Supporting information

LTA zeolite characterization based on pore type distribution

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S1 – Cation location proposed by literature force fields

To investigate the effects of other cation positions we used the force fields proposed by Fang et al., 2013[1] and by Martin-Calvo et al., 2014[2]. We extracted samples of the LTA framework during CO_2 adsorption at a pressure of 40 kPa at 303 K[1] and 293 K[2]. This strategy is based on the study reported by Vujic and Lyubartsev, 2016.[3] The authors demonstrated that the position of the cations does not vary much along the adsorption isotherm. To achieve such conclusion, they simulated CO_2 isotherms with the structure already optimized, with mobile cations and then with fixed cations, obtaining very similar results.

The force field proposed by Fang et al. (2013)[1] – CCFF – was developed from extensive quantum chemistry calculations. The great advantage of this force field is that it was developed specifically for the adsorption of CO_2 in LTA and successfully transferred, without reparameterizations, to zeolites NaY and NaX[1], in addition to reproducing adsorption isotherms at different temperatures[4] and Si/Al ratio[5].

The force field of Martin-Calvo et al. (2014)[2] was reparametrized from parameters developed by Garcia-Sanchez et al. (2009)[6], which was validated for FAU, MFI and MOR. The reparameterization consisted basically of increasing the absolute value of the sodium cation charges from 0.383 to 0.6633.

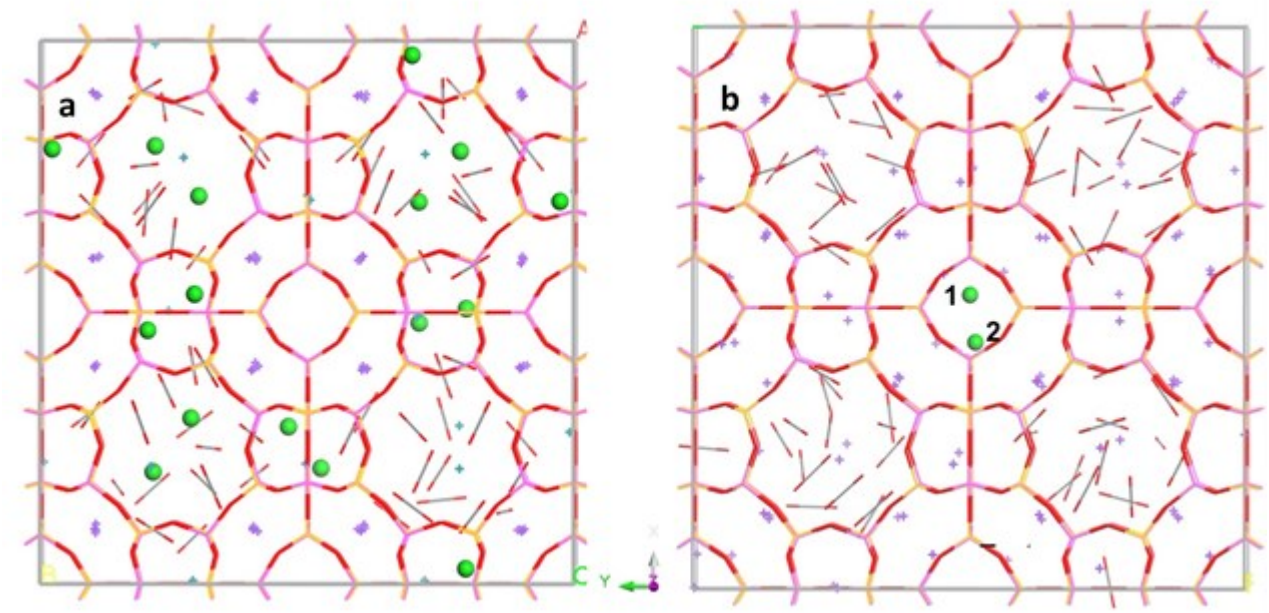

Figure S1 – (a) Positions of the Na^+ cations from the S3 site that migrated to the S2 site as predicted by the force field of Fang et al., 2013[1]. The green colored spheres show the position of the eight sodium cations in the 8-membered windows. When moving from site S3 to site S2, the cations occupy diagonal positions in the same 8-membered window. (b) Positions of the Na^+ cations from the S3 site that migrated into the sodalite as predicted by the force field of Martin-Calvo et al., 2014[2]. The green colored spheres show examples of the off-center (1) and anomalous (2) position of some of the cations in the vicinity of the 4-membered windows within the sodalite cavities. Part of the cations remained in the S3 positions inside the supercages.

