**Ab Initio** investigation of the interaction of \( \text{H}_2 \) with lithium exchanged low-silica chabazites.

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**Abstract.** The interaction of molecular hydrogen with the polarizing centers of lithium-exchanged chabazites with low Si/Al ratio (CHA-5/1 and CHA-3/1) was theoretically studied within a periodic approach at the B3LYP level of theory. The cation site preferences in both zeolites were determined and the \( \text{H}_2 \) interaction was then studied by adding a molecule in the proximity of the different polarizing centers. The energetic features of the \( \text{H}_2\text{-Li}_2\text{CHA-5/1} \) complex were also refined by using a cluster model cut out from the periodic structure in which the contribution of the dispersive forces to the interaction (absent at the B3LYP level of theory) were estimated at the MP2 level by means of an ONIOM2-like approach. Overall results show that the position of the polarizing center in the structure and its stability in the site is crucial for determining the sorption capacity of the zeolite. Lithium exchanged low-silica zeolites are shown to be only effective for hydrogen storage when the Al loading is at least 3:1. However, the highest predicted enthalpy of adsorption is around 10 kJ/mol and this indicates that they are far from fulfilling the conditions for practical applications.

1. **Introduction**

During the last years, intensive research have been dedicated to use hydrogen as an energy carrier.[1–3] However, a safe and easy to handle media for hydrogen storage is a primary requirement to link efficiently production and end use.[4] Among the proposed methods for hydrogen storage, molecular adsorption in microporous materials has been proposed as a viable solution[5–8] because the simplicity and the moderate temperature and pressure conditions of the process. For the role of host material in this process, Al-substituted chabazites[9] have been suggested as promising candidates. They belong to the category of aluminosilicate zeolites and, thus, are characterized by two main aspects i.e. (i) the Si/Al ratio in the framework; and (ii) the presence of extraframework cations necessary to compensate the effect of the substitution of silicon by aluminum atoms.

In a previous work, a theoretical study of the hydrogen interaction with the protons of variable Si/Al ratio acidic chabazites was reported with the objective to determine the potential of these materials as media for molecular hydrogen storage.[10] B3LYP computed binding energies were very low, being in the range: 1.8/2.4 kJ/mol, and even the inclusion of electron correlation at the MP2 level yielded small values around \( \sim \)5.0 kJ/mol. This weak interaction suggested that the...
presence of more polarizing sites in the framework might improve the hydrogen storage capacity. Hydrogen adsorption in Li-, Na- and K-exchanged high-silica zeolites was then investigated.\cite{11} In both cluster and periodic calculations, it was observed that the presence of cations slightly increases the sorption capacity of the zeolites, in particular for the LiCHA-11/1 case. This fact was also confirmed in recent publications on the theoretical investigation of hydrogen adsorption in cation exchanged mordenite\cite{12} and ferrierite\cite{13,14}. We also studied the interaction of hydrogen with a Mg-exchanged chabazite \cite{15} showing that a stronger polarizing center further increases the interaction, in agreement with experimental findings.\cite{16} Computed results from all of those studies have recently been gathered and discussed in a short review.\cite{17}

In this work, we extend upon previous studies by considering the role played by a higher Al loading (i.e. Si/Al: 5/1 and 3/1) on hydrogen adsorption and consequently by the presence of more than one polarizing center in the zeolitic framework. Only Li$^+$ cations are considered as charge balancers since results obtained for LiCHA-11/1 zeolite \cite{11} showed that lithium has a higher affinity with hydrogen in comparison with sodium and potassium.

2. Computational details
All the periodic calculations were performed with the program CRYSTAL06\cite{18}. The B3LYP Hamiltonian\cite{19} was adopted together with an all-electron Gaussian type basis set of double-zeta quality, hereafter referred as BSA. It consists in 31G(p), 88-31G(d), 8-411G(d) and 88-31G(d) contractions for H, Si, O and Al, respectively. The exponents (in Bohr$^{-2}$ units) of the most diffuse shell are: $\zeta_s$(H) = 0.16, $\zeta_{sp}$(Si) = 0.19, $\zeta_{sp}$(O) = 0.18 and $\zeta_{sp}$(Al) = 0.14; and the exponents for the polarization functions are: $\zeta_p$(H) = 1.1, $\zeta_d$(Si) = 0.61, $\zeta_d$(O) = 0.60 and $\zeta_d$(Al) = 0.51. For the Li$^+$ cations, a 61G contraction was adopted as used in a previous study on the cation selectivity in alkali exchanged high-silica chabazites.\cite{20} As regards the numerical integration of the exchange-correlation term, a pruned grid of 75 radial points and a maximum of 434 angular points in a Lebedev scheme was used. The condition for the SCF convergence was set to $10^{-8}$ Hartree. For the sampling of the irreducible Brillouin zone, the shrinking factor of the reciprocal space mesh was set to 2, which corresponds to 8 k-points. For the zeolite alone, lattice parameters and atomic positions were fully relaxed. Default convergence criteria were used for the geometry optimization.\cite{18,21}

