J. Mater. Chem A.

Supporting Information

Achieving Ultrahigh Energy Storage Density at Low Operating Fields in Lead Hafnate-Based Novel Perovskite Solid Solutions

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Figure S.1. (a-e) Room temperature variation of relative permittivity as a function of frequency of the (1-x)$PbHfO_3$-x$AgNbO_3$ ceramics: (a) x = 0.01, (b) x = 0.02, (c) x = 0.025, (d) x = 0.027 and (e) x = 0.03.

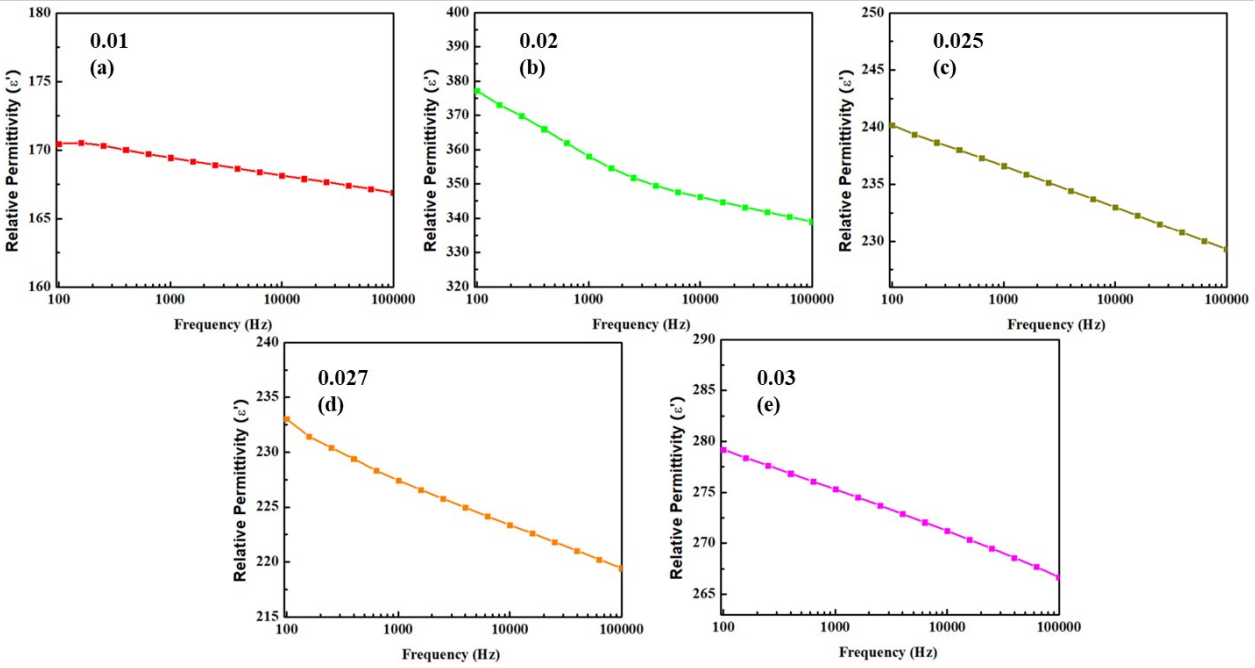
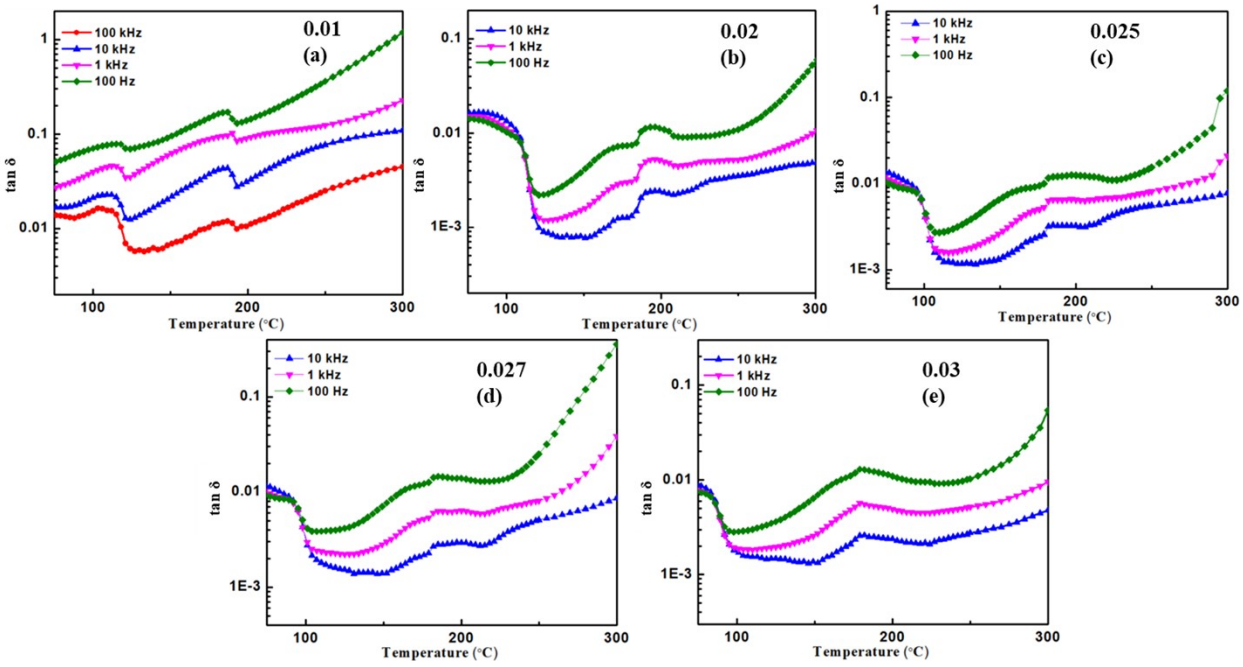
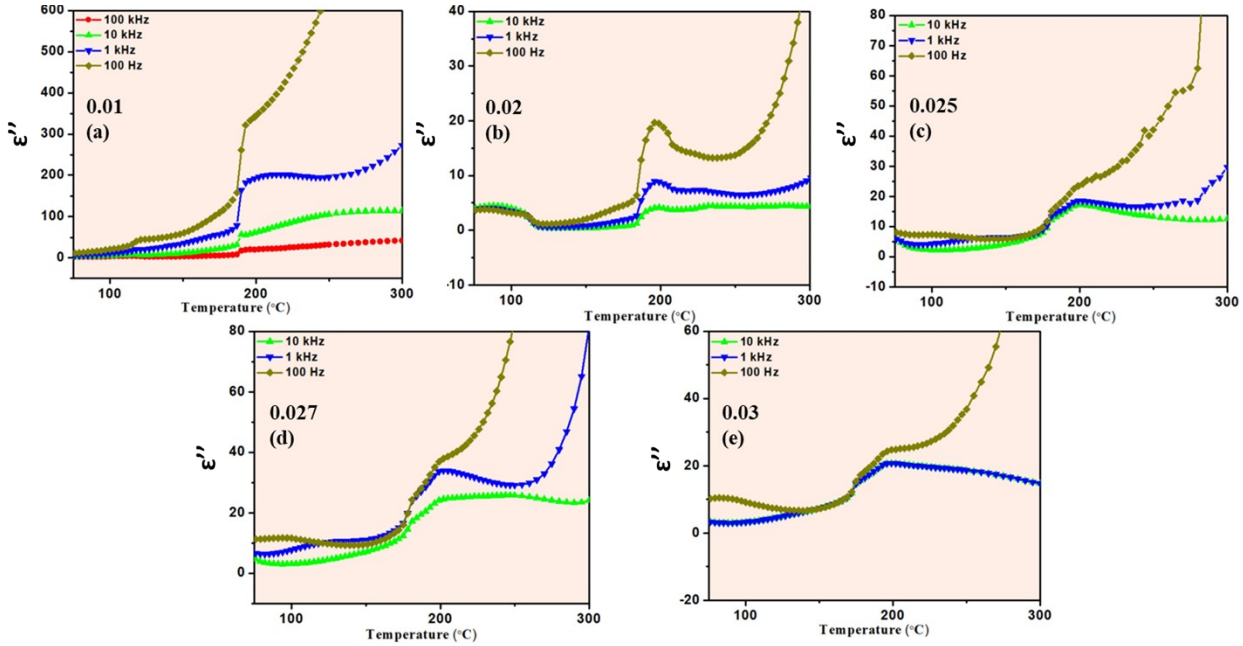