S2 – Comparison between simulated values of isotherms obtained by different force fields

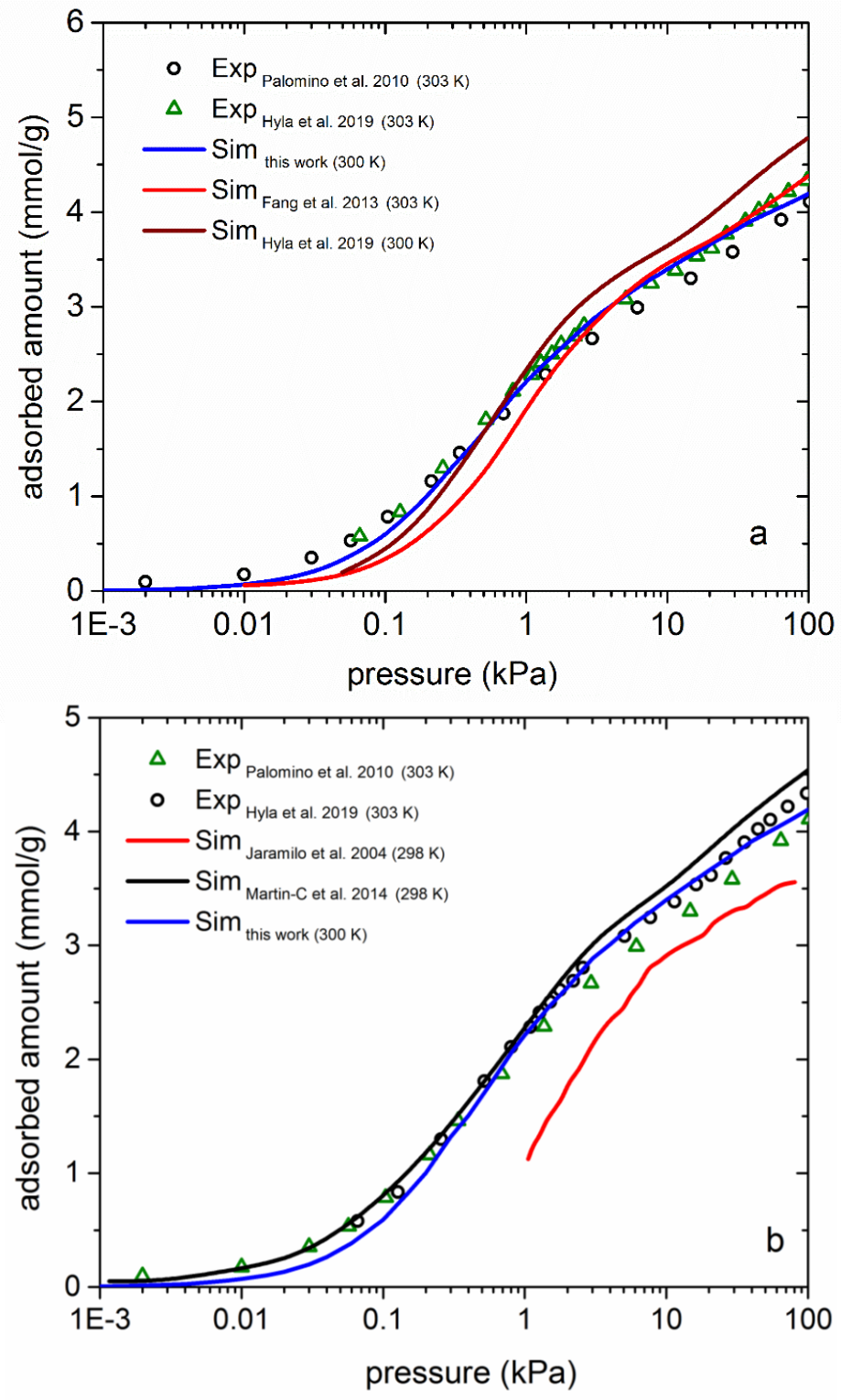

Figure S2 – (a) Experimental (Palomino et al., 2010; Hyla et al., 2019)[4,7] and simulated (Fang et al., 2013; Hyla et al. 2019; This work)[1,4] isotherms of CO_2 at 300 K and 303 K on zeolite Na-LTA. (b) Experimental (Palomino et al., 2010; Hyla et al., 2019)[4,7] and simulated (Jaramillo et al. 2004; Martin-Calvo et al. 2014; This work)[2,8] isotherms of CO_2 at 300 K and 303 K on zeolite Na-LTA.

