

# Novel dielectric relaxation behaviors driven by host-guest interactions in intercalated compounds of Kaolinite with amino-pyridine derivatives

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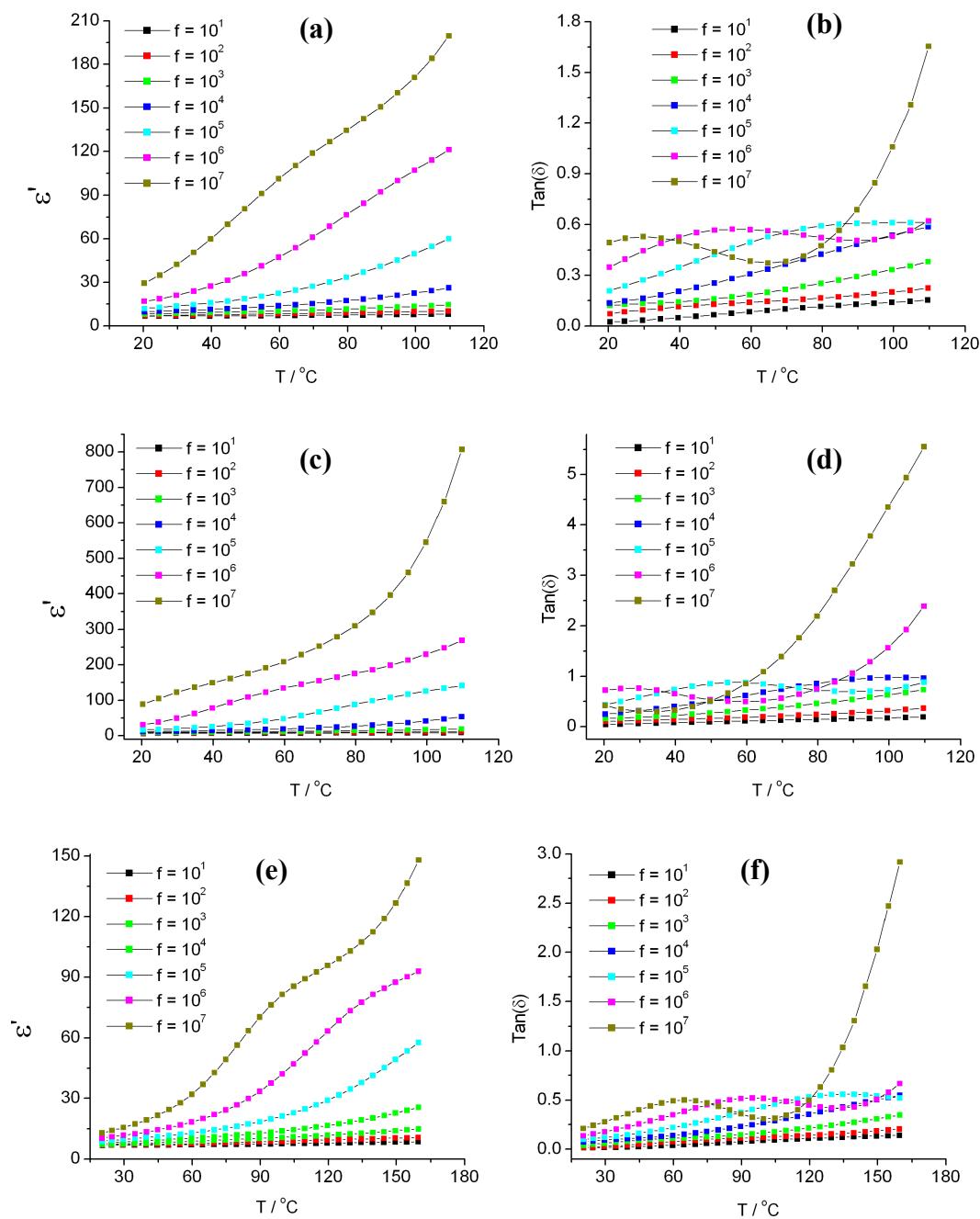
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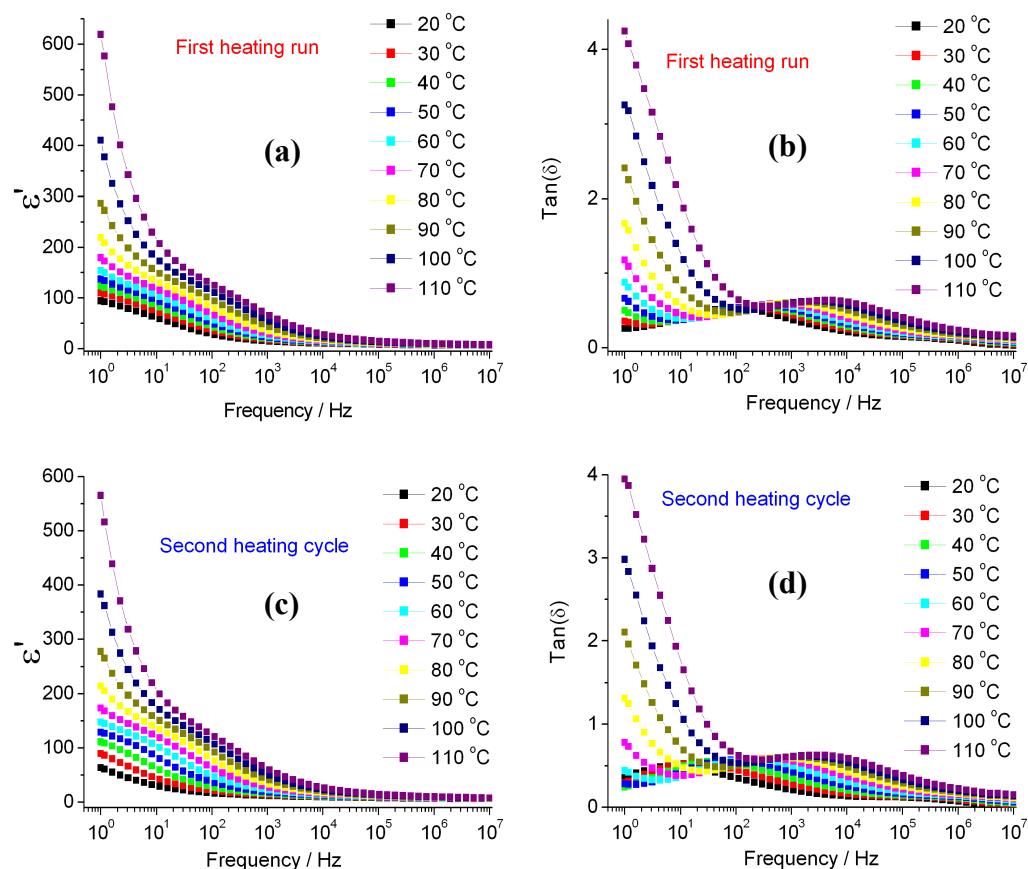
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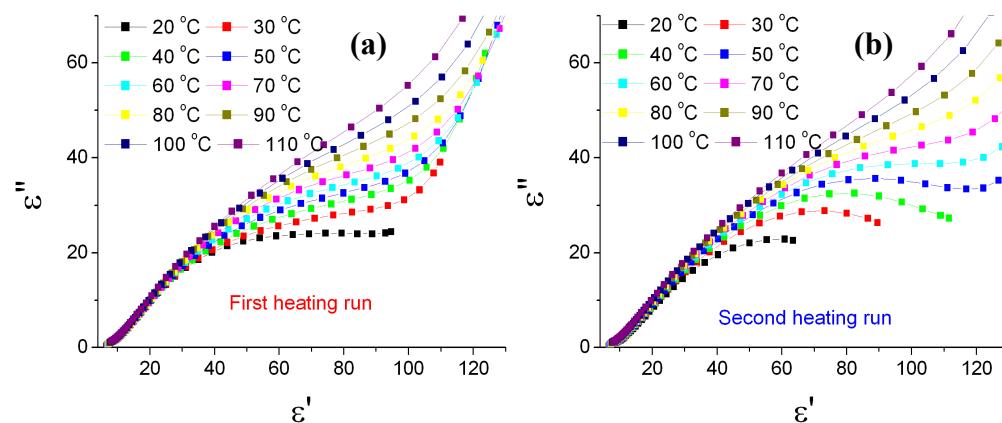