In order to study the interaction between H$_2$ and the zeolites, one molecule per unit cell was inserted in the zeolitic cavity close to the different cation sites. By using a further optimization stage the most stable position of the hydrogen molecule inside the cavity was obtained. Because the molecule has a very small effect on the geometry of the host material, only the position of the hydrogen atoms and the counterions were fully relaxed, keeping fixed the rest of the framework. On the resulting geometry, the binding energies were computed as:

$$BE_{[H_2-MCHA]} = E_{[MCHA]} + E_{[H_2]} - E_{[H_2-MCHA]}$$

All reported binding energies were corrected for the Basis Set Superposition Error (BSSE) by using the Boys-Bernardi counterpoise method.\cite{22}

The anharmonic H-H stretching was computed by considering the H-H bond as an independent oscillator. The total potential energy was calculated and a polynomial curve of sixth degree used to fit the energy points. The first vibrational transition $\omega_{01}$ was obtained by solving numerically the one-dimensional Schrödinger equation.\cite{23}

Since present DFT functionals cannot cope with dispersive interactions\cite{24,25} a more appropriate scheme was tested to better describe the interaction of H$_2$ with the Li$_3$CHA-5/1 zeolite using a ONIOM2-like approach. Details are discussed in previous works\cite{10,11} and here we only report the ONIOM2 definition of the binding energy $BE_{\text{ONIOM2}}$:

$$BE_{\text{ONIOM2}} = BE_{[\text{High,Model}]} + BE_{[\text{Low,Real}]} - BE_{[\text{Low,Model}]}$$
where \( BE_{\text{High,Model}} \), \( BE_{\text{Low,Real}} \) and \( BE_{\text{Low,Model}} \) are the binding energies computed, with equation (1), on the model system at the HIGH level of theory, and on both the real and model systems at the LOW level of theory, respectively. For the LOW level of theory the B3LYP/BSA has been adopted, while both B3LYP/aug-cc-pVDZ and MP2 combined with both aug-cc-pVDZ and aug-cc-pVTZ basis sets were adopted as the HIGH level of theory. MP2 calculations were carried out by using the GAUSSIAN03 software package.[26]

As proposed by Otero-Areán et al.[13] to arrive at a final estimate of the adsorption enthalpy at 100 K (\( \Delta H_{100}^{o} \)), the zero-point energy correction and thermal effects were calculated within the harmonic approximation for the cluster adopted as model system at the B3LYP/BSA level of theory. The resulting value was added to the computed \( BE^{\text{ONIOM2}} \) to get an estimate of the \( \Delta H_{100}^{o} \).

3. Results and discussion

3.1. Structural models and relative stability of \( \text{Li}_2\text{CHA-5/1} \)

The structure of CHA-5/1 zeolite was constructed using as starting geometry the primitive cell of the pure silica chabazite[27] (\( R\bar{3}m \) space group). It consists in a hexagonal prism with 12 equivalent \( \text{SiO}_4 \) groups in which two Si atoms were substituted with aluminum to obtain the desired Si/Al ratio 5/1. The distribution of two Al atoms among the twelve available sites was previously investigated for acidic chabazites.[10] Here, we adopt the most stable arrangement that corresponds to two Al atoms positioned at opposite vertices of the hexagonal prism (see Figure 1). The resulting negatively charged framework was neutralized by adding lithium cations as counterions. Recently, Smith et al.[28] determined that in Li-exchanged low-silica chabazites there are three cationic sites corresponding to framework rings of different size, namely: four-, six- and eight-membered rings. Hereafter, they will be referred to as SI, SII and SIII, respectively. SI and SII were experimentally determined as the most favourable sites, and according to that finding, two \( \text{Li}_2\text{CHA-5/1} \) zeolitic models were constructed. The first distribution, denoted as DI, envisages both \( \text{Li}^+ \) cations in the SII sites (i.e. the two parallel six-member rings that compose the unit cell), whereas in the second distribution, named DII, both SI and SII sites are occupied. Both distributions are shown in Figure 1. At the B3LYP/BSA level, the DI distribution results to be more stable than DII by about 80 kJ/mol. This is clearly due to the semi-ionic nature of the zeolitic framework that allows Li to be more favourably accommodated in the SI site than the SII one.