S3 – Force field transferability to different temperatures

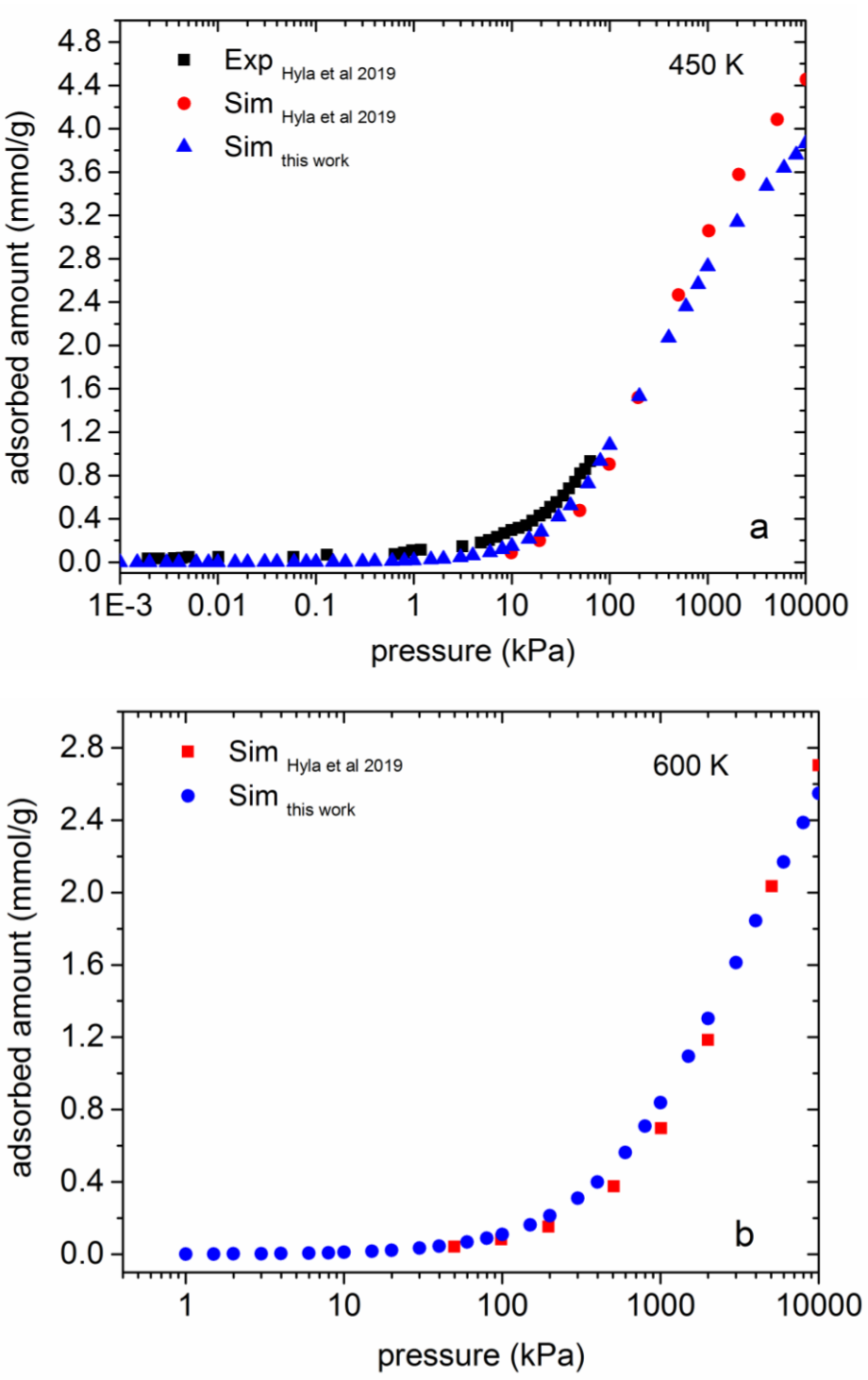

Figure S3 – (a) Experimental (Hyla et al., 2019)[4] and simulated (Hyla et al., 2019; this work)[4] isotherms of CO_2 at 450 K and (b) Simulated (Hyla et al. 2019; this work)[4] isotherms of CO_2 at 600 K on zeolite Na-LTA.

S4 – Comparison between simulated values of adsorption heat obtained by different force fields.

