

SUPPLEMENTARY INFORMATION

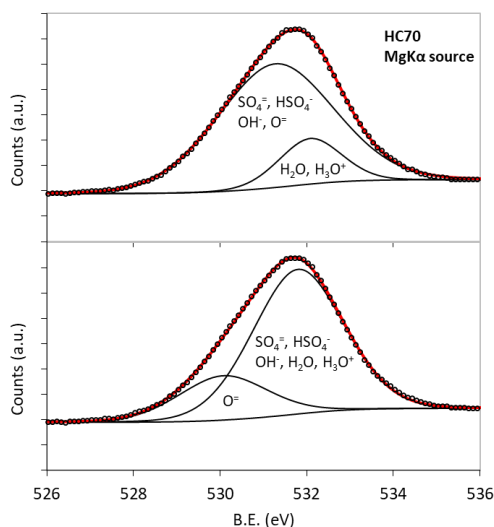


Figure S1. Two ways of performing the deconvolution of the O1s region for the HC70 sample. Symbols: experimental curve; red lines: deconvolution results. Tentative species for each peak indicated in the figure.

The region was subjected to deconvolution in two different ways (top and bottom plots in Figure S1). Both deconvolutions fit the experimental curve to perfection and, according to standard assignments that are usually provided within a rather elastic B.E. range in the scientific literature, the chemical correctness of both proposals is assured. For quantification purposes, it is however imperative to combine the deconvolution procedure with the fulfilment of mass and charge balances generated in accordance with the results of parallel techniques (e.g. FTIR), in order to select among the different deconvolution options. In other words, the purpose of the deconvolution procedure must be to minimize the sum of the fitting error and the mass and charge balance errors. For instance, the first peak at 530.1 eV in the bottom plot of Figure S1, which is commonly assigned to $O^=$ in Fe_2O_3 or $FeOOH$, (Grosvenor et al. 2004; Liu et al. 2014) has significant area. When the value of this is translated into actual $O^=$ concentration, and other constrains for the second peak are assumed (O/S atomic ratio in sulfates, presence of OH^- and H_2O/H_3O^+ , etc.), then a negatively charged surface results. When the unlikely presence of bisulfates, whose oxygen atoms are known to show the same binding energies as those of sulfates in both the O1s and S2p regions (Audi and Sherwood 2000) is assumed, the negative charge of the surface is partly alleviated, but in no case the charge balance is satisfied. If the peak at 530.1 eV is partly assigned to interstitial oxygen, as is occasionally the case, (Fernández-Pérez and Marbán 2020) the amount of this species would need to surpass that of sulfate for achieving a neutrally charged surface. In fact, only a deconvolution procedure similar to that presented in the top plot in Figure S1 permits to satisfy the charge balances for all the samples, as is shown below, without the need of including unlikely species.

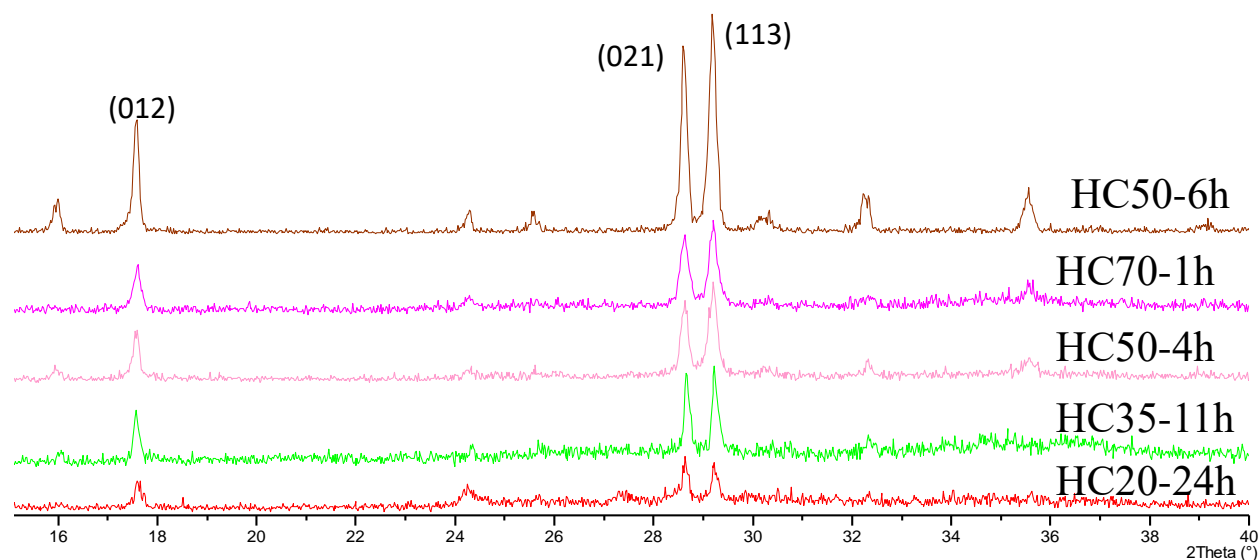


Figure S2: X-ray diffraction patterns of the solid obtained with increasing temperature (from 20 to 70°C) at times (24, 11, 4 and 1 h) when the transformation rate is considered to be 95%. A representative XRD pattern of a crystalline solid (HC50-6h) has been also included in this figure. As can be observed, the main reflections of natrojarosite are sharp and more intense when the transformation has been accomplished (HC50-6h). Moreover, minor reflections are clearly identified at the 2Theta interval in the crystalline phase.