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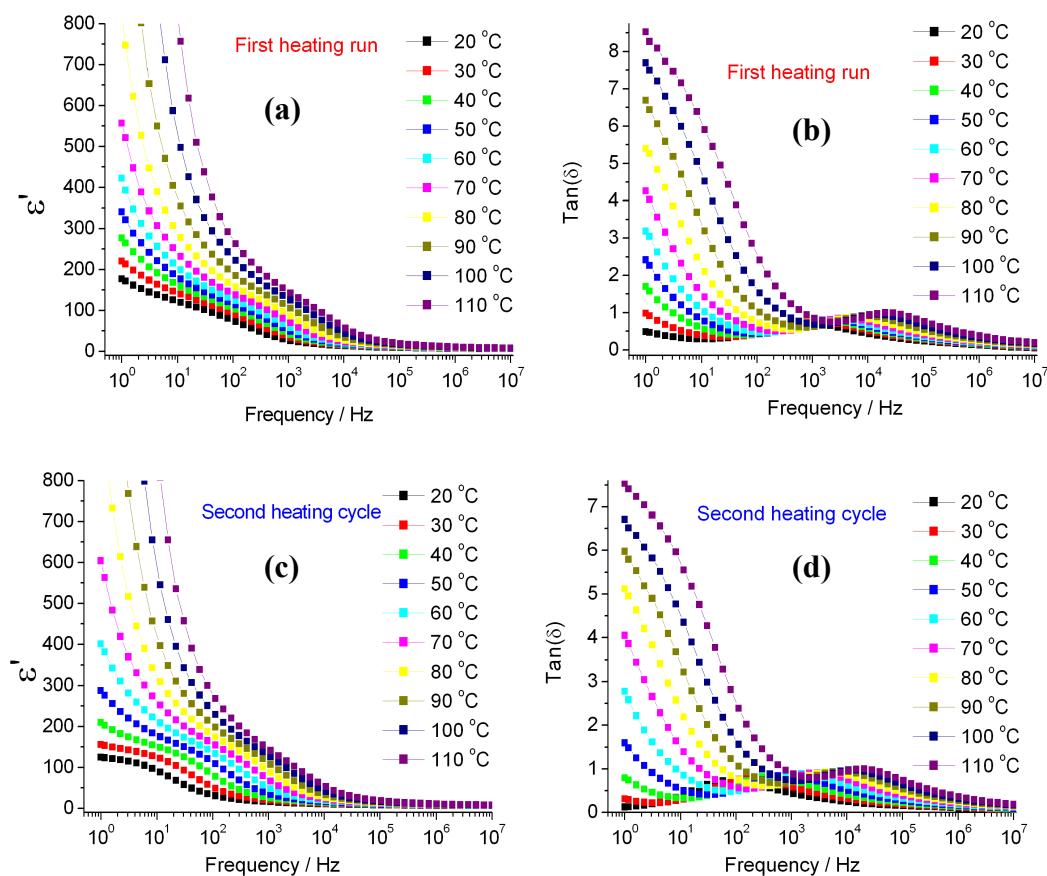
**Figure S1** Temperature dependences of  $\epsilon'$  and  $\tan(\delta)$  in the frequency range of  $1-10^7\text{ Hz}$  for (a, b) **2-APy-K** (c, d) **3-APy-K** and (e, f) **4-APy-K** in the second heating run.



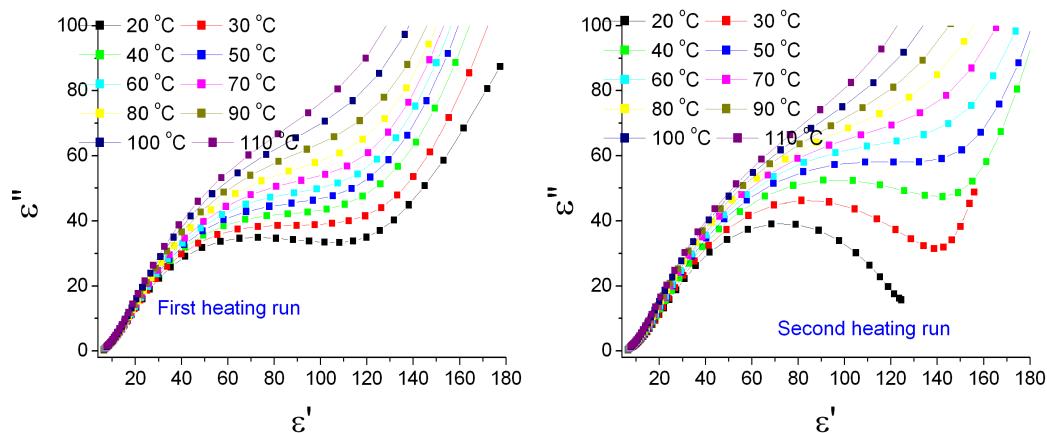
**Figure S2** Frequency dependences of  $\epsilon'$  and  $\text{tan}(\delta)$  in the temperature range of 20–110 °C for 2-APy-K in the first heating cycle and the second heating cycle. Both plots of  $\epsilon'$ -f and  $\text{tan}(\delta)$ -T in two sequential heating cycles are quite similar to each other, indicating that the intercalant molecules were not de-intercalated at 110 °C even though the measurement temperature is only 2 °C below the de-intercalation temperature).