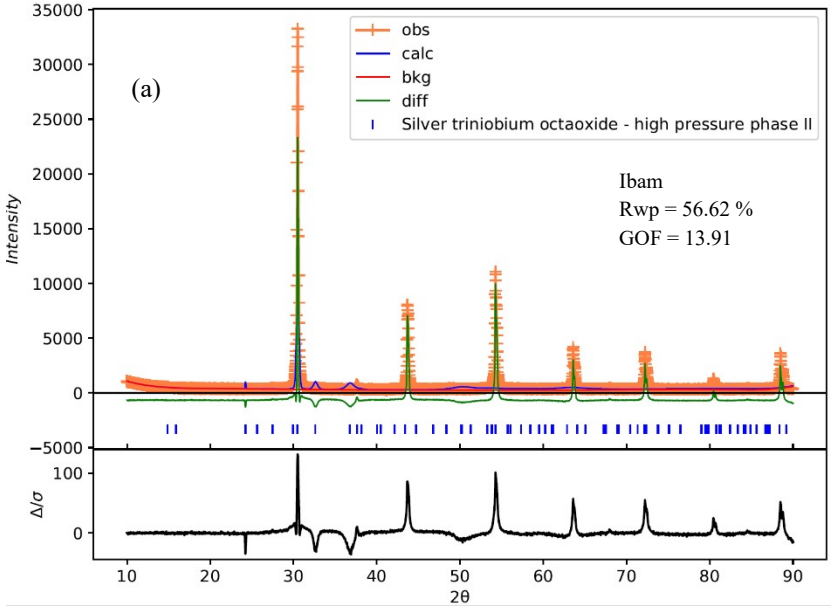
Figure S.2. (a-e) Temperature dependencies of loss tangent of the (1-x)$PbHfO_3$-x$AgNbO_3$ ceramics measured at various frequencies : (a) x = 0.01, (b) x = 0.02, (c) x = 0.025, (d) x = 0.027 and (e) x = 0.03.

