

## ESI

### **A facile and efficient strategy for design of ferroelectric and giant dielectric hybrids via intercalating polar molecules into noncentrosymmetric layered inorganic compound**

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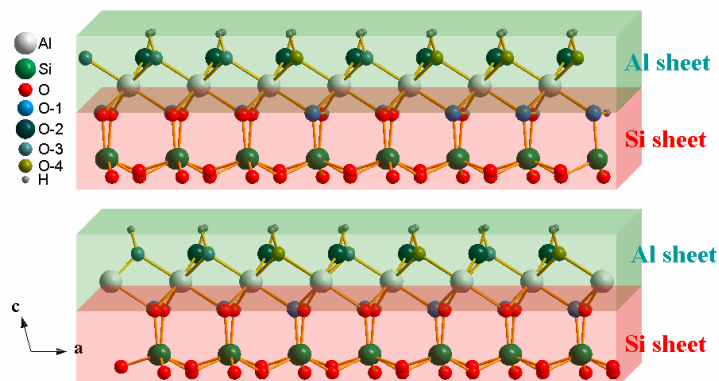
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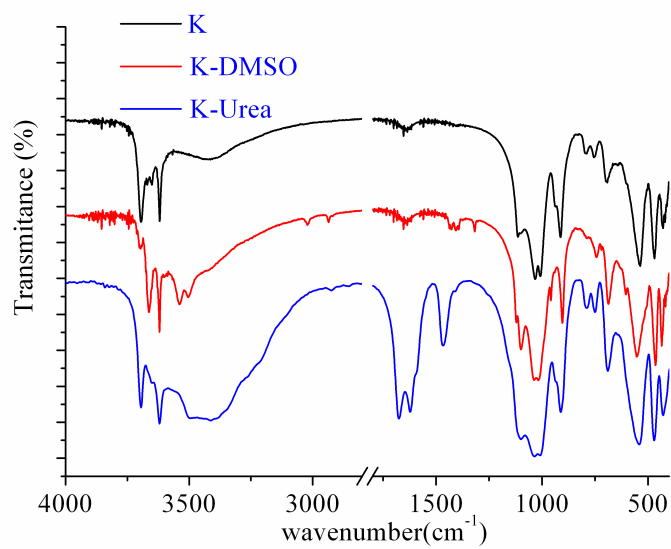
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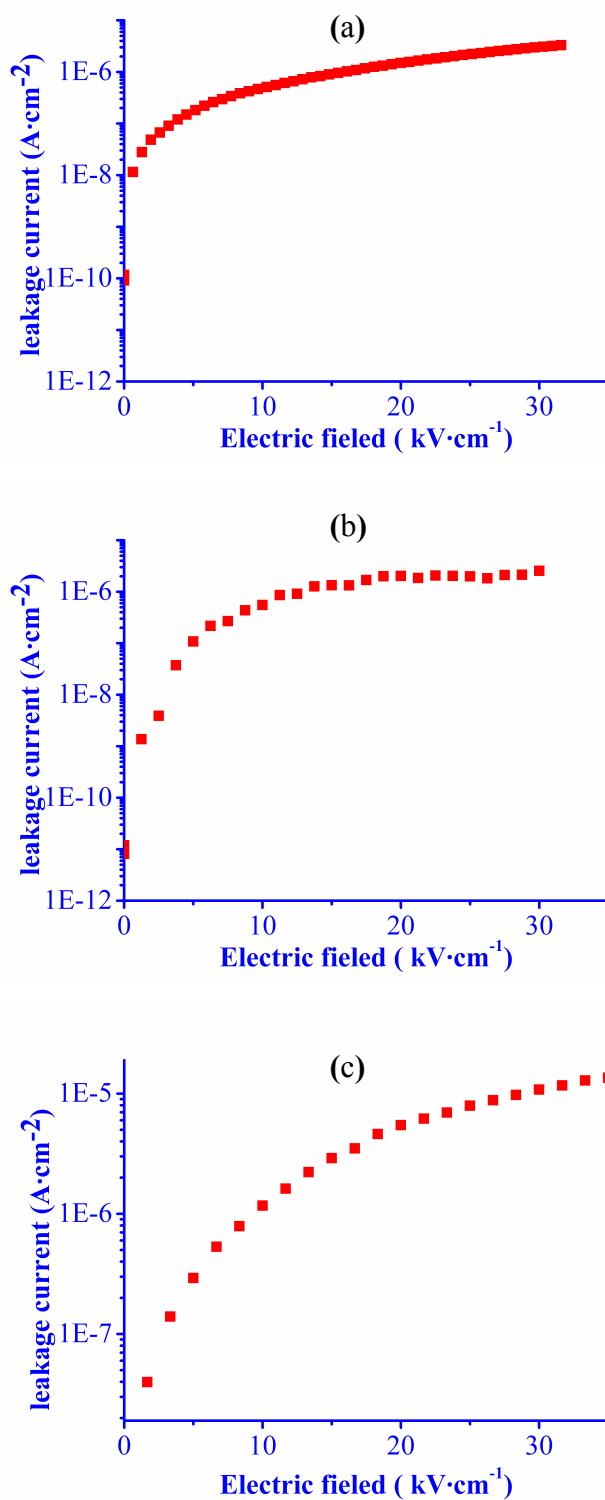
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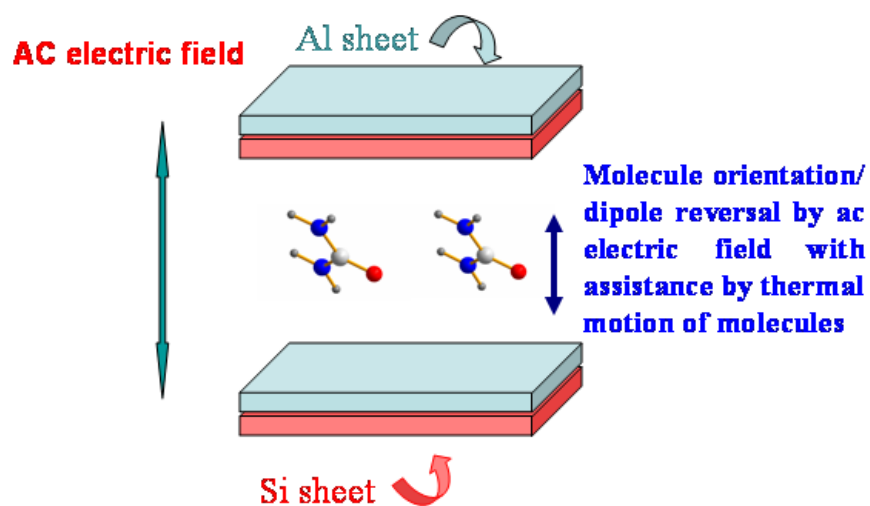
**Figure S1** Layered structure of Kaolinite.