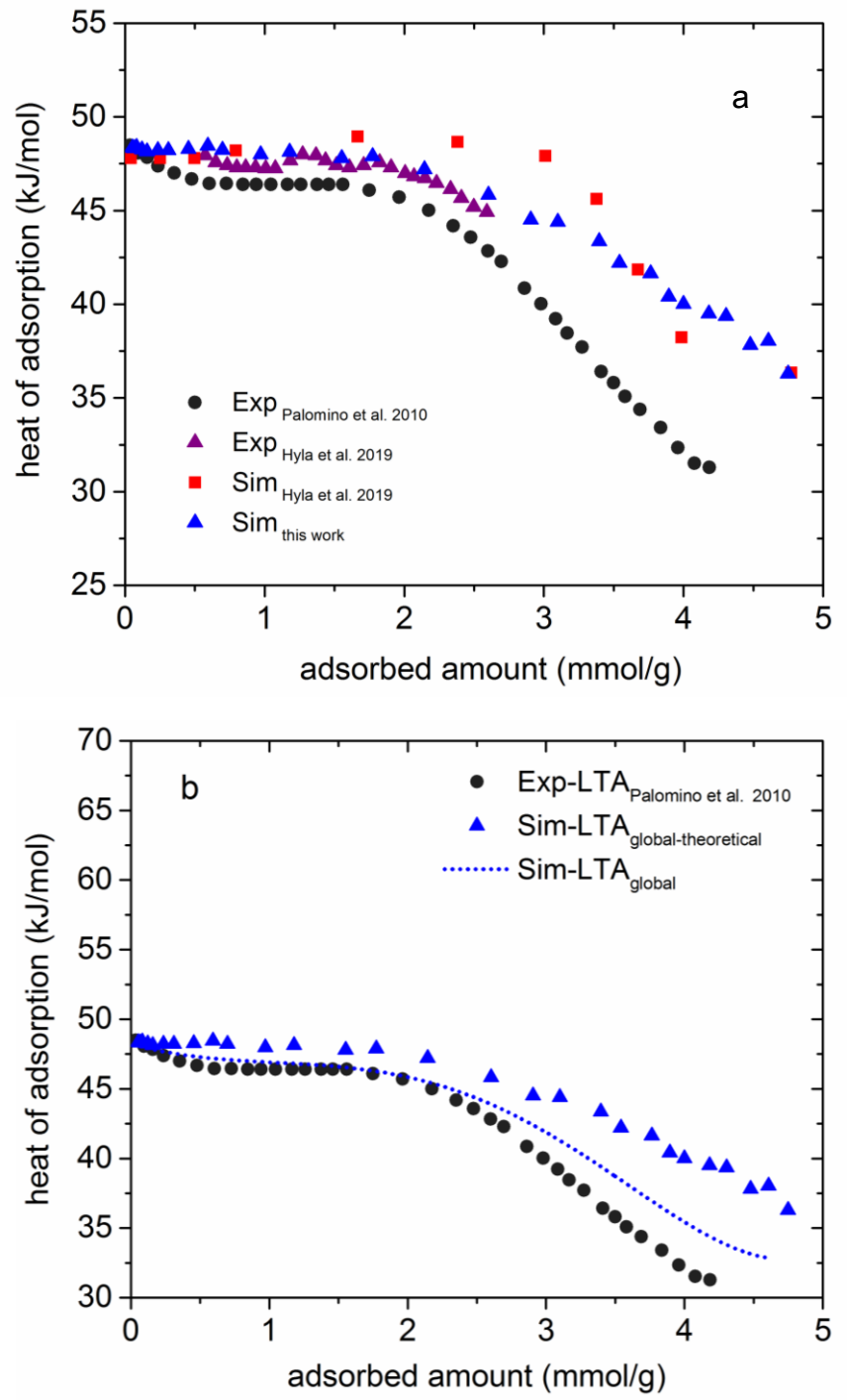

Figure S4 – (a) Experimental (Hyla et al. 2019; Palomino et al., 2010)[4,7] and simulated heat of adsorption (Hyla et al., 2009; this work)[4] of CO_2 at 300 K on zeolite Na-LTA. (b) New heat of adsorption (dotted line) predicted with the simulated global isotherm (LTA_{global}) for the sample LTA_{Prime1}.
The experimental data were obtained by microcalorimetry (Hyla et al. 2019)[4] and by fitting the Clausius-Clapeyron equation (Palomino et al., 2010)[7].

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