

EVIDENCE OF NITROGEN AS ACCEPTOR IN NITROMETHANE-FORMALDEHYDE HETERODIMERS CHARACTERIZED USING MATRIX ISOLATION INFRARED SPECTROSCOPY AND COMPUTATIONAL METHODS

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Formaldehyde (FA) is a fascinating molecule for astrochemists due to the complex mechanistic pathways leading to its formation. Being a very important prebiotic precursor, understanding its participation in various weak interactions is crucial. FA is well established to form hydrogen bonds. In the present work, the interaction of FA with Nitromethane (NM) was studied at low temperature and supported by *ab initio* theoretical calculations. The heterodimers of NM and FA, NM-FA, were generated within Ar and N₂ matrices and characterized using infrared spectroscopy. Perturbation in the ν_3 mode of NM and ν_2 mode of FA due to the formation of heterodimers has been investigated, as these infrared spectral signatures were shifted from the monomer absorption of NM and FA. The variation of the intensity of these features, in response to the variation in concentration of FA and NM, additionally supported with computations, affirms the formation NM-FA. The red shifts observed, agree well with the predictions by harmonic frequency calculations on the pnictogen-hydrogen-tetrel bound geometry. Computations indicated three minima on the potential energy surface at MP2/CBS and B2PLYP-GD3/CBS levels of theory. The most stable heterodimer, observed experimentally, was stabilized by cooperative pnictogen (O...N), hydrogen (O...H) and tetrel (O...C) bonds as confirmed by QTAIM and NBO analyses. Dominance of electrostatics over other effects in forming the bonds has been established by energy decomposition analysis (EDA). The ability of FA, as a potential electron donor to pnictogen bonding while being a weak tetrel donor too, in addition to its expected participation in hydrogen bonding, stands established experimentally and computationally.