d) 3-ton (e) 5-ton (f) 7-ton, and (g) 10-ton, respectively.

4.2.3 Dielectric Loss

Figure S9: The dielectric loss for MIL-100-MG pellets prepared under a compression load of: (a) 0.5-ton, (b) 1-ton, (c) 2-ton, (d) 3-ton (e) 5-ton (f) 7-ton, and (g) 10-ton, respectively.

4.3 Comparative dieletric loss

Figure S10: Dielectric loss of MOF pellets as a function of pelleting pressure and temperature: (a) Basolite F300 and, (b) MIL-100-MG pellets specific frequencies (0.01, 0.1, and 1 MHz).

5. Reflectivity spectra $R(\omega)$ in the far-IR and mid-IR regions

Figure S11: Reflectance spectra of MIL-100-MG pellets. Inset: (a) reflectivity spectra in the near-IR region, (b) joining of the far-IR and mid-IR spectra at 517 cm⁻¹ (~15.5 THz), (c) pelleting pressure-dependent redshift in transition mode of peaks obtained from the Gaussian peak fitting and (d) plot for pressure-dependent peak shift in peak positions.

6. Refractive index in THz region

Figure S12: (a) Real and (b) imaginary parts of the refractive index of MIL-100 pellets in IR frequency range. Inset (c)-(d) shows the optically insensitivity of the framework in near-IR region.

7. The imaginary part of dielectric constant in the THz region

Figure S13: The imaginary part of dielectric constant (ε'') in the IR frequency range for MIL-100 pellets. Inset (a) shows the ε'' spectrum in the near-IR region. Inset (b) doesn't show any shift in the transition mode, whereas, in inset (c)-(d) the redshift is evident in the transition modes caused by the pelleting force-induced amorphization.

8. AC conductivity

8.1 Basolite F300

Figure S14: The AC conductivity of Basolite F300 pellets prepared under a compression load of: (a) 0.5-ton, (b) 1-ton, (c) 2-ton, (d) 3-ton (e) 5-ton (f) 7-ton, and (g) 10-ton, corresponding to the pressure of 36.96, 73.92, 147.84, 221.76, 369.6, 517.44 and 739.20 MPa, respectively.

8.2 MIL-100-MG

Figure S15: The AC conductivity of MIL-100-MG pellets prepared under a compression load of: (a) 0.5-ton, (b) 1-ton, (c) 2-ton, (d) 3-ton (e) 5-ton (f) 7-ton, and (g) 10-ton, corresponding to the pressure of 36.96, 73.92, 147.84, 221.76, 369.6, 517.44 and 739.20 MPa, respectively.