1.1 Introduction

1.1.1. History of Gel

The history of gels is quite fascinating, as these versatile substances have been used for a variety of purposes throughout human history[1]. The ancient Egyptians used a gel-like substance made from a combination of plant extracts and natural gums as a binding agent for their cosmetics and perfumes[2][3]. Additionally, the ancient Greeks and Romans utilized gels made from natural ingredients for medicinal purposes[4][5], such as soothing skin ailments and wounds. In the 19th century the chemist Thomas Graham coined the term "colloid" to describe substances with particles dispersed in a medium, which included gels[6]. He also studied colloidal systems and their properties, making important contributions to the scientific understanding of gels[7]. The development of modern gel technology accelerated during the early 20th century[8][9]. Chemists and scientists began synthesizing new types of gels using synthetic polymers. One notable example is the invention of "jelly" or "gelatin dessert" in 1897 by Pearle Wait, which eventually became known as Jell-O[10]. Gels found various applications during World War II. For example, synthetic gels were used in military equipment[11][12], such as radar systems[13][14], as damping materials to reduce vibrations and shocks[15][16]. From the mid-20th century onwards, gels have found extensive applications in various industries[17][18], including pharmaceuticals, cosmetics, food, and technology[19]. Gel-based formulations have become an integral part of consumer products, ranging from personal care items like shampoos and toothpaste to medical products such as topical ointments and drug delivery systems[20]. Additionally, in molecular biology and biotechnology, gels are vital tools, with agarose and polyacrylamide gels frequently employed in electrophoresis techniques for separating DNA, RNA, and proteins according to their size and charge[21][22][23].

1.1.2. An Overview of the Gel

Gel is a substance that can exist in various forms, including a semi-solid or solid-like state[24]. It is characterized by its ability to retain a significant amount of liquid within its three-dimensional network of molecules[25]. Gels and other jelly-like materials can be composed of natural or synthetic materials and are commonly used in a wide range of applications, including personal care products, pharmaceuticals, and scientific research[26]. A gel comprises two phases: a liquid phase and a solid phase made up of gelator molecules that enclose the liquid phase (solvent)[27]. Achieving the gel state involves a delicate equilibrium. It's important to note that not every molecule is suitable as a gelator, and not every solvent can facilitate gel formation[28][29]. When molecules become solubilized in a solvent, three primary outcomes can occur. First, crystals may develop when molecules begin to self-assemble in an extremely organized manner. The second possibility is that molecules will randomly group, which could result in an amorphous. Third, the self-assembly in an orderly manner by entrapping the solvent, gel material is formed[30][31].

Supramolecular chemistry is still a young field and related scientists are keenly involved in developing a variety of applications for the benefit of mankind. Thanks to the great contribution of researchers like James Fraser Stoddart, who developed molecular machinery and self-assembled structures, supramolecular chemistry advanced during the 1990s[32]. Supramolecular systems were merged with electrochemical and photochemical themes, synthetic self-replicating systems, and molecular information processing at this time. In addition to its versatility, nanotechnology is a strong output of supramolecular chemistry.

The term "supramolecular chemistry" was introduced by J. M. Lehn[33], and his colleagues C. J. Pedersen and D. J. Cram in 1978 and awarded the Nobel Prize for Chemistry in 1987[34].

Low molecular weight gelator (LMWG) molecules and different solvent molecules spontaneously come together through a variety of non-covalent interactions, including hydrogen bonding, p-stacking, and van der Waals forces, to create supramolecular gels. Small organic compounds with a molecular mass of typically less than 3000 are known as LMWGs[35][36]. LMWGs immobilize organic solvents for organogels and aqueous solvents for hydrogels, resulting in the formation of a supramolecular gel network. Supramolecular gels have a wide range of potential uses, including as sensors[37][38], structure-directing agents[39][40], cosmetics[41], art conservation, bio-medical applications, lithography[42][43], optoelectronic devices[44][45], electrochemical devices, cell culture[46], medical diagnostics, biomineralization[47][48], drug delivery, tissue engineering and also used in molecular biology for DNA analysis organic electronics[49], semiconducting molecular gelator[50], optical filter, and photoconductivity[51], among others.

Figure 1.1 Gels materials that find extensive use in various field.

Overall, gels are versatile materials that find extensive use in various industries due to their unique properties and ability to hold and deliver liquids within a solid or semi-solid matrix shown in **Figure 1.1**.