3.2. Interaction of molecular hydrogen with the periodic models of \( \text{Li}_2\text{CHA-5/1} \)

The interaction of \( \text{H}_2 \) with \( \text{Li}_2\text{CHA} \) was studied by adding a molecule in the cavity of the host material close to the polarizing centers. In spite of the different stability, both DI and DII distribution were considered to analyse the role of the cation location on the dihydrogen adsorption. A preference of the hydrogen molecule to form side-on complexes was observed. Relevant structural features of the complexes are reported in Table 1. The distances between the molecule’s center of masses and the counterions at the SII sites are larger than 2.4 Å while for the SI site it decreases to 2.14 Å as an indication of the better sorption capacity of the lithium cation when located in the four-member ring. Indeed, from Table 1, it can be seen that the highest binding energy (4.2 kJ/mol) is predicted for that site. This finding also agrees with previous theoretical studies [11–14] in which it was shown that exposed cations tend to interact strongly with dihydrogen. On the contrary, binding at the SII site is lower because of the shielding effect of the neighboring atoms in the zeolitic framework, in agreement with calculations by Torres et al.[11].

The H–H anharmonic frequency shifts due to interactions with the cationic sites are also reported in Table 1. The bathochromic shifts are within a range of 30–50 cm\(^{-1}\), and as already
Table 1. Relative stability of the adopted cation distributions ($\Delta E$ - kJ/mol), optimal distance from the Li$^+$ cations to the molecule’s center of masses ($d_{M-H_2}$ - Å), BSSE corrected binding energies (BE$_c$ - kJ/mol), and changes in the H-H bond distance ($\Delta d_{HH}$ - Å) and in the H-H stretching frequency ($\Delta \omega_{01}$ - cm$^{-1}$) for H$_2$ adsorbed on different cation sites.

| Cationic Distribution | $\Delta E$ | Cationic Site | $d_{M-H_2}$ | BE$^a$ | $\Delta d_{HH}$ | $\Delta \omega_{01}$ | $\Delta \omega_{01}$ |
|-----------------------|-----------|--------------|-------------|--------|----------------|----------------|---------------|
| Distribution I        | 0.0       | SII          | 2.41        | 0.4 (2.5) | 0.002         | -31            |               |
|                       |           | SII          | 2.47        | 0.3 (2.4) | 0.001         | -34            |               |
| Distribution II       | 80.0      | SI           | 2.14        | 4.2 (9.2) | 0.001         | -50            |               |
|                       |           | SII          | 2.54        | 0.2 (1.4) | 0.002         | -31            |               |

$^a$ Values in parentheses are the BE computed by single-point calculations with an aug-cc-pVDZ basis set on H$_2$. $^b$ The differences referred to the corresponding values for molecular hydrogen in the gas phase: $d_{H-H}^{ref} = 0.7438$ Å and $\omega_{01}^{ref} = 4238$ cm$^{-1}$.

discussed in our previous work,[10] there is no correlation between the computed binding energies and the changes in the frequency. For instance, values around -30 cm$^{-1}$ were computed with a corresponding negligible binding energy values, so they might be associated to a pure effect of the electrostatic field on the very sensitive hydrogen molecule.

