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Nature-inspired delamination of

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Abstract: Inspired by the natural frost weathering of rocks and minerals, layered double hydroxides (LDHs) were delaminated by a facile temperature shock method, without the usual involvement of organic solvents and sonication. The temperature shock was carried out with cycles of freezing the LDH solution in liquid nitrogen and subsequent melting the frozen solution in a 80 °C water bath. Around 67 % LDHs were delaminated into nanosheets after 16 such cycles. Moreover, the LDH monolayers thus obtained retain their lateral size at least three times better than that prepared in formamide under sonication. The present work provides a green and general approach to obtain high quality LDH nanosheets, which are finding application across a wide spectrum of areas.

Keywords: Layered double hydroxides; Multilayer structure; Exfoliation; Nanoparticles; Temperature shock; Nature inspiration.

1. Introduction

In many applications, layered double hydroxides (LDHs) [1] in the form of delaminated nanosheets are desirable, e.g. as a basic building blocks for constructing functional nanocomposites. However, the delamination or exfoliation of LDHs proves to be challenging, because of their unusually strong electrostatic interlayer interactions[2, 3]. While there were rare cases of delamination of LDHs in water[4], LDHs were usually delaminated in pure or mixed organic solvents, e.g. formamide[5, 6], isopropanol [7], acrylates[8], toluene[9], dimethylformamide[10], *etc.* Moreover, the intercalation of extraneous anions[11] and/or sonication[12] were a necessity to achieve LDH delamination in most cases. However, the resultant solvent leakage and organic waste are hazardous to the health of people and the environment. Appropriate extraneous anions for intercalation may be hard to choose. Sonication may

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