1.2. Classification of Gel

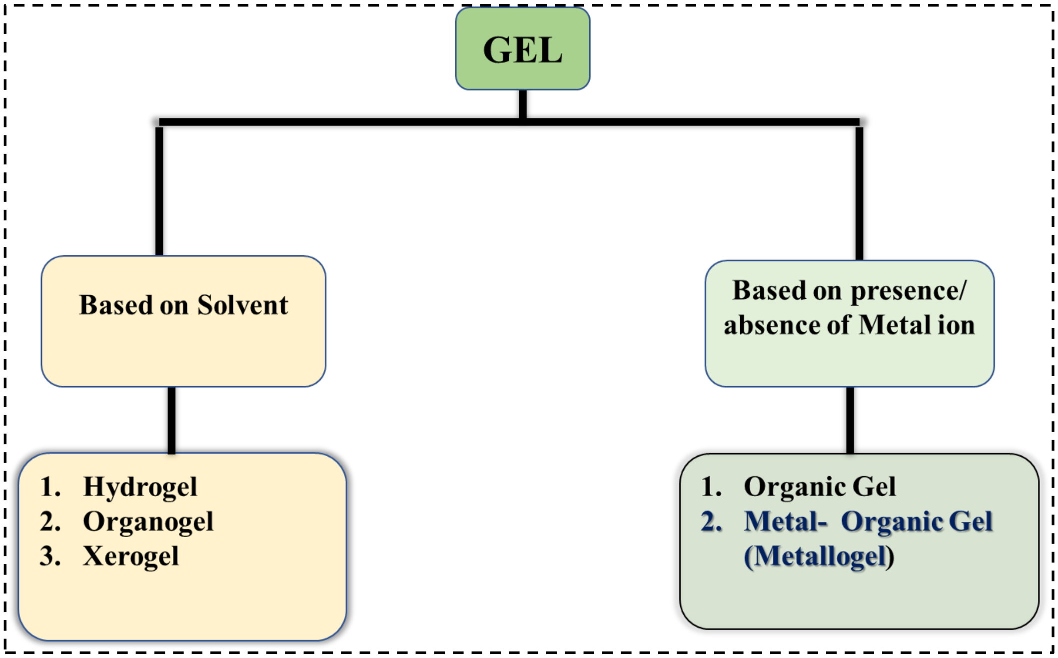

Figure 1.2 A detailed chart for the classification of gels

1.2.1 Hydrogel

Hydrogel is a type of gel composed of a three-dimensional network of hydrophilic polymers that can absorb and retain a large amount of water or aqueous solutions[52]. One of the notable characteristics of hydrogels is their ability to swell in the presence of water or other aqueous solutions while maintaining their three-dimensional structure[53]. The swelling behavior is primarily due to the hydrophilic nature of the polymer chains, which have a strong affinity for water molecules[54]. The unique properties of hydrogels, including their high water content, biocompatibility[55], and tunable mechanical properties, make them versatile materials with a wide range of applications in different industries, and hydrogels are extensively used in biomedical research and healthcare[56]. They are employed as scaffolds for tissue engineering and regenerative medicine, drug delivery systems[52][57], wound dressings, contact lenses, and biosensors[58], personal care product.

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1.2.2. Organogel

An organogel is a type of gel formed by organic molecules that can self-assemble into a three-dimensional network structure in an organic solvent. Unlike hydrogels, which are formed in water or aqueous solutions, organogels are formed in non-aqueous or organic solvents, that make up organogels are typically amphiphilic[59], meaning they have both hydrophilic (water-attracting) and hydrophobic (water-repelling) parts.

Organogels can be composed of various organic compounds, such as low-molecular-weight gelators (LMWGs) or small organic molecules, as well as some polymers. These gelators have the ability to form aggregates or fibrillar structures in the solvent, which then entangle or form a network, resulting in the gel-like consistency. The properties of organogels can be tuned by altering the composition of the gelators and the organic solvent used. The gel structure, mechanical strength, and gelation properties can be modified to suit specific applications. The gel properties can be affected by factors such as temperature, concentration, pH, and the nature of the organic solvent[60][61].

1.2.3. Xerogel

The term "xerogel" comes from the Greek words "Xero" meaning dry and "gel" referring to the gel-like nature of the material.

The drying process of a xerogel involves removing the liquid component of a gel under carefully controlled conditions, typically through methods such as evaporation, supercritical drying, or freeze-drying. The result is a solid material with a highly porous structure that retains the original shape and three-dimensional network of the gel.

Xerogels are known for their extremely low moisture content and high surface area due to the porous nature of their structure[62]. Overall, xerogels offer a versatile platform for the development of materials with tailored properties, particularly in applications that require high surface area, low moisture content, and controlled porosity[63].

1.2.5. Metallogel

Metallogels are supramolecular self-assembled molecules in which metal ions get assimilated with the suitable organic gelators into the 3-D soft gel-structure[64]. The gelation process in metallogels is typically achieved through the coordination or cross-linking of metal ions or metal complexes with ligands or polymers. The metal-ligand interactions lead to the formation of a gel network, which can exhibit diverse properties depending on the choice of metal and ligand components. Metallogels have attracted significant research interest due to their unique properties and potential applications. The field of metallogels is still an active area of research, with ongoing efforts to understand their properties, design new gelators, and explore novel applications. Metallogels offer a promising platform for developing functional materials with unique metal-based properties and applications.