3.3. Interaction of molecular hydrogen with a cluster model of Li$_2$CHA-5/1

The high binding energy value computed in SI site for the H$_2$–Li$_2$CHA-5/1 complex is an interesting result; therefore, as a further refinement, a careful analysis of the effect of the Li
cation environment and of the dispersion forces (not fully accounted for by the B3LYP level of theory) was performed through complementary calculations carried out by means of a cluster approach. To that purpose, a cluster model was cut out from the optimal B3LYP/BSA periodic structure identifying the fragment of the zeolite in which the molecular adsorption takes place. The adopted cluster model is shown in Figure 2. The nearest all silica four-member ring to lithium was also considered as part of the cluster. To include dispersive contributions, MP2 single point calculations were carried out. Errors in the results are not expected to be large because B3LYP provides geometries close to those obtained at MP2 level.[30]

Table 2. BSSE corrected binding energies (in kJ/mol) of the cluster model of the H$_2$–Li$_2$CHA-5/1 system at the different levels of theory required for the evaluation of the BE$^{\text{ONIOM2}}$. Low level: B3LYP/BSA.

| High level       | BE$^{\text{[Low,Real]}}$ | BE$^{\text{[Low,Model]}}$ | BE$^{\text{[High,Model]}}$ | BE$^{\text{ONIOM2}}$ | $\Delta H_{100}^o$ $^a$ |
|------------------|--------------------------|---------------------------|---------------------------|-----------------------|--------------------|
| B3LYP/aug-cc-pVDZ| 4.2                      | 3.4                       | 9.3                       | 10.1                  | -5.7               |
| MP2/aug-cc-pVDZ  | 4.2                      | 3.4                       | 11.8                      | 12.6                  | -8.2               |
| MP2/aug-cc-pVTZ  | 4.2                      | 3.4                       | 11.9                      | 12.7                  | -8.3               |

$^a$ Obtained from BE$^{\text{ONIOM2}}$ by including the zero-point energy and thermal effects at 100 K.

Within the adopted periodic ONIOM2 approach, the MP2 contribution to the binding energy was computed with the different basis sets (see Computational Details section for definitions). Results are reported in Table 2. The MP2/aug-cc-pVDZ binding energy for the cluster model is 11.8 kJ/mol. It is interesting to compare it with the B3LYP one of 9.3 kJ/mol which indicates that the contribution of dispersive interactions included at the MP2 level is of the order of 2.5 kJ/mol (i.e. around 25% of the B3LYP binding energy). With the aug-cc-pVTZ, the MP2 binding energy slightly increases thus showing that the convergence with the basis set is reached at this level. It is worthy to note that the difference between BE$^{\text{[Low,Model]}}$ (i.e. cluster model) and BE$^{\text{[Low,Real]}}$ (i.e. periodic model) due to long range effects accounts for less than 1.0 kJ/mol. Although this contribution seems rather small, it represents almost $\sim$20% of the total BE$^{\text{[Low,Model]}}$ binding energy.

Values computed at the High level were also employed to get BE$^{\text{ONIOM2}}$ (see Table 2 and eq. (2)). The BE$^{\text{ONIOM2}}$ value of 12.7 kJ/mol, computed at the MP2/aug-cc-pVTZ high level, is remarkably higher than the value of 8.8 kJ/mol reported previously for H$_2$–LiCHA-11/1 [11] where Li$^+$ is located in the six-membered ring. This confirms the importance of the cation location for the hydrogen sorption capacity of alkali-exchanged zeolites.

In order to get a more thermodynamically sound value for the interaction energy, the zero-point energy and thermal effects at 100K were also considered. As an estimate, the B3LYP/BSA level correction of 4.4 kJ/mol computed for the same cluster model of Figure 2 was used. The predicted enthalpies of adsorption ($\Delta H_{100}^o$) are also reported in Table 2. The final estimate of -8.3 kJ/mol obtained for the adsorption of H$_2$ on the SI site in Li$_2$CHA-5/1 is in good agreement with available experimental $\Delta H_{100}^o$ values from FTIR spectroscopic measurements reported by Otero-Areán and co-workers [9, 13, 14] on H$_2$–Li-FER and H$_2$–Li-ZSM-5 (i.e. -4.1 and -6.5 kJ/mol, respectively). The slight overestimation of $\Delta H_{100}^o$ can be due to the small size of the adopted cluster model.

