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Donor-acceptor organo-imido polyoxometalates: new materials for photonics and solar energy conversion

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Polyoxometalates (POMs) are a class of anionic, molecular metal oxide clusters, whose range of properties commensurate with their enormous variety of structural types. Derivatization of POMs with organic groups results in hybrids. For example, connecting a multi-electron accepting POM (such as the lacunary Keggin, $XW_{11}O_{39}^{n-}$, or Dawson, $X_2W_{17}O_{61}^{n-}$ cluster) with a light harvesting chromophore results in systems where photoexcitation results in multiple charge accumulation on the POM, and reduction of protons to dihydrogen. In most such examples, however, electronic isolation of the POM from the organic subunits means that these properties are an enhancement (due to enforced spatial proximity) of behaviour that occurs with unconnected POMs and chromophores. By contrast, we are interested in studying new optical and photophysical behaviours that emerge when strong electronic coupling occurs between POMs and appended organic groups. Such strong coupling is observed with the POM organoimido bond, and while the synthetic methods to obtain this class of POM derivatives are now quite well developed, there have so far been few efforts to address the properties that may emerge from this strong communication. Here, the author will discuss our work developing these materials as a new class of second and third order non-linear optical (NLO) chromophores building an understanding of their charge-transfer transitions and also exploiting them as co-adsorbents in p-type dye-sensitized solar cells (p-DSSC). This work shows that POM chromophores can break through empirical NLO performance limits that apply to most purely organic materials, but that the extent of charge transfer on to the POM is strongly influenced by the organic donor group and bridge. It is also showing that POMs can enhance p-DSSC photovoltages by as much as 100%.

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