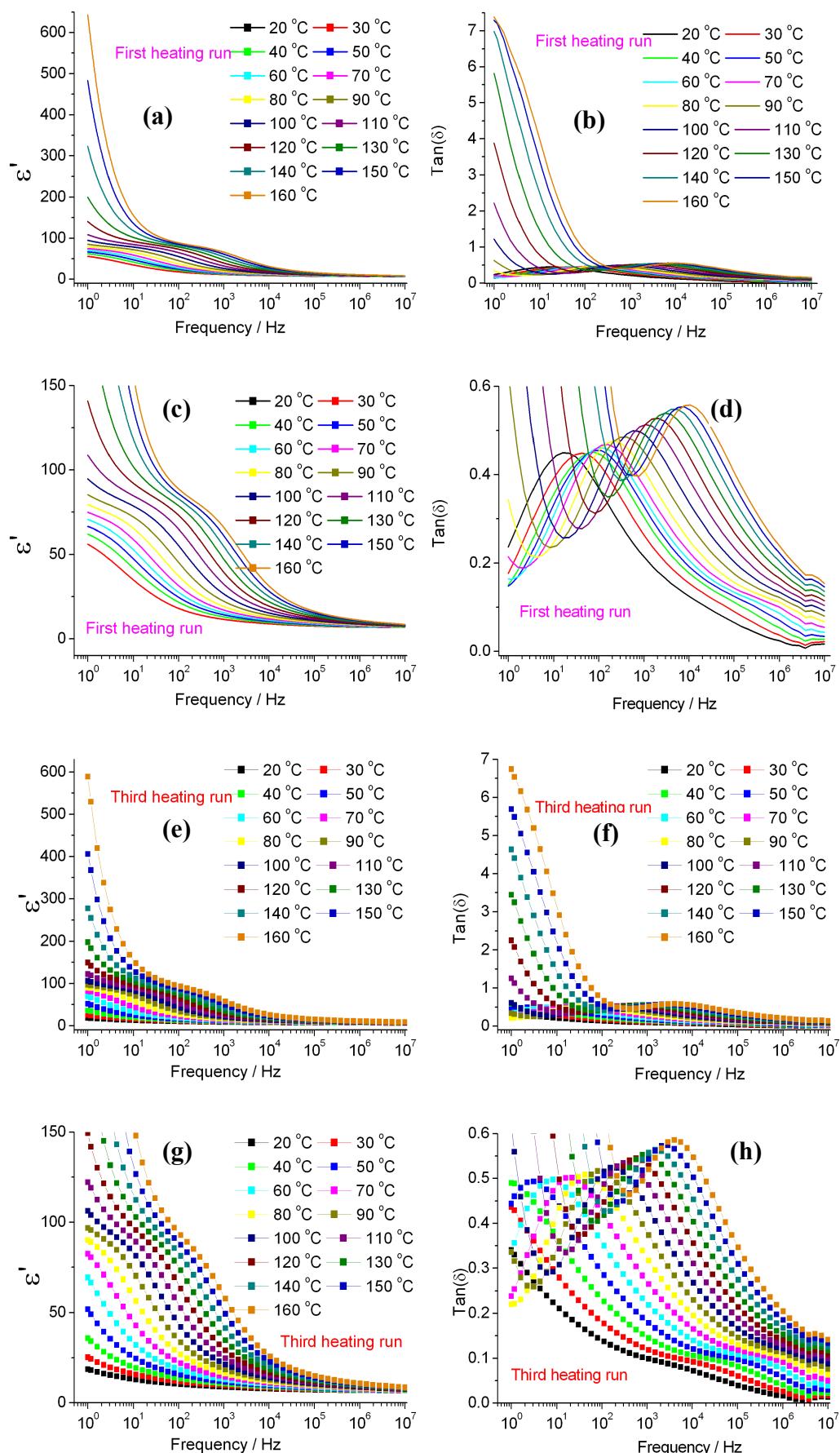
**Figure S3** Cole-Cole plots at 20–110 °C in (a) the first heating run (b) the second heating run for 2-APy-K.



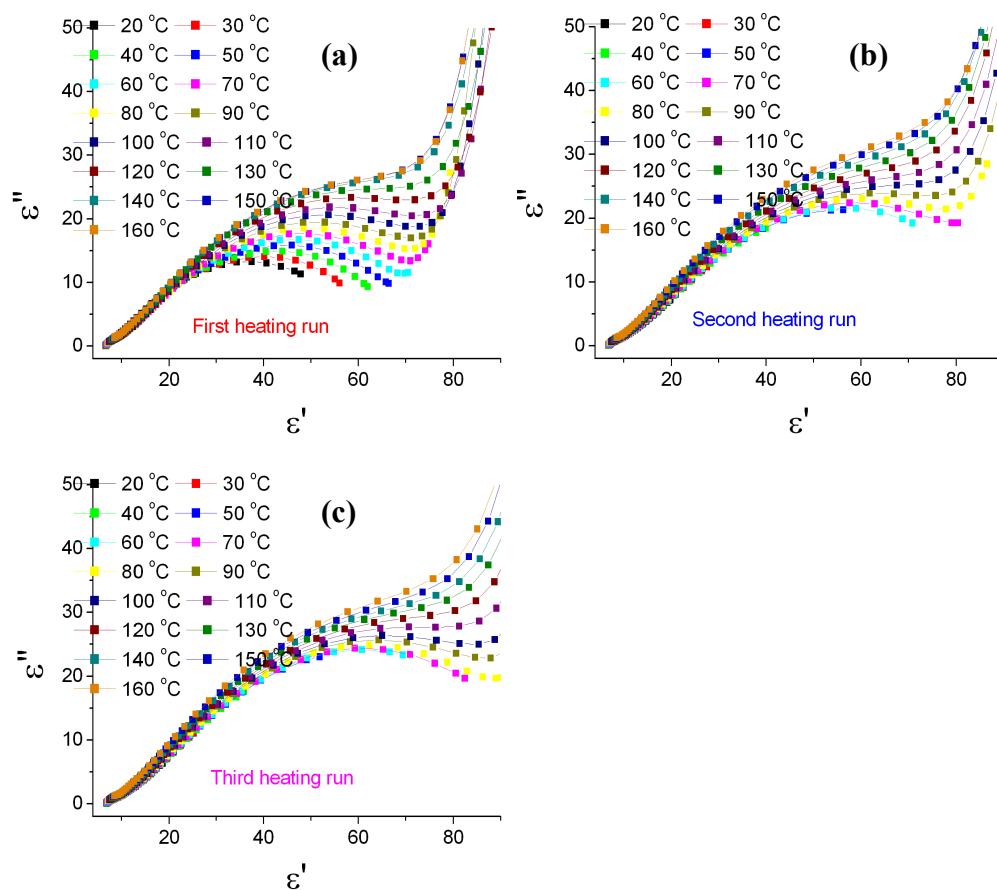
**Figure S4** Frequency dependences of  $\epsilon'$  and  $\tan(\delta)$  in the temperature range of 20–110 °C for **3-APy-K** (a, b) the first heating cycle (c, d) the second heating cycle. The results in two sequential heating cycles are comparable to each other.



**Figure S5** Cole-Cole plots at 20–110 °C in (a) the first heating run (b) the second heating run for **3-APy-K**.



**Figure S6** Frequency dependences of  $\epsilon'$  and  $\tan(\delta)$  in the temperature range of 20–160 °C for **4-APy-K** (a-d) the first heating cycle (e-h) the third heating cycle. The results in two heating cycles are comparable to each other as well as the second heating run.



**Figure S7** Cole-Cole plots at 20–160 °C in (a) the first heating run (b) the second heating run and (c) the third heating run for **4-APy-K**.