3.4. Structural models and relative stability of Li$_3$CHA-3/1
The high binding energy predicted for hydrogen at the SI site in the DII distribution is a promising result. Unfortunately, the large difference in energy stability with respect to the DI one makes this prediction useless. However, at variance with the Li$_2$CHA-5/1, it is expected that
with a higher aluminum loading (i.e. Si/Al=3:1) a stable zeolitic framework can be obtained with lithium located in one of the SI sites; thus we considered Li$_3$CHA-3/1 zeolite in our investigation. The structural models were constructed from the all-silica chabazite by substituting three Si atoms with aluminum. The most stable Al distribution in the hexagonal prism for this Si/Al ratio was reported in Ref. [10] and adopted in the present work (see Figure 3). Firstly, the two SII sites of Li$_3$CHA-3/1 were occupied by lithium atoms and then four different cationic distributions (i.e DI, DII, DIII and DIV) were defined by varying the position of the third Li$^+$ cation among the available four-member rings of the cell. Of those distributions, DII envisages the Li$^+$ cation coordinated by two neighbouring oxygen atoms at the edge between two four-membered rings and, in accordance with the notation adopted in the present work, it closely resembles a SIII site. After full relaxation of the structure, the relative stability of the four cationic distribution follows the trend DIII > DIV > DII > DI with energy differences of 6.6, 15.9 and 37.0 kJ/mol with respect to DIII. As for Li$_2$CHA-5/1, the relative stability of the cation sites is mainly ruled by electrostatics with the Li ion being more stable in the four-membered rings containing two Al atoms (i.e. more ionic) and then in those with only one Al atom. It is worth noting that distribution DII with the SIII site occupied is more stable than the SI site in the DI distribution.

![Figure 3. Schematic representation of the cation sites in pure lithium-exchanged CHA-3/1 structure. The lithium atoms fixed in the more stable SII sites are represented by the light-blue spheres, whereas the dark-blue ones represent the more exposed Li$^+$ cation. On the other hand, the zeolitic framework is represented by gray (silicon) and yellow (aluminum) colours. DI, DII, DIII and DIV cationic distributions obtained by varying the position of the more exposed Li$^+$ among the available four-membered rings are shown.](image)

3.5. Interaction of molecular hydrogen with the periodic model of Li$_3$CHA-3/1

The B3LYP/BSA binding energies of H$_2$ in interaction with Li$_3$CHA-3/1 are summarized in Table 3. For the DI case, the hydrogen adsorption was also investigated for the Li$^+$ cation located at the SII site. In agreement with the results obtained for the H$_2$–Li$_2$CHA-5/1 system, very small binding energies were computed, whereas, as expected, H$_2$ interacts more strongly with the Li$^+$ cation at the SI site. For the latter, a binding energy of 4.2 kJ/mol was obtained very close to the reported value for Li$_2$CHA-5/1 (see DII distribution in Table 1). For the DIII and DIV distributions, binding energies of 2.9 and 3.6 kJ/mol were obtained, respectively. Such lower values are due to the presence of two aluminum atoms in the four-member rings that makes the neighbouring oxygen atoms more ionic and more prone to coordinate the lithium ion. The highest computed binding energy was obtained for the DII distribution in which lithium is well exposed, located at the corner of an eight-membered ring and then readily accessible to the H$_2$ molecule. Accordingly, the shortest distance between the molecule’s center of masses and the Li$^+$ cation is observed for that case. The computed value of 1.99 Å is quite similar to the distance computed at the B3LYP level on an adduct of dihydrogen with bare Li$^+$[29] and indicates that the cation in this site can be regarded as almost free. Furthermore, data in Table 3 show an unusually large anharmonic H-H frequency shift of -193 cm$^{-1}$, in contrast with the smaller shifts of -48 and -31 cm$^{-1}$ obtained for the DIII and DIV distributions, respectively.
The latter values are in the range of frequency shifts usually observed in our investigations so far.

Table 3. B3LYP/BSA relative stability of the adopted cation distributions (\(\Delta E - \text{kJ/mol}\)), optimal distance from the cations to the molecule’s center of masses (\(d_{M-H_2} - \text{Å}\)), BSSE corrected binding energies (\(\text{BE}_c - \text{kJ/mol}\), and changes in the H-H bond distance (\(\Delta d_{HH} - \text{Å}\)) and in the H-H stretching frequency (\(\Delta \omega_{01} - \text{cm}^{-1}\)) for H\(_2\) adsorbed on the cationic sites of the different cationic distributions of Li\(_3\)CHA-3/1 zeolite.