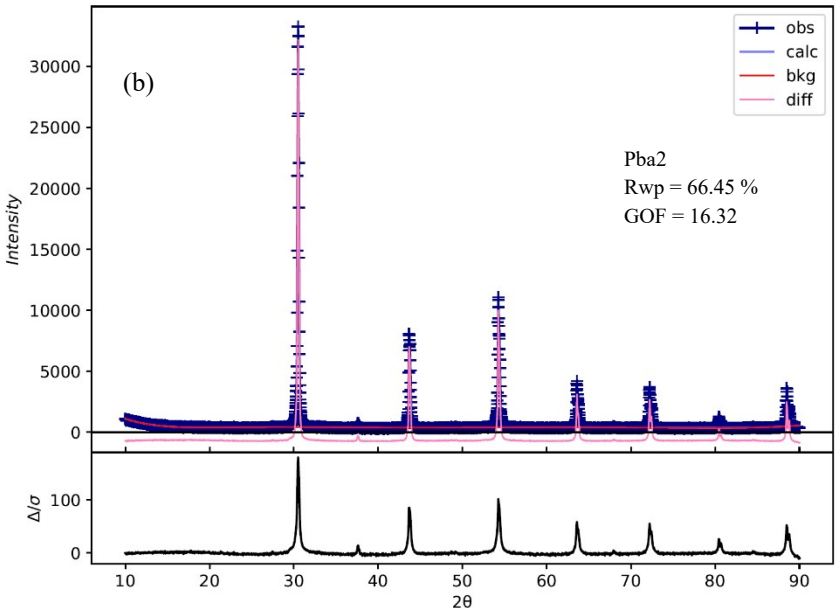


The temperature and frequency dependencies of loss tangent of the as-sintered (1-x)$PbHfO_3$-x$AgNbO_3$ ceramics are presented in Figure S-2 in logarithmic scale. Two sharp anomalies are clearly visible in the loss tangent data, which represent T_{C1} and T_{C2}.

Figure S.3. (a-e) Variation of Imaginary part of permittivity (ε'') as a function of temperature for various compositions of the (1-x)$PbHfO_3$-x$AgNbO_3$ ceramics.




(a)
obs
calc
bkg
diff
Silver triniobium octaoxide - high pressure phase II
Ibam
Rwp = 56.62 %
GOF = 13.91
Intensity
Δ/σ
2θ


