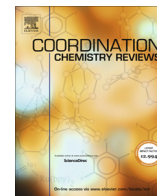




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Review

Energy materials based on metal Schiff base complexes

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ABSTRACT

Metal Schiff base complexes represent a class of compounds that have become a field of immense interest because of their intriguing chemical and physical properties, and their wide-ranging applications in a number of scientific areas. The presence of transition metal elements with a polydentate Schiff base ligand to form metal complexes offers a good platform for combining the chemical, electronic, magnetic, optical and redox properties of metal complexes with those of the organic materials. Metal Schiff base complexes can be incorporated into discrete small molecules, oligomers or polymers, generating new functional materials with useful mechanical, catalytic, thermal, chemical and optoelectronic properties. This review presents the contemporary research development of the field, with emphasis on the fundamental concepts, facile tuning of the photophysical properties and possible energy-related applications of various functional metal Schiff base complexes. To date, many soluble conjugated materials based on metal Schiff base complexes have been generated and studied. They have found an array of energy applications, for example, as converters for light/electricity signals in organic light-emitting diodes and dye-sensitized solar cells, energy storage and potential conductive thermoelectric materials.

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Abbreviations: A, electron acceptor; acac, acetylacetonate; Alq₃, tris(8-hydroxyquinolino)aluminum; BALq, bis(2-methyl-8-quinolinolato)-mono(4-phenylphenolato)aluminum; BCP, 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline; Bepp₂, bis[2(2'-hydroxyphenyl)pyridine]beryllium; CBP, 4,4'-bis(N-carbazolyl)-1,1'-biphenyl; CE, current efficiency; CIE, Commission International de L'Eclairage; CuPc, copper phthalocyanine; D, electron donor; Da, Dalton unit; DFT, density functional theory; DMF, dimethylformamide; E_g, optical bandgap; EIS, electrochemical impedance spectroscopy; E_{ox}, oxidation potential; EL, electroluminescence; ett, 1,1,2,2-ethenetetrathiolate; EQE, external quantum efficiency; FF, fill factor; Flrpic, bis(4,6-difluorophenyl)pyridine(picolate)iridium(III); FK306, iridium(III) bis[4-(tert-butyl)-2',6'-difluoro-2,3'-bipyridine]acetylacetonate; GPC, gel permeation chromatography; HOMO, highest occupied molecular orbital; ITO, indium-tin oxide; ICT, intramolecular charge transfer; IL, intraligand; ILCT, intraligand charge transfer; IPCE, incident photon-to-electron conversion efficiency; LC, ligand-centered; LCD, liquid crystal display; LEC, light-emitting electrochemical cell; LUMO, lowest unoccupied molecular orbital; MLCT, metal-to-ligand charge transfer; NIR, near-infrared; NPB, 4,4'-bis[N-(1-naphthyl)-N-phenylamino]biphenyl; M_n, number-average molecular weight; OEP, octaethylporphyrin; OLED, organic light-emitting diode; PBD, 2-(4-biphenyl)-5-(4-tert-butylphenyl)-1,3,4-oxadiazole; PCE, power conversion efficiency; PE, power efficiency; PEDOT:PSS 3, 4-poly(ethylenedioxythiophene):poly(styrenesulfonate); PF, power factor; PF2/6, poly[9,9-bis(2-ethylhexyl)fluorene]; PL, photoluminescence; PLED, polymer light-emitting diode; PMMA, poly(methyl methacrylate); PVK, poly(N-vinylcarbazole); salen, N,N'-bis(salicylimine)-1,2-ethylenediamine; salen-Me₄, N,N'-bis(salicylimine)-1,1,2,2-tetramethylethylenediamine; salphen, N,N'-bis(salicylimine)-1,2-phenylenediamine; salpn, N,N'-bis(salicylimine)-1,3-propanediamine; TCTA, 4,4',4''-tri(9-carbazolyl)triphenylamine; TE, thermoelectric; TEMPO, tetramethyl-1-piperidinyloxy; THF, tetrahydrofuran; UV-vis, ultraviolet-visible; M_w, weight-average molecular weight; WOLED, white organic light-emitting diode; ZFS, total zero-field splitting; Φ_{em}, photoluminescence quantum yield; λ_{max}, absorption wavelength maximum; λ_{em}, peak emission wavelength; τ, emission lifetime; J_{sc}, short-circuit current density; LT₅₀, device lifetime; V_{oc}, open-circuit voltage; S, Seebeck coefficient; T_d, decomposition temperature; σ, electrical conductivity; K, thermal conductivity.