| Cationic Distribution | \(\Delta E\) | Cationic Site | \(d_{M-H_2}\) | \(\text{BE}_c^a\) | \(\Delta d_{HH}^b\) | \(\Delta \omega_{01}^b\) |
|-----------------------|-------------|--------------|---------------|----------------|----------------|----------------|
| Distribution I        | 37.2        | SI           | 2.17          | 4.2 (8.6)     | 0.002          | -69            |
|                       |             | SII          | 2.45          | 0.3 (1.9)     | 0.001          | -30            |
|                       |             | SII          | 2.52          | 0.0           | 0.001          | -              |
| Distribution II       | 15.9        | SIII         | 1.99          | 6.2 (13.5)    | 0.007          | -193           |
| Distribution III      | 0.0         | SI           | 2.20          | 2.9 (6.6)     | 0.002          | -48            |
| Distribution IV       | 6.6         | SI           | 2.20          | 3.6 (7.5)     | 0.003          | -31            |

\(a\) Values in parentheses are the BE computed by single-point calculations with an aug-cc-pVDZ basis set on H\(_2\). \(b\) The differences referred to the corresponding values for molecular hydrogen in the gas phase: \(d_{H-H}^{ref} = 0.7438 \text{Å}\) and \(\omega_{01}^{ref} = 4238 \text{cm}^{-1}\).

For the H\(_2\)–Li\(_3\)CHA-3/1 complex, a trustworthy cluster model (i.e. neutral and without strong dipole moments) could not be cut out from the periodic structure; therefore, the contribution of the dispersive forces to the interaction by means of the ONIOM2-approach was not directly computed in this case. To arrive at a better estimate of the binding energy, a different approach was adopted. A single-point calculation was carried out at the B3LYP level by using an aug-cc-pVDZ basis set on H\(_2\) (see values in parentheses in Table 3). As a check, similar calculations were done on H\(_2\)–Li\(_2\)CHA-5/1 with hydrogen interacting with Li at the different sites and are also reported in Table 1. A binding energy of 9.2 kJ/mol was computed for the SI site in very good agreement with the B3LYP/aug-cc-pVDZ high level calculation reported for the adopted cluster model in Table 2. This confirms the goodness of this simple approach. For H\(_2\)–Li\(_3\)CHA-3/1, binding energies increase significantly with a rather high value for the DII distribution. Finally, by considering that an increment of roughly 25% in the binding energy is obtained for H\(_2\)–Li\(_2\)CHA-5/1 at the MP2/aug-cc-pVTZ level of theory (see Table 2), values of 10.7, 16.8, 8.2 and 9.4 kJ/mol were estimated for the DI, DII, DIII and DIV distributions, respectively. The estimated value of 16.8 kJ/mol for the DII distribution show that, even including thermal correction to 100K, the enthalpy of adsorption could be larger that 10 kJ/mol.

4. Conclusions
In the present work, the interaction of a hydrogen molecule with the polarizing sites of Li\(_2\)CHA-5/1 and Li\(_3\)CHA-3/1 zeolites was theoretically studied at the B3LYP level of theory. The periodic models were derived from experimental available data on low-silica alkali exchanged chabazites\[28\]. In addition, for the H\(_2\)–Li\(_2\)CHA-5/1 system with the highest computed binding energy a further analysis was carried out by combining the periodic results with MP2 calculations on a cluster model cut out from the zeolitic framework.

Present results allow us to draw some conclusions on the role played by the Al content of the zeolite on the interaction with molecular hydrogen. In agreement with experimental findings\[31\], computed data show that the adsorption capacity increases with increasing the Al content of the zeolite. However, the overall effect is more related to the availability of exposed cation than
to the role played by the increased ionicity of the zeolitic framework. Indeed, binding energies increase when H$_2$ interacts with Li$^+$ at site SI, SII and SIII, respectively. On the contrary, the polarizing capacity of the cation reduces as much as the cation is effectively coordinated to the zeolitic framework. This can be highlighted by considering that the binding energy of hydrogen with the SII site, where Li is located in a six-memebered ring, is small and tends to decrease when passing from a Si/Al ratio of 11:1 to 5:1 and 3:1 with the BE estimated at the B3LYP/aug-cc-pVDZ level being 3.1[11], 2.5, 1.9, respectively. Furthermore, our results show that most stable cation distribution of the Li$_2$CHA-5/1 cannot be used to store H$_2$ and the Al loading must be increased at least as in the Li$_3$CHA-3/1 chabazite or more. Therefore, even though low-silica zeolites should be more effective as possible hydrogen storage material, for lithium exchanged chabazites, the binding energies are still too far from the required range between -20 and -40 kJ/mol necessary for technological applications[32].

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