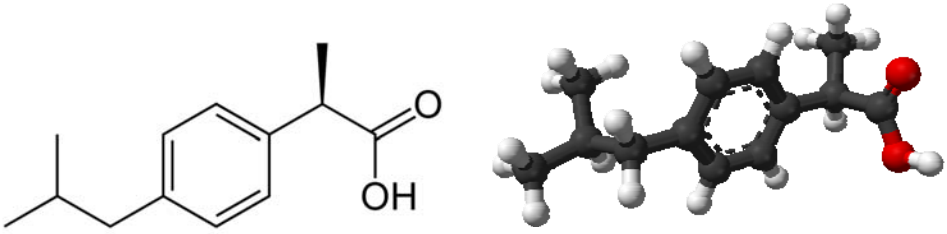
Electronic Supplementary Information

A.C.S. Alcântara et al. (Bionanocomposites based on alginate-zein.……)

Ibuprofen formula

(α-methyl-4-(2-methylpropyl) benzene-acetic acid)



The $[Mg_2Al]Cl$ LDH was characterized by means of XRD to verify the formation of the searched phase, and by EDX to confirm that the Mg/Al ratio is approximately 2:1, i.e. the theoretical elemental ratio. Considering that the XRD reflections can be indexed to a hexagonal lattice with rhombohedral symmetry, commonly used for description of LDH structures (A. Vaccari, *Catal. Today,* 1998, **41**, 53-71) this observed first diffraction peak can be ascribed to the (003) reflection, which gives a basal spacing (d_{003}) of about 0.77 nm characteristic of the $[Mg_2Al]Cl$ LDH (Z. Gu, A.C. Thomas, Z.P. Xu, J.H. Campbell and G.Q. Lu, *Chem. Mater.*, 2008, **20**, 3715-3722; Z.P. Xu, N.D. Kurniawan, P.F. Bartlett, G.Q. Lu. *Chem. Eur. J.*, 2007, **13**, 2824- 2830)

Structural information on the arrangement of the ibuprofen anions in the interlayer region can be derived from the XRD results and so, assuming a thickness of 0.48 nm for the LDH layer a basal spacing increase of 1.77 nm can be calculated, which is in the range reported by other authors (P. Gunawan and R. Xu, *J. Pharm. Sci*., **97**, 2008, 4367–4378). The ibuprofen molecule has a length of about 1.0 nm, estimated with HyperChem^{TM} Release 3 molecular modelling program, which is coherent with the literature (M. Vallet-Regi, A. Rámila, R.P. Del Real, J. Pérez-Pariente. *Chem. Mater.*, 2001, **13**, 503-510). In this way, it can be proposed that the intercalated ibuprofenate anions are arranged in a bi-layer with the negative charge of the drug molecule in close interaction with the positive charged sites of the layers and the molecular plane slightly tilted with respect to the LDH layers.

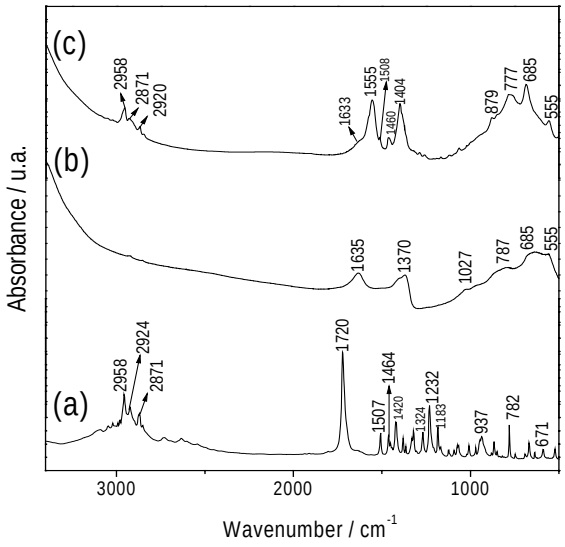


Fig. S1. FTIR spectra (4000-500 cm^{-1} region) of (a) ibuprofen, (b) $[Mg_2Al]Cl$ LDH and (c) LDH-IBU

