



Review

Metal-containing crystalline luminescent thermochromic materials

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ABSTRACT

Stimuli-responsive functional materials with specific properties have been garnering recent attention. Various external stimulations, such as light, mechanical or pressure, vapor, solvent or temperature, have been extensively utilized to induce physical property changes. Temperature, the most fundamental parameter, is believed to serve as an efficient stimulus for triggering luminescence changes. This thermal, stimuli-responsive luminescence change is known as luminescence thermochromism. This review focuses on the systematic developments of metal-containing crystalline luminescence thermochromic materials, which will be of interest to researchers attempting to design and develop new temperature-induced luminescence-changing materials. Additionally, focusing on crystalline materials provides direct insight into the chromic mechanisms of luminescence thermochromism, which are also discussed.

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Abbreviations: LE, low-energy; ³CC, triplet cluster-centered; XMCT, halide-to-metal charge transfer; XLCT, halogen-to-ligand charge transfer; MLCT, metal-to-ligand charge transfer; HE, high-energy; DFT, density functional theory; SCO, spin crossover; bib, 1,4-bis(1-imidazolyl)benzene; bix, 1,4-bis-(imidazol-1-ylmethyl)benzene; ³Py, 3-pyridyl; ³Qy, 3-quinolyl; MBI, 1,1'-methylene-bis(imidazole); MIM, 1-methylimidazole; LMCT, ligand-to-metal charge transfer; Bu₂DABCO, N,N'-dibutyl-1,4-diazabicyclo-[2.2.2]octane; py, pyrazole; DMBDC, 2,5-dimethoxy-1,4-benzenedicarboxylate; H₂PIA, 5-(pyridin-4-yl)isophthalic acid; CIE, Commission Internationale de l'Eclairage; H₂BPDA, biphenyl-3,5-dicarboxylic acid; H₂DSTP, 2,4-(2,2':6',2''-terpyridin-4'-yl)-benzenedisulfonic acid; OA, oxalic acid; BDC, 1,4-benzene dicarboxylic acid; H₄QPTCA, 1,1':4',1'':4'',1'''-quaterphenyl-3,3''',5,5''''-tetracarboxylic acid; H₂FDC, 9-fluorenone-2,7-dicarboxylic acid; Pip, piperazine; MePip, N-methylpiperazine; Me₂Pip, N,N'-dimethylpiperazine; EtPip, N-ethylpiperazine; Et₂Pip, N,N'-diethylpiperazine; BzPip, N-benzylpiperazine; Bz₂Pip, N,N'-dibenzylpiperazine; Ph₂CHPip, N-(diphenylmethyl)piperazine; HMTA, hexamethylenetetramine; ³MC, metal-centered transfer; pyz, pyrazine; H₂bdc, benzene-1,3-dicarboxylic acid; VTSCXRD, variable-temperature single-crystal X-ray crystal structure; HTB, thiobenzoic acid; C₂H₄N₄, 4-NH₂-1,2,4-triazole; C₁₉H₁₂N₄, 1-pyrenecarboxaldehyde; C₃₀H₃₃N₆O₂Cl, Rhodamine B; 4-mbaH, 4-(methoxycarbonyl)benzenethiolate; P⁺S, 2-(diphenylphosphino)-benzenethiolate; PPh₃, triphenylphosphine; dpd, dipyridin-4-yl-diazene; bipy, 4,4'-bipyridine; dpbz, 1,4-bis(4-pyridyl)benzene; PyS⁻, pyridine-4-thiolate; HPyS, 1H-pyridine-4-thione; H₂mbs, 2-mercaptobenzoic acid; H₂sbs, 2-sulfobenzoic acid; ^tPr, isopropyl; Pyr, pyrrole; dppy, diphenylphosphino-2-pyridine; HMTZ, 1-{2-(N,N-dimethylamino)ethyl}-5-mercapto-1H-tetrazole; BTB, 1,3,5-tris(4-carboxyphenyl)-benzene; ATZ, 5-amino-1-H-tetrazolate; ILCT, intra-ligand charge transfer; LLCT, ligand-to-ligand charge transfer; H₆TTHA, 1,3,5-triazine-2,4,6-triamine hexaacetic acid; Hpt, pyridinium-4-thiolate; dps, 4,4'-dipyridylsulfide; H₂hpi₂cf, 5-(2-(5-fluoro-2-hydroxyphenyl)-4,5-bis(4-fluorophenyl)-1H-imidazol-1-yl)isophthalic acid.

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1. Introduction

Stimuli-responsive functional materials with specific properties have attracted increasing attention due to their use in a wide variety of applications, such as memory, chemical sensors, optical recording, display devices, and data storage [1]. Various external stimulations, such as light [2–9], mechanical or pressure [10–19], vapor [20–29], solvent [30–37] and temperature [38–150], have been extensively utilized to induce physical property changes. Among these stimulations, physical property changes can result from a combination of multiple external stimuli. Thermal triggers are probably the most widespread external stimuli found in nature, while color changes are the most striking phenomena. Usually, color changes are collectively termed ‘chromotropism’.