(b)
obs
calc
bkg
diff
Pba2
Rwp = 66.45 %
GOF = 16.32
Intensity
Δ/σ
2θ

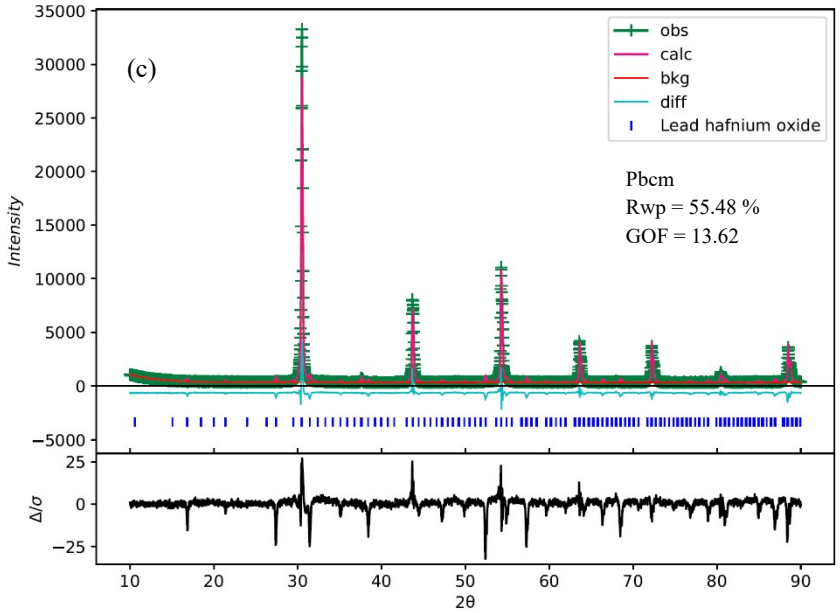


Figure S.4. (a-c) Rietveld refinement results of the $0.973PbHfO_3$-$0.027AgNbO_3$ ceramics measured at 195 °C by using different structural models: (a) Ibam, (b) Pba2 and (c) Pbcm.

Table S.1 Refined structural parameters of $0.973PbHfO_3$-$0.027AgNbO_3$ ceramics at different temperatures

Temperature	Space group	Ions	Positional Coordinates X	Y	Z	Cell Parameters (Å)
25 °C	Pbam	Pb^{2+} (1)	0.7157	0.1303	0.0000	a = 5.8383 (1)
		Pb^{2+} (2)	0.7136	0.1247	0.5000	b = 11.6901 (2)
		Hf^{4+}	0.2449	0.1244	0.2512	c = 8.1911 (1)
		O^{2-} (1)	0.2859	0.0982	0.0000	
		O^{2-} (2)	0.2681	0.1532	0.5000	
		O^{2-} (3)	0.0229	0.2568	0.2222	
		O^{2-} (4)	0.0000	0.5000	0.2880	
		O^{2-} (5)	0.0000	0.0000	0.2692	
		Ag^{1+} (1)	0.7157	0.1303	0.0000	
		Ag^{1+} (2)	0.7136	0.1247	0.5000	
		Nb^{5+}	0.2449	0.1244	0.2512	
105 °C	Imma	Pb^{2+}	0.0120 (7)	0.0410 (4)	0.0330 (5)	a = 5.8431 (3)
		Hf^{4+}	0.0220 (5)	0.008 (3)	-0.0070 (4)	b = 8.2110 (2)
		O^{2-} (1)	0.610 (3)	0.2500 (8)	-0.0200 (9)	c = 5.8379 (2)
		O^{2-} (2)	-0.153 (3)	0.020 (1)	0.000 (3)	
		Ag^{1+}	0.0120 (7)	0.0410 (4)	0.0330 (5)	
		Nb^{5+}	0.0220 (5)	0.0088 (30)	-0.0070 (4)	
175 °C	Imma	Pb^{2+}	0.0100 (7)	0.0620 (3)	0.0220 (7)	a = 5.8416 (2)
		Hf^{4+}	0.0250 (6)	0.003 (3)	-0.0010 (5)	b = 8.2381 (2)
		O^{2-} (1)	0.01800	0.00900	0.00140	c = 5.8362 (2)
		O^{2-} (2)	0.01800	0.03100	0.01000	
		Ag^{1+}	0.0100 (7)	0.0620 (3)	0.0220 (7)	
		Nb^{5+}	0.0250 (6)	0.003 (3)	-0.0010 (5)	

195 °C	Imma	Pb^{2+}	0.040 (1)	0.056 (1)	0.0150 (7)	a = 5.8430 (5)
		Hf^{4+}	0.0230 (8)	0.0120 (9)	-0.0030 (6)	b = 8.2522 (5)
		O^{2-} (1)	0.0180	0.0090	0.0014	c = 5.8366 (3)
		O^{2-} (2)	0.0180	0.0310	0.0110	
		Ag^{1+}	0.0400 (10)	0.0560 (10)	0.0150 (7)	
		Nb^{5+}	0.0230 (8)	0.0120 (9)	-0.0030 (6)	
205 °C	Imma	Pb^{2+}	0.0168	0.0291	0.0201	a = 5.8428 (4)
		Hf^{4+}	0.0140 (9)	0.013 (1)	0.0000 (7)	b = 8.2552 (7)
		O^{2-} (1)	-0.9500 (9)	0.5300 (9)	0.5100 (9)	c = 5.8363 (4)
		O^{2-} (2)	-0.7700 (5)	-0.2200 (4)	0.5600 (6)	
		Ag^{1+}	0.0168	0.0291	0.0201	
		Nb^{5+}	0.0140 (9)	0.013 (1)	0.0000 (7)	