

Hydrogen Bonds in Crystalline Imidazoles Studied by ^{15}N NMR and *ab initio* MO Calculations

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Intermolecular hydrogen bonds of the type N–H...N in crystals of imidazole and its 4-substituted and 4,5-disubstituted derivatives were studied by ^{15}N CP/MAS NMR and an *ab initio* molecular orbital (MO) calculation. In the ^{15}N CP/MAS NMR spectrum of each of the imidazole derivatives, two peaks due to the two different functional groups, >NH and =N–, were observed. The value of the ^{15}N isotropic chemical shift for each nitrogen atom depends on both the length of the intermolecular hydrogen bond and the kind of the substituent or substituents. It was found that the difference between the experimental chemical shifts of >NH and =N– varies predominantly with the hydrogen bond length but does not show any systematic dependence on the kind of substituent. The *ab initio* MO calculations suggest that the hydrogen bond formation influences the ^{15}N isotropic chemical shift predominantly, and that the difference between the ^{15}N isotropic chemical shift of >NH and =N– varies linearly with the hydrogen bond length.

Key words: ^{15}N CP/MAS NMR; Imidazoles; Hydrogen Bond; ^{15}N Chemical Shift; *ab initio* MO Calculation.