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1. Introduction

A major global problem confronting the world today is the energy crisis. While the quality of life for the humankind depends largely on the availability of energy, the world is becoming increasingly dependent on new energy sources because of the upcoming depletion of fossil fuels [1]. One of the grand challenges of this century is to develop renewable energy systems that can give efficient and effective utilization of energy. Renewable energy sources such as solar power can provide a large amount of energy and could potentially solve our immediate energy needs. There is a growing concern that combustion of fossil fuels is causing an accumulation of CO₂ in the atmosphere, leading to many detrimental effects on the environment. Recent predictions show that it is necessary to find an additional 14–20 TW by 2050 when our energy reserves based on fossil fuels are vanishing [2]. Thus, scientists are looking for other sustainable energy sources to meet our future energy demand and fight for a cleaner environment. There is much research attention in developing renewable resources and improving the technologies for energy interconversions. The transformations of light into electricity (electricity generation in photovoltaic cells) and electricity into light (light generation in light-emitting diodes) are two important interrelated areas that have attracted considerable research interest in recent years [3]. Coordination and organometallic compounds have been sought and investigated for both of these transformations and these metal complexes and polymers will undoubtedly play key roles in the efficient production, transformation and utilization of energy [4]. The chemical and physical properties of such metal-based material can be easily fine-tuned simply by varying its chemical structure (both the metal center and/or organic ligand) to develop a suitable material to fit a particular energy application.

New functional materials with tailor-made properties, low cost and straightforward synthesis are of central importance in the topical themes of material sciences. Transition metal complexes with interesting optoelectronic properties are increasingly used in the design of novel functional molecular materials [5–10]. In general, the advantages of metalation includes (i) the capability to generate active species for charge transport through the redox activity of both the metal atoms and the organic ligand; (ii) easy fine-tuning of the highest occupied molecular orbital–lowest unoccupied molecular orbital (HOMO–LUMO) energy gap through the interaction of the d-orbitals of the transition metal with the HOMO and/or LUMO of the ligand; (iii) high diversity of the molecular framework based on the coordination number, geometry and valence shell of the selected metal atom and (iv) improvement of the solubility and ease of tuning the intermolecular π -stacking and/or metallophilicity in the solid state. Among these, metal Schiff base complexes are a broad class of compounds that have received increasing attention due to their attractive chemical and physical properties, and their wide-ranging applications in various fields [11–13].

The class of compounds, so-called imines, are often referred to as Schiff bases, named after the German chemist Hugo Schiff. Schiff bases contain the azomethine group ($-\text{RC}=\text{N}-$) and are typically formed by the condensation of a primary amine with an active carbonyl compound [14]. They have drawn considerable research attention of scientists owing to the ease of synthesis and metal complexation. Transition metal complexes of N₂O₂ Schiff base derived from salicylaldehyde and diamines have attracted intense

interest as a promising class of luminescent materials [12]. Among these, metalated *N,N'*-bis(salicylimine)-1,2-ethylenediamine (salen), **I** (abbreviated as [M(salen)]), is the prototype of this family of compounds, which are known to exhibit intriguing electroluminescent (EL) [12], nonlinear optical (NLO) [15] and catalytic properties [16,17]. Other related compounds, such as [M(salphen)] **II** (salphen = *N,N'*-bis(salicylimine)-1,2-phenylenediamine), [M(salpn)] **III** (salpn = *N,N'*-bis(salicylimine)-1,3-propane diamine) and [M(salen-Me₄)] **IV** (salen-Me₄ = *N,N'*-bis(salicylimine)-1,1,2,2-tetramethylethylenediamine) (Fig. 1), demonstrate the breadth of chemical structures available for this class of molecules, which possess the central N₂O₂ binding pocket formed by two imines and two phenoxides [13]. In the past few decades, because of their preparative accessibility and high thermal stability, M(salen)-type motifs have been commonly introduced into small discrete molecules, oligomers and polymers in the hope of generating new functional materials with desired features [12,13].

In this review, we highlight the types and photophysics of various metal Schiff base systems, with special emphasis placed on those with transition metal elements such as platinum(II), nickel (II) and zinc(II). As the synthetic chemistry of metal Schiff base complexes is prevalent, the study of their photophysical and electronic properties would provide a good access to new classes of functional molecular materials of practical interest. A survey of the reported metal Schiff base compounds for various energy-related applications is summarized here, which are useful for organic light-emitting diodes (OLEDs), dye-sensitized solar cells, energy-storing and potential thermoelectric applications.

2. Electrical to light energy conversion in organic light-emitting diodes

Recent research on organic light-emitting diodes (OLEDs) has been focused on devices composed of thin films containing organometallic molecules that can directly convert electricity into light [18–20]. OLEDs have the great potential of revolutionizing display technologies in the scientific community. The key advantages of OLEDs for flat-panel display applications are their self-emitting property, high luminous efficiency, full color capability, wide viewing angle, high contrast, low power consumption, low weight, potentially large-area color displays and flexibility [21]. They are also more energy-efficient and are generally lower in cost than liquid crystal displays (LCDs). In view of this, the rapidly growing market for OLED technology is driving both the academic and industrial sectors toward the development of new functional materials for advanced manufacturing technology. Heavy metal complexes have recently gained tremendous research interest for fabricating highly efficient phosphorescent OLEDs by taking advantage of the 1:3 exciton singlet/triplet ratio predicted by simple spin statistics [18–21]. These phosphorescent emitters are mainly derived from the family of the third-row transition metal (Re(I), Os(II), Ir(III) and Pt(II)) complexes [22–24], although examples with some second-row transition metals such as Ru(II) [22] and first-row transition metals such as Cu(I) are also known [25,26]. The three key electronic processes, namely, (i) charge injection, (ii) charge transport, and (iii) electron-hole recombination (i.e., exciton formation) efficiency must be individually optimized in order to improve the overall OLED performance. The pioneering

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