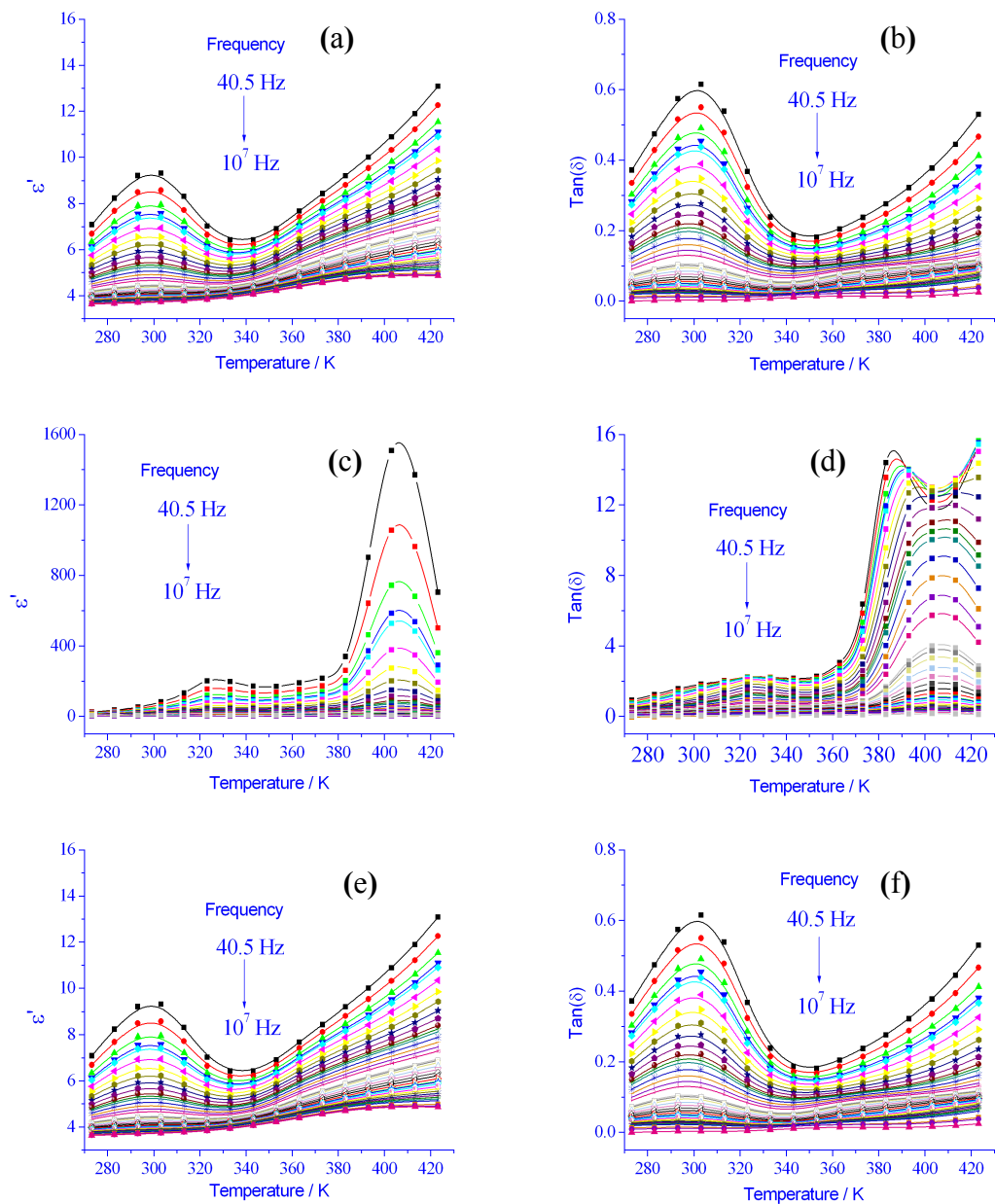
**Figure S2** IR spectra of intercalated and raw Kaolinites.



**Figure S3** Plots of leakage current versus applied electric field for (a) K-DMSO, (b) K-Urea samples and (c) raw Kaolinite.



**Figure S4** Schematic illustration for the dipole reversal of urea molecules resulted from the motions of urea molecule flipping motion by electric field with assistance by thermal motion of molecules for K-Urea.



**Figure S5** Temperature dependence of  $\epsilon'$  and  $\tan\delta$  of (a, b) K-DMSO (c, d) K-Urea intercalation compounds and (e, f) raw Kaolinite at  $f = 40.5 \sim 10^7$  Hz and 273~423K.