1.3. Why Metallogel???

The metallogels as a material of interest can offer several advantages and opportunities in various applications. A few of them can be described as follows.

Structural Diversity: Metallogels exhibit a wide range of structural morphologies, including fibers, particles, and interconnected networks. This structural diversity allows for the design and fabrication of materials with tailored properties and functionalities.

Tunable Properties: Metallogels can possess tunable mechanical, optical, and chemical properties. By selecting different metal ions, ligands, or coordination chemistry, the properties of the metallogel can be adjusted to meet specific requirements for a particular application.

Stimuli-Responsiveness: Metallogels can be engineered to respond to external stimuli such as changes in temperature, pH, or light. This responsiveness enables the development of smart materials and systems that can adapt, sense, or release substances in response to environmental cues.

Enhanced Catalytic Activity: Incorporating metal catalysts or metal nanoparticles into metallogels can lead to enhanced catalytic activity and selectivity. This makes metallogels attractive for applications in catalysis and chemical reactions.

Energy Storage: Metallogels can be utilized in energy storage devices such as batteries and supercapacitors. The high surface area and porosity of metallogels, along with the presence of redox-active metal centers, can contribute to improved energy storage performance.

Biomedical Applications: Metallogels can be tailored to exhibit biocompatibility and controlled release properties, making them valuable in biomedical applications. They can be used for drug delivery, bioimaging, tissue engineering scaffolds, and other applications in the field of healthcare.

Unique Metal-Based Properties: Metals have unique properties such as conductivity, magnetism, and optical characteristics. Incorporating metals into gels can leverage these properties for applications in electronics, sensors, and optics.

Fundamental Research: Metallogels present opportunities for fundamental research in materials science and chemistry. Understanding the self-assembly processes, coordination chemistry, and gelation mechanisms of metallogels can contribute to advancements in the field and the development of new gelation strategies.

It's important to note that the choice of material depends on the specific requirements and objectives of the intended application. While metallogels offer numerous advantages, their selection should be based on a thorough consideration of the desired properties, compatibility with the targeted environment, and feasibility for fabrication and scale-up.

1.4. IMPORTANT APPLICATIONS OF METALLOGELS

1.4.1 Electronic devices

Metallogel is a unique hybrid material that combines the conductivity of metals with the structural properties of gels. Due to the presence of metal ions, metallogels typically exhibit self-healing and electrical conductivity, making them attractive for use in Schottky diodes and other electronic devices. A Schottky diode is a semiconductor device that consists of a metal-semiconductor junction. It is named after the German physicist Walter H. Schottky. The metal part of the diode, known as the Schottky contact or Schottky barrier, is typically made of metal with a low work function, such as gold, silver, platinum, or aluminum[65]. When a metallogel is used in a Schottky diode, it is typically employed as the metal electrode or contact material[66]. The metallogel provides a conductive path for the current flow and forms a barrier at the metal-semiconductor junction[67]. The key advantage of using a metallogel in this application is its ability to provide stable and reliable contact with the semiconductor material.

Recently, Dey et al[67]. developed a Schottky barrier diode device using metallogel thin film. This device is composed of layers of indium tin oxide (ITO), metallogel, and aluminium in a sandwich-like configuration. In order to explore its electrical behavior, electrical characterizations were conducted on the developed device. As a result of the I-V characteristic of the device, it was revealed that the device behaved non-linearly which is a characteristic of diodes. The metallogel was synthesized from oxalic acid incorporating Mn(II) via coordination complexation

1.4.2. Electrolytic material

Metallogels as an electrolyte have important applications in supercapacitors, solid oxide fuel cells (SOFC), and batteries[68]. Since liquid electrolytes faces problems such as leakage, internal corrosion, volatilization, etc., therefore metallogel based electrolytes are of immense interest in resolving the problem associated with liquid electrolytes[69]**.** Metallogels offer unique properties that can be advantageous for supercapacitor applications, such as high surface area, porosity, and the potential for incorporating redox-active species. The specific metallogel synthesized by Bhattacharjee et.al.[70]. The synthesized metallogel has specific organic ligands (l-2-(3,5-ditert-butyl-2-hydroxybenzyl amino)-succinic acid) and zinc sulfate ($ZnSO_4$). It have exhibited properties that made it suitable for supercapacitor applications, such as high capacitance, cycling stability, or other desirable characteristics.

1.4.3. Sensors

You-Ming Zhang and co-workers report a novel supramolecular metallogel-based high-resolution anion