

Supporting Information for Synthesis, Spectral Characterization, and Crystal Structures of Organophosphonic Diamides: Pyramidal Nitrogen Centers and Hydrogen Bonding in  $[\text{PhP}(\text{O})(\text{NH}^t\text{Bu})_2]$ ,  $[\text{PhP}(\text{O})(\text{NHDipp})_2]$  (Dipp = 2,6- $^i\text{Pr}_2\text{C}_6\text{H}_3$ ) and  $[\text{}^t\text{BuP}(\text{O})(\text{NH}^i\text{Pr})_2]$

Ramaswamy Murugavel\* and Ramasamy Pothiraja

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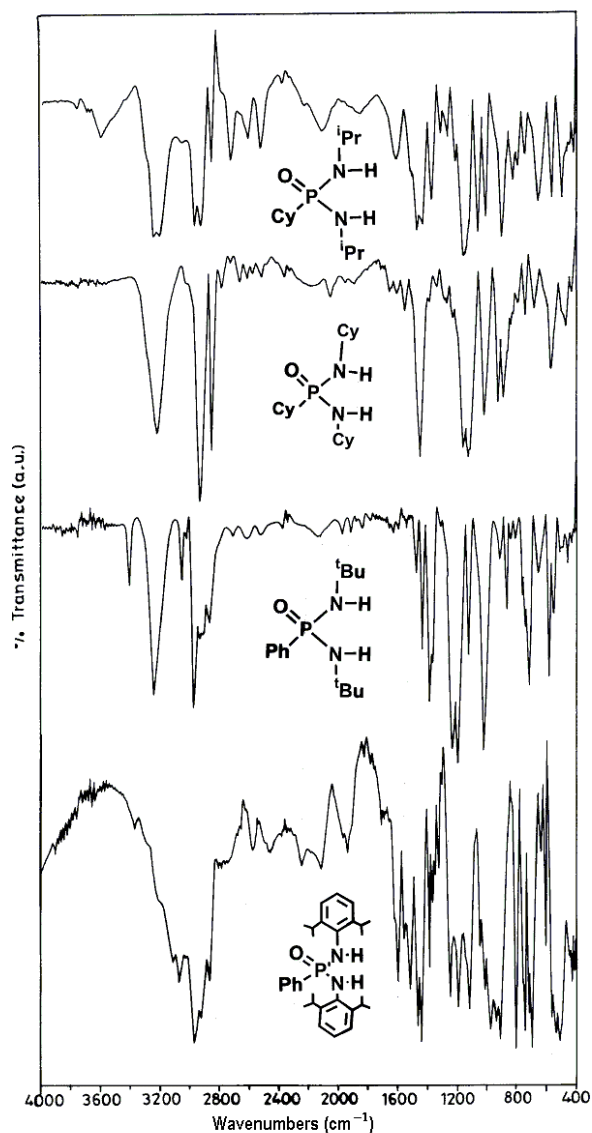


Figure S1. Representative IR spectra of phosphonic diamides (KBr discs)

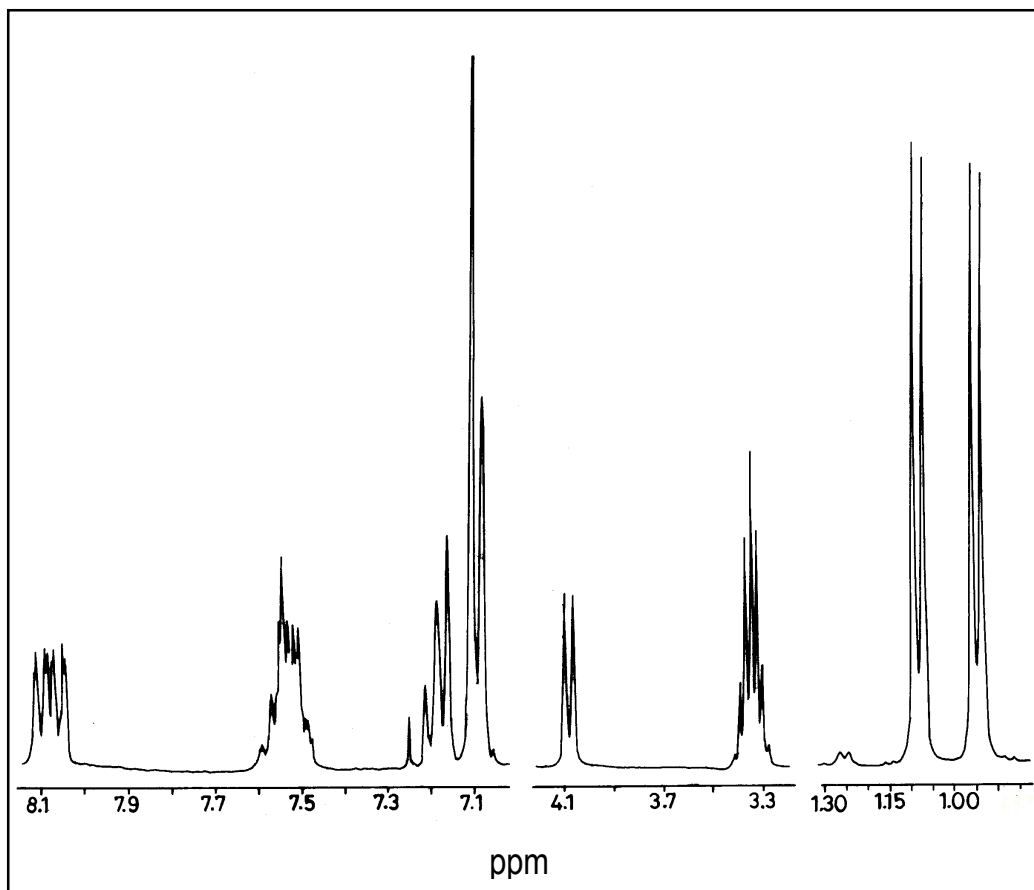


Figure S2. <sup>1</sup>H NMR spectrum of [PhP(O)(NHDipp)<sub>2</sub>] (**2**) in CDCl<sub>3</sub>.