

## Supporting Information

Effect of Al-Si-Al and Al-Si-Si-Al Pairs in the ZSM-5 Zeolite Framework on the  $^{27}\text{Al}$  NMR Spectra. A Combined High Resolution  $^{27}\text{Al}$  NMR and DFT/MM Study

Jiří Dědeček,<sup>†</sup> Stepan Sklenak,<sup>\*,†</sup> Chengbin Li,<sup>†</sup> Blanka Wichterlová,<sup>†</sup> Vendula Gábová,<sup>†</sup> Jiří Brus,<sup>‡</sup> Marek Sierka,<sup>§</sup> and Joachim Sauer<sup>§</sup>

*J. Heyrovský Institute of Physical Chemistry, Academy of Sciences of the Czech Republic, Dolejškova 3, 182 23 Prague, Czech Republic, Institute of Macromolecular Chemistry of the Academy of Sciences of the Czech Republic, Heyrovsky Sq. 2, 162 06 Prague 6, Czech Republic, and Institut für Chemie, Humboldt-Universität zu Berlin, Unter den Linden 6, 10099 Berlin, Germany*

\* Corresponding author. E-mail: [stepan.sklenak@jh-inst.cas.cz](mailto:stepan.sklenak@jh-inst.cas.cz)

<sup>†</sup> J. Heyrovsky Institute of Physical Chemistry.

<sup>‡</sup> Institute of Macromolecular Chemistry.

<sup>§</sup> Humboldt-Universität zu Berlin.

