Synthesis of Germanosilicate Zeolite HPM-12 Using a Short Imidazolium-based Dication:
Structure-Direction by Charge-to-Charge Distance Matching

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Fig. S1. XRD patterns of as-made products with Ge/ of 0.1 under different times.

Fig. S2. XRD patterns of as-made HPM-12 obtained with different Ge/ under a crystallization time of 5 days.

Fig. S3. XRD patterns of the products obtained at different dilution levels for Ge/ = 0.3 at 3 days crystallization.

Fig. S4. XRD patterns of as-made (top) and calcined HPM-12 (bottom) with Ge/ of 0.2.

Fig. S5. Detail of the 1H NMR spectra of the blank experiments in Table 3, showing the displacement and change in shape of the two protons directly attached to the imidazole ring (b,c). This AB system suffers a downfield shift of 0.37 ppm and converts into an almost perfect AZ system by a 20 time increase in concentration. The presence of HF also changes the shape of the multiplet and slightly displaces the signal (only for the highest concentration in HF). Apparently, the effect of pH (or chloride concentration) is smaller.

Fig. S6. TG curves of as-made HPM-12: sample a and b were synthesized with the same Ge/ of 0.3 but different H2O/TO2 of 10 and 15, respectively; sample c was synthesized with Ge/ of 0.5 and H2O/TO2 of 10.

Fig. S7. Conformations of 3BDMI cations in vacuo (two views, left and right, are shown). Relative energies are also given (in kcal/mol with respect to the most stable conformer), as calculated in vacuo.

Fig. S8. Void volume (blue and white surfaces) of the UOE framework; 3BDMI cations can locate along the 10-R channels (perpendicular to the paper) or along the ‘a’ axis, spanning two consecutive channels (white arrow).

Fig. S9. Location of 3BDMI dications (‘ss’ conformer) in [010] orientation (top, two views at left and right) or in [100] orientation (bottom, two views).

Table S1. 13C NMR chemical shifts (in ppm) of the dissolved zeolite and of blank solutions designed to show the dependencies on concentration and presence of other species.
Fig. S1. XRD patterns of as-made products with Ge\textsubscript{f} of 0.1 under different heating times.

Fig. S2. XRD patterns of as-made HPM-12 obtained with different Ge\textsubscript{f} under a heating time of 5 days.
Fig. S3. XRD patterns of the products obtained at different dilution levels for Ge\textsubscript{f} = 0.3 with a heating time of 3 days.

Fig. S4. XRD patterns of as-made (top) and calcined HPM-12 (bottom) synthesized with Ge\textsubscript{f} of 0.2.
Table S1. $^{13}$C NMR chemical shifts (in ppm) of the dissolved zeolite and of blank solutions designed to show the dependencies on concentration and presence of other species.

<table>
<thead>
<tr>
<th>Experiment</th>
<th>3BDMI&lt;sup&gt;a&lt;/sup&gt;</th>
<th>HF&lt;sup&gt;a&lt;/sup&gt;</th>
<th>SiO&lt;sub&gt;2&lt;/sub&gt;&lt;sup&gt;a&lt;/sup&gt;</th>
<th>a</th>
<th>b,c</th>
<th>f</th>
<th>d</th>
<th>e</th>
<th>g</th>
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<td>1</td>
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<td>123,121</td>
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<td>123,121</td>
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<sup>a</sup>Relative to the concentration in the dissolved zeolite NMR experiment, <sup>b</sup>HCl to achieve the same pH as in the zeolite NMR experiment, <sup>c</sup>Standard NMR experiments to check the purity of the SDA (concentration not determined and variable in different experiments)
Fig. S5. Detail of the $^1$H NMR spectra of the blank experiments in Table 3, showing the displacement and change in shape of the two protons directly attached to the imidazole ring (b,c). This AB system suffers a downward shift of 0.37 ppm and converts into an almost perfect AZ system by a 20 time increase in concentration. The presence of HF also changes the shape of the multiplet and slightly displaces the signal (only for the highest concentration in HF). Apparently, the effect of pH (or chloride concentration) is smaller.
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