Supplementary material (ESI) for Journal of Materials Chemistry

Supporting Information (containing description and 4 figures) for manuscript entitled "Enhanced Performance of Dye-Sensitized Solar Cells by an Al_2O_3 Charge-Recombination Barrier formed by Low-Temperature Atomic Layer Deposition"

The following supporting materials explain how the graphical model shown in Fig. 4 was constructed. The lattice constants i and ii in Fig. 4a are the reported values for anatase TiO_2.[14] The molecular sizes of TMA, O-Al$(OH)_2$, and O-Al(O-Al$(OH)_2)_2$ were computed using *Chem3D Ultra* (www.cambridgesoft.com), where the computation was based on energy-minimization using the *ab initio* method with STO-3G as the basis set. The molecules were treated in the model as spheres, whose radius was such that the whole of the molecule was encompassed, as illustrated in Fig. (i)-(iii). The molecular size of the N719 molecule was obtained from the reported crystallographic data of the N3 molecule.[14] Note that the N719 dye is identical to the N3 dye during the sensitization process, where the N719 dye is dissociated in acetonitrile into $N3^{2-}$ ions [while releasing $(C_4H_9)_4N^{2+}$ ions], which then sensitize the electrodes. The N719 molecule was also treated as a sphere encompassing the whole of the molecule, as shown in Fig. (iv).

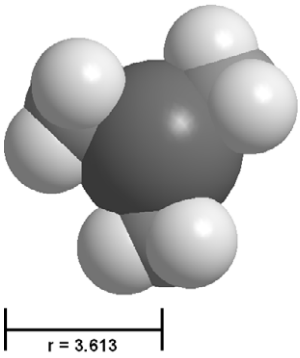

Fig. (i) The computed TMA molecule for the model shown in Fig. 4, where the length of 3.613 Å is the radius of the sphere representing the TMA molecule.

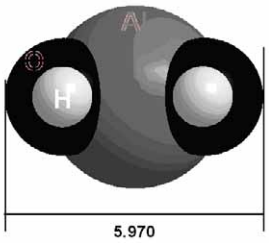

Fig. (ii) The computed O-Al$(OH)_2$ molecule for the model shown in Fig. 4, where the

length of 5.970 Å is the diameter of the sphere representing the O-Al$(OH)_2$ molecule.

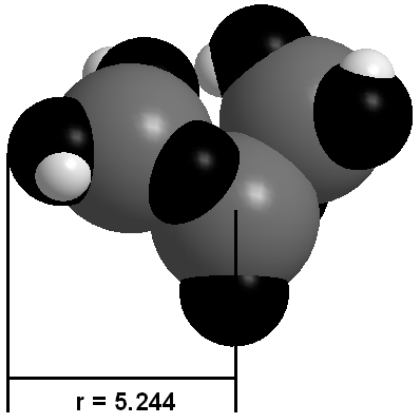

Fig. (iii) The computed O-Al(O-Al$(OH)_2)_2$ molecule for the model shown in Fig.4, where the length of 5.244 Å is the radius of the sphere representing the O-Al(O-Al$(OH)_2)_2$ molecule.

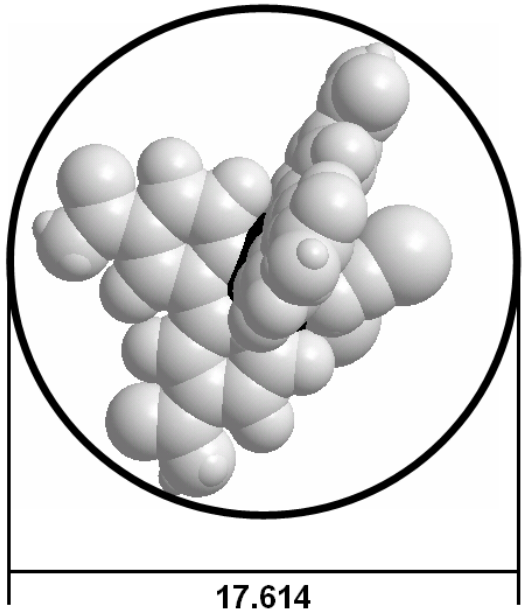

Fig. (iv) The $N3^{2-}$ ion, which is the functioning species of the N719 dye, treated in the model shown in Fig. 4, where the length of 17.614 Å is the diameter of the sphere representing the N719 molecule.