**TABLE S1: Al-O-Si angles (degrees) for Al-Si-Al pairs**

| Sequences                    | T-O-T angles |       |       |       | Average |
|------------------------------|--------------|-------|-------|-------|---------|
| <b>Al(1)</b> -Si(16)-Al(17)  | 136.3        | 144.8 | 153.7 | 160.7 | 148.9   |
| Al(1)-Si(16)- <b>Al(17)</b>  | 135.3        | 139.4 | 142.1 | 148.2 | 141.3   |
| <b>Al(1)</b> -Si(16)-Si(17)  | 141.7        | 141.8 | 143.4 | 146.1 | 143.3   |
| Si(1)-Si(16)- <b>Al(17)</b>  | 134.6        | 137.7 | 137.9 | 153.1 | 140.8   |
|                              |              |       |       |       |         |
| <b>Al(1)</b> -Si(16)-Al(15)  | 141.2        | 146.0 | 156.9 | 164.1 | 152.1   |
| Al(1)-Si(16)- <b>Al(15)</b>  | 142.2        | 142.6 | 148.9 | 152.9 | 146.7   |
| <b>Al(1)</b> -Si(16)-Si(15)  | 141.1        | 142.5 | 159.2 | 163.8 | 151.7   |
| Si(1)-Si(16)- <b>Al(15)</b>  | 134.9        | 142.6 | 168.2 | 174.0 | 154.9   |
|                              |              |       |       |       |         |
| <b>Al(8)</b> -Si(12)-Al(11)  | 138.9        | 141.7 | 153.0 | 161.2 | 148.7   |
| Al(8)-Si(12)- <b>Al(11)</b>  | 139.6        | 145.6 | 146.0 | 151.4 | 145.7   |
| <b>Al(8)</b> -Si(12)-Si(11)  | 141.3        | 147.2 | 151.1 | 159.4 | 149.8   |
| Si(8)-Si(12)- <b>Al(11)</b>  | 139.5        | 146.2 | 147.0 | 150.5 | 145.8   |
|                              |              |       |       |       |         |
| <b>Al(9)</b> -Si(10)-Al(11)  | 126.8        | 141.0 | 161.0 | 166.7 | 148.9   |
| Al(9)-Si(10)- <b>Al(11)</b>  | 144.5        | 144.7 | 149.9 | 155.0 | 148.5   |
| <b>Al(9)</b> -Si(10)-Si(11)  | 124.6        | 144.1 | 150.6 | 152.6 | 143.0   |
| Si(9)-Si(10)- <b>Al(11)</b>  | 137.1        | 142.6 | 150.5 | 150.8 | 145.3   |
|                              |              |       |       |       |         |
| <b>Al(8)</b> -Si(9)-Al(10)   | 143.0        | 144.7 | 149.0 | 159.9 | 149.2   |
| Al(8)-Si(9)- <b>Al(10)</b>   | 138.0        | 144.9 | 150.7 | 153.6 | 146.8   |
| <b>Al(8)</b> -Si(9)-Si(10)   | 143.0        | 148.2 | 151.1 | 158.8 | 150.3   |
| Si(8)-Si(9)- <b>Al(10)</b>   | 138.3        | 147.8 | 148.7 | 150.9 | 146.4   |
|                              |              |       |       |       |         |
| <b>Al(14)</b> -Si(20)-Al(24) | 131.3        | 136.5 | 141.4 | 157.7 | 141.7   |
| Al(14)-Si(20)- <b>Al(24)</b> | 133.4        | 135.4 | 139.6 | 154.6 | 140.8   |
| <b>Al(14)</b> -Si(20)-Si(24) | 132.3        | 139.4 | 146.0 | 155.2 | 143.2   |
| Si(14)-Si(20)- <b>Al(24)</b> | 131.2        | 136.7 | 141.6 | 148.1 | 139.4   |
|                              |              |       |       |       |         |
| <b>Al(21)</b> -Si(20)-Al(24) | 140.8        | 151.6 | 160.1 | 173.1 | 156.4   |
| Al(21)-Si(20)- <b>Al(24)</b> | 126.2        | 141.6 | 143.6 | 158.0 | 142.3   |
| <b>Al(21)</b> -Si(20)-Si(24) | 144.7        | 145.0 | 159.3 | 159.9 | 152.2   |
| Si(21)-Si(20)- <b>Al(24)</b> | 130.9        | 137.4 | 141.9 | 148.2 | 139.6   |
|                              |              |       |       |       |         |
| <b>Al(22)</b> -Si(23)-Al(24) | 136.9        | 137.0 | 138.6 | 148.9 | 140.3   |
| Al(22)-Si(23)- <b>Al(24)</b> | 140.4        | 141.6 | 142.4 | 153.5 | 144.5   |
| <b>Al(22)</b> -Si(23)-Si(24) | 137.2        | 139.7 | 142.9 | 145.5 | 141.3   |
| Si(22)-Si(23)- <b>Al(24)</b> | 133.2        | 137.1 | 140.0 | 152.2 | 140.6   |

