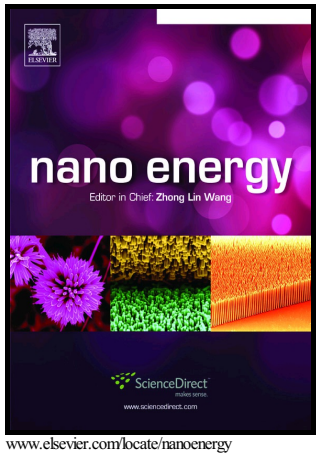

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Abstract

Three-dimensional (3D) hierarchical heterostructures have been widely studied for energy storage because of their amazing synergistic effect. However, a detailed characterization how the branched structure affects the backbone structure during electrochemical cycling, and the specific relationship between the backbone and the branched heterogeneous structure (namely synergistic effect) have been rarely revealed. In addition, the controllable synthesis of this system still remains a great challenge. Herein, we developed a one-step gradient hydrothermal method to obtain a series of 3D hierarchical heterogeneous nanostructures, including V_2O_5/NaV_6O_{15}, V_2O_5/ZnV_2O_6 and V_2O_5/CoV_2O_6, through controlling the sequence of nucleation and growth processes of different structural units in the same precursor. On the basis of time-resolved *in situ* X-ray diffraction (XRD) characterizations, we clearly elucidated the synergistic effect between the branched and backbone structure. During the synergistic effect, the branched NaV_6O_{15} helps to reduce the potential barrier during lithium-ion insertion/extraction, buffers the impact of crystal-system transformations during the charge/discharge process; the backbone V_2O_5 is beneficial to increase the charge/discharge capacity, inhibits the self-aggregation of branched NaV_6O_{15} and maintains the stability of 3D structure. Consequently, 3D V_2O_5/NaV_6O_{15} hierarchical heterogeneous microspheres exhibit the best electrochemical performance than pure

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