Supplementary Information

**MgAl LDH nanosheets loaded with Ni nanoparticles: a multifunctional filler for improving energy storage performance of PVDF-based nanocomposites**

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**Fig. S1** TEM image of the MgAl LDH nanosheets.

The thickness of MgAl LDH nanosheets is about 15 nm, as shown in Fig. S1.

**Fig. S2** EDS of the Ni-MgAl LDH nanoparticles.

The EDS spectrum was used to analyze the elemental distribution, as exhibited in Fig. S2, the presence of C, O, Mg, Al and Ni elements was observed.
To verify the chemical composition and electronic states of the Ni-MgAl LDH nanoparticles, XPS measurements were carried out. As shown in Fig. S3 (a), there are obvious peaks at 73 eV, 284 eV, 532 eV and 1300 eV, which represent Al 2p, C 1s, O 1s and Mg 1s, respectively. In addition, a weak peak at around 853 eV appeared in the XPS results of Ni-MgAl LDH nanoparticles, which was attributed to Ni 2p. The high resolution XPS spectrum of the Ni 2p was shown in Fig. S3 (a), and the peaks of Ni 2p\(_{3/2}\) and Ni 2p\(_{1/2}\) can be observed at 853 eV and 870 eV, respectively. This result confirmed that Ni nanoparticles were in the metal state instead of oxide in Ni-MgAl LDH nanoparticles\(^1\).

Fig. S4 XRD patterns of the pure PVDF, MgAl LDH/PVDF and Ni-MgAl LDH/PVDF nanocomposite films.
Fig. S5 XRD patterns of the pure PVDF, MgAl LDH/PVDF and Ni-MgAl LDH/PVDF nanocomposite films.

Table S1 Calculation results of melting enthalpy and crystallinity

| Sample           | Melting enthalpy $\Delta H_f$ (J g$^{-1}$) | Crystallinity (%) |
|------------------|------------------------------------------|-------------------|
| pure PVDF        | -42.38                                   | 40.5              |
| MgAl LDH/PVDF    | -43.20                                   | 41.3              |
| Ni-MgAl LDH/PVDF | -44.66                                   | 42.7              |

Fig. S6 (a) Frequency dependent dielectric constant and (b) dielectric loss of pure PVDF and MgAl LDH/PVDF nanocomposites with varied MgAl LDH contents.
Finite element simulations of the nanocomposites

We used COMSOL Multiphysics 5.4 with MATLAB to simulate the distribution of space charge density of the nanocomposites. The selected physical field was electric current field, and the research type was steady state. In the simulation, the MgAl LDH nanosheets were 200 nm in diameter and 15 nm in thickness, and the diameter of Ni nanoparticles were 2 nm. The fillers were randomly distributed in a three-dimensional block with side length 1 μm at a volume ratio of 0.4 vol.%, and the randomness of the filler position was realized by MATLAB code. The dielectric constants of the MgAl LDH nanosheets, Ni nanoparticles, and PVDF were 40, 1, and 9.2, respectively. The conductivities of the MgAl LDH nanosheets, Ni nanoparticles, and PVDF were $10^{-5}$ S m$^{-1}$, $1.43 \times 10^{7}$ S m$^{-1}$ and $10^{-9}$ S m$^{-1}$, respectively. An electric field of 500 kV mm$^{-1}$ was applied.

References

[1] H. Nesbitt, D. Legrand and G. Bancroft, Physics and Chemistry of Minerals, 2000, 27, 357-366.