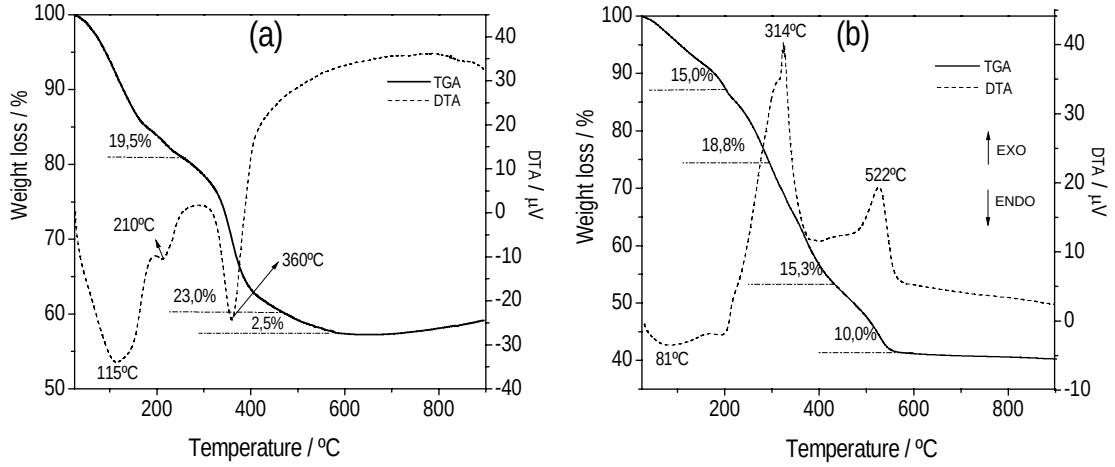


Fig. S2. TG and DTA curves obtained for (a) $[Mg_2Al]Cl$ LDH material and (b) LDH-IBU hybrid.

The TG curve of the pristine $[Mg_2Al]Cl$ LDH (Fig. S2a) shows a mass loss with an endothermic peak at 115ºC, attributed to the loss of physically adsorbed water molecules located at the exterior of the LDH particles and in the interlayer space (19.6% between 25 and 295ºC). A second mass loss with also an endothermic effect centred at

360ºC is related to a partial dehydroxylation of the brucite- type layers and release of intercalated chloride anions (23.0% between 295 and 450ºC). The last process involves the complete dehydroxylation of the material to form the respective oxides $MgAlO_4$ y MgO (2.5%, temperatures above 500ºC). The product obtained from the $[Mg_2Al]Cl$ – LDH, based on the data from the thermal analysis, is consistent with the expected formula $[Mg_{0.67}Al_{0.33}(OH)_2]Cl_{0.33}\cdot 1.08\ H_2O]$.

The thermal stability of ibuprofen is slightly increased in the LDH-IBU hybrid (Fig. S2b) in comparison to pure ibuprofen which is known to completely decompose in a single combustion process at about 220ºC. (C. R. Gordijo, C.A.S. Barbosa, A.M.C. Ferreira, V. R.L. Constantino and D.O. Silva, *J. Pharm. Sci.*, 2005, **94**, 1135-1148). Moreover, thermal decomposition of the organic molecule is a complex process that overlaps with other effects ascribed to the hydroxide matrix. In this way, the first weight loss (about 15%) up to 200ºC is ascribed to water elimination. Between that temperature and 350ºC there is an important weight loss (18.8%) accompanied by a strong exothermic peak associated to the elimination of the intercalated ibuprofen molecules. The second weight loss (15.3%) between 350 and 450ºC can be related to removal of water from the layers dehydroxylation and to a possible release of interlayer chloride anions. (C. R. Gordijo, C.A.S. Barbosa, A.M.C. Ferreira, V. R.L. Constantino, and D.O. Silva, *J. Pharm. Sci.*, 2005, **94**, 1135-1148). Finally, between 450 and 550ºC, a third weight loss of about 10.0% is observed, which can be related to decomposition of species derived from ibuprofen still present in the solid according with other authors (C. R. Gordijo, C.A.S. Barbosa, A.M.C. Ferreira, V. R.L. Constantino and D.O. Silva, *J. Pharm. Sci.*, 2005, **94**, 1135-1148)