In theory, due to the Boltzmann distribution of excited-state electrons, the luminescence of all materials is dependent on the temperature [151,152]. The relationship between the nonradiative transition rate (k_{nrt}) and temperature (T) fits the Arrhenius equation:

$$k_{\text{nrt}} \sim (-\Delta E/kT)$$

Here, ΔE is the energy separation between the bottom of the excited state and the overlap point, and k is the Boltzmann constant. According to this equation, k_{nrt} is directly proportional to T . Upon increasing the temperature, the nonradiative transition rate increases, which results in a diminished luminescence intensity. However, the luminescence thermochromic mechanisms of metal-containing materials remain elusive for multiple excited bands that possess different overlap energies, which results in complicated electronic band structures and temperature-dependent luminescence properties.

Luminescence thermochromic materials change luminescence in response to temperature stimuli. Excellent reviews concerning temperature-dependent optical properties of a discrete molecular entity have been reported [153–155,157]. Metal-containing complexes constructed by inorganic and organic moieties possess great advantages in constructing luminescence thermochromic materials: (1) Both the inorganic and organic moieties can generate luminescence, and the metal-ligand charge transfer can result in diversity luminescence change, (2) Polynuclear metal clusters or coordination polymers based on d^8/d^{10} electronic structure provide metallophilic interactions, and slight changes of these interactions result in marked changes in their photophysical properties, and (3) complexes based on lanthanide(III) ions can exhibit unique intrinsic optical properties due to their intra-4f electronic transitions of the metal ions or ion-to-ligand back energy transfer. To assemble stable, efficient, easily visible luminescence thermochromic materials, understanding the relationship between the molecular structures and their resultant properties is essential. Crystalline states enable monitoring of the structural changes at different temperatures. Therefore, it is advantageous to probe the mechanism of external stimulation involving temperature to gain an in-depth understanding of structure-property relationships. With the aim to provide an enhanced understanding of metal-containing, crystalline luminescence thermochromic materials, detailed, temperature-based luminescent changes are summarized in this review. In addition, some guided perspectives for future research are also provided.

2. Luminescence thermochromism

Thermochromic luminescence properties are characterized by a reversible change of the emission wavelength with a temperature change. Hardt and coworkers first suggested the term ‘fluorescence thermochromism’ to refer to temperature-dependent emissive behavior [38]. To date, many efforts have been devoted to developing new luminescence thermochromic materials. Additionally, the luminescence thermometer, or thermochromism, has been investigated in the fields of biochemical activity, polymers and nanoparticles [39–47]. Although a review provided a comprehensive summary covering many aspects of temperature-dependent luminescent probes and sensors [48], the aim of this review is to provide several large families of metal-containing luminescence thermochromic substances, such as Cu-halide clusters [47–75], lanthanide and mixed-lanthanide coordination polymers [78–101], coinage metals [102–138], and complexes with other transition metals [139–150]. Usually, luminescence thermochromism based on Cu(I)-halide complexes is attributed to a low-energy (LE) triplet cluster-centered (^3CC) excited state and halide-to-copper charge transfer ($^3\text{XMCT}$) transition at room temperature or a high-energy (HE) triplet halide-to-ligand charge transfer ($^3\text{XLCT}$) excited state and metal-to-ligand charge transfer ($^3\text{MLCT}$) transition at low temperature. Ion-to-ligand back-energy transfer or effective sensitization of organic ligands from Tb^{3+} to Eu^{3+} ions may be the main reason for the luminescence thermochromism of lanthanide and mixed-lanthanide coordination polymers. For coinage metals, temperature-dependent metallophilicity variations can be utilized to explain luminescence thermochromism. In addition, spin crossover (SCO) complexes are usually accompanied by luminescence variations triggered by temperature changes.

2.1. Cu-halide clusters

Complexes based on Cu(I)-halide clusters have captured the attention of researchers for their highly emissive nature and high quantum yields. Most copper compounds and clusters have exhibited multistimuli-responsive behaviors [55–77]. For more general discussions of multistimuli-responsive luminescent materials, refer to the excellent reviews from Cariati, Ford, and Vogler [49–52]. Among these materials, polynuclear copper-iodine clusters are promising candidates, as they present appealing and peculiar cubane-like Cu_4I_4 clusters associated with rich photophysical properties (Table 1). Examples of copper-iodine cluster-based hybrid inorganic-organic materials with intriguing luminescent thermochromic behaviors are highlighted. The relative intensity change of two distinct emission bands (HE and LE) dependent on the temperature was the origin of luminescence thermochromism in copper-iodine clusters. At room temperature, the LE emission band at approximately 550–650 nm is dominant, corresponding to a ^3CC consisting of a mixture of $^3\text{XMCT}$ s, which can be affected by the outer environment, such as changes to the solid-state structure, temperature, pressure or solution. At low temperature, the HE emission band at approximately 400–450 nm is dominant, corresponding to an $^3\text{XLCT}$ and an $^3\text{MLCT}$. The final emission color can be effectively controlled by the intensity ratio of the LE and HE bands.

Perruchas and coworkers prepared a series of compounds based on Cu_4I_4 or Cu_6I_6 clusters, $[\text{Cu}_4\text{I}_4\{\text{PPh}_2(\text{OCH}_2\text{CH}_3)\}_4]$ (**1**) [69], $[\text{Cu}_6\text{I}_6(\text{PPh}_2(\text{CH}_2)_3\text{PPh}_2)_3]$ (**2**), $[\text{Cu}_4\text{I}_4(\text{PPh}_3)_4]$ (**3**), and $[\text{Cu}_4\text{I}_4(\text{PPh}_3)_4]$

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