**TABLE S2. Al-O-Si angles (degrees) for Al-Si-Si-Al pairs**

| Sequences                          | T-O-T |       |       |       | Average |
|------------------------------------|-------|-------|-------|-------|---------|
| <b>Al(2)</b> -Si(1)-Si(17)-Al(23)  | 143.6 | 146.1 | 150.0 | 158.5 | 149.6   |
| Al(2)-Si(1)-Si(17)- <b>Al(23)</b>  | 143.6 | 144.3 | 147.6 | 150.1 | 146.4   |
| <b>Al(2)</b> -Si(1)-Si(17)-Si(23)  | 138.4 | 143.3 | 145.7 | 145.8 | 143.3   |
| Si(2)-Si(1)-Si(17)- <b>Al(23)</b>  | 142.8 | 144.8 | 146.0 | 147.2 | 145.2   |
|                                    |       |       |       |       |         |
| <b>Al(8)</b> -Si(2)-Si(1)-Al(17)   | 137.9 | 142.9 | 147.7 | 157.4 | 146.5   |
| Al(8)-Si(2)-Si(1)- <b>Al(17)</b>   | 135.6 | 136.5 | 139.4 | 143.5 | 138.8   |
| <b>Al(8)</b> -Si(2)-Si(1)-Si(17)   | 138.5 | 156.3 | 158.9 | 159.2 | 153.2   |
| Si(8)-Si(2)-Si(1)- <b>Al(17)</b>   | 134.7 | 135.5 | 140.9 | 141.8 | 138.2   |
|                                    |       |       |       |       |         |
| <b>Al(17)</b> -Si(16)-Si(7)-Al(23) | 137.8 | 141.9 | 142.6 | 143.3 | 141.4   |
| Al(17)-Si(16)-Si(7)- <b>Al(23)</b> | 143.0 | 144.1 | 145.7 | 147.8 | 145.2   |
| <b>Al(17)</b> -Si(16)-Si(7)-Si(23) | 135.6 | 138.0 | 138.7 | 147.5 | 139.9   |
| Si(17)-Si(16)-Si(7)- <b>Al(23)</b> | 142.6 | 144.5 | 148.0 | 148.5 | 145.9   |
|                                    |       |       |       |       |         |
| <b>Al(7)</b> -Si(23)-Si(22)-Al(1)  | 133.6 | 140.7 | 141.8 | 153.2 | 142.3   |
| Al(7)-Si(23)-Si(22)- <b>Al(1)</b>  | 138.0 | 140.2 | 141.6 | 146.2 | 141.5   |
| <b>Al(7)</b> -Si(23)-Si(22)-Si(1)  | 135.4 | 141.1 | 144.4 | 152.4 | 143.3   |
| Si(7)-Si(23)-Si(22)- <b>Al(1)</b>  | 140.5 | 140.7 | 141.1 | 144.8 | 141.8   |
|                                    |       |       |       |       |         |
| <b>Al(16)</b> -Si(7)-Si(23)-Al(22) | 134.1 | 144.3 | 144.8 | 150.4 | 143.4   |
| Al(16)-Si(7)-Si(23)- <b>Al(22)</b> | 139.6 | 140.0 | 146.0 | 147.0 | 143.2   |
| <b>Al(16)</b> -Si(7)-Si(23)-Si(22) | 133.6 | 145.5 | 146.5 | 148.2 | 143.4   |
| Si(16)-Si(7)-Si(23)- <b>Al(22)</b> | 137.3 | 141.0 | 144.0 | 144.6 | 141.7   |

Reference 52 with the complete list of authors:

Frisch, M. J.; Trucks, G. W.; Schlegel, H. B.; Scuseria, G. E.; Robb, M. A.; Cheeseman, J. R.; Montgomery, J., J. A. ; Vreven, T.; Kudin, K. N.; Burant, J. C.; Millam, J. M.; Iyengar, S. S.; Tomasi, J.; Barone, V.; Mennucci, B.; Cossi, M.; Scalmani, G.; Rega, N.; Petersson, G. A.; Nakatsuji, H.; Hada, M.; Ehara, M.; Toyota, K.; Fukuda, R.; Hasegawa, J.; Ishida, M.; Nakajima, T.; Honda, Y.; Kitao, O.; Nakai, H.; Klene, M.; Li, X.; Knox, J. E.; Hratchian, H. P.; Cross, J. B.; Bakken, V.; Adamo, C.; Jaramillo, J.; Gomperts, R.; Stratmann, R. E.; Yazyev, O.; Austin, A. J.; Cammi, R.; Pomelli, C.; Ochterski, J. W.; Ayala, P. Y.; Morokuma, K.; Voth, G. A.; Salvador, P.; Dannenberg, J. J.; Zakrzewski, V. G.; Dapprich, S.; Daniels, A. D.; Strain, M. C.; Farkas, O.; Malick, D. K.; Rabuck, A. D.; Raghavachari, K.; Foresman, J. B.; Ortiz, J. V.; Cui, Q.; Baboul, A. G.; Clifford, S.; Cioslowski, J.; Stefanov, B. B.; Liu, G.; Liashenko, A.; Piskorz, P.; Komaromi, I.; Martin, R. L.; Fox, D. J.; Keith, T.; Al-Laham, M. A.; Peng, C. Y.; Nanayakkara, A.; Challacombe, M.; Gill, P. M. W.; Johnson, B.; Chen, W.; Wong, M. W.; Gonzalez, C.; Pople, J. A., *Gaussian 03*, Revision C.02, Gaussian, Inc., Wallingford CT, 2004.