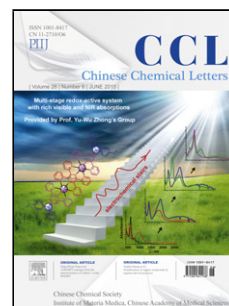


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Communication

Crystallization-induced phosphorescence, remarkable mechanochromism, and grinding enhanced emission of benzophenone-aromatic amine conjugates

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ABSTRACT

Pure organic luminogens with efficient room temperature phosphorescence (RTP) and remarkable mechanochromism are highly desired in view of their fundamental significance and technical applications. Herein, four twisted pure organic luminogens based on benzophenone and aromatic amines were synthesized and their photophysical properties were thoroughly investigated. They exhibit crystallization-induced phosphorescence (CIP), giving bright fluorescence and phosphorescence dual emission in crystals. Upon grinding, they become amorphous and emit predominantly red-shifted fluorescence, demonstrating remarkable mechanochromism. Furthermore, three of them even demonstrate greatly enhanced emission upon grinding, which is rarely observed in twisted D-A structured luminogens.

The intensive pursuit of materials with efficient room temperature phosphorescence (RTP) have been continuous these years for their essential applications in organic light-emitting diodes, data recording, security protection, photodynamic therapy [1-8], *etc.* Although great progress has been achieved in inorganic and organometallic phosphors [9-14], taking advantage of heavy atom effect to boost intersystem crossing (ISC), less attention has been paid to pure organic luminogens with relatively poor spin-orbital coupling (SOC).

Compared with inorganic and organometallic compounds, pure organic luminogens take advantages of facile preparation, easy functionalization, low toxicity, and low cost. In the past, the studies on pure organic phosphors, however, were normally carried out at cryogenic conditions owing to the susceptibility of triplet excitons to molecular motions and external quenching [15,16]. To obtain efficient pure organic RTP, on one hand, people normally endeavored to increase the SOC and thus facilitate the ISC process through incorporation of aromatic carbonyls, heavy atoms, and heteroatoms [15-20]; on the other hand, they tried to depress the nonradiative deactivation pathways through absorption on solid matrix [21], crystallization [22-27], embedding into rigid matrix [28], supramolecular interactions [29], and metal-organic framework coordination [30,31]. Among these approaches, the discovery of crystallization-induced phosphorescence (CIP) in pure organics such as benzophenone and 4,4'-dibromobiphenyl suggested a possible crystal engineering way to efficient RTP [17].

Meanwhile, efficient luminogens with high contrast mechanochromism also attracted great attention because of their fundamental implications on the molecular packing and promising applications in optical recording, mechanical sensors, security papers [32-39], *etc.* CIP-active pure organic RTP luminogens are also promising mechanochromic candidates due to the susceptibility of triplets to molecular motions and external quenchers [22,27]. With continuous efforts to construct pure organics with efficient RTP and remarkable mechanochromism, in this work, a series of benzophenone-aromatic amine conjugates were synthesized (Fig. 1). Such molecular design based on the following considerations: (1) Aromatic carbonyl groups can effectively promote SOC and share rapid ISC rates [15,16,40]; (2) Nitrogen-containing diphenylamine (DPA) and triphenylamine (TPA) may enhance $n-\pi^*$ transition to facilitate ISC process [18-20]; (3) Such electron donating-accepting (D-A) structure may narrow the energy gap between excited singlet and triplet states, thus facilitating the ISC process; (4) Twisted D-A structure and possible RTP emission may synergistically endow the compounds with remarkable mechanochromism [41]. Herein, their detailed photophysical properties were investigated.

The target compounds were facilely prepared according to the synthetic routes shown in Scheme S1 (Supporting information) in moderate to high yields (62%~74%). They were characterized by spectroscopic methods, with satisfactory results obtained (Figs. S1-S7 in Supporting information). Particularly, single crystal structures definitely suggest the successfully preparation of these compounds [CCDC 1845357 (BPDPA), 1845358 (BPTPA), 1845359 (BP2DPA), and 1845360 (BP2TPA)]. When dissolved in *n*-hexane (Hex), toluene (Tol), trichloromethane (TCM), and tetrahydrofuran (THF), their absorption maximum are generally at about 356~382 nm (Table 1, Fig. S8 in Supporting information), with slightly varied peaks due to the poor tuning effect on ground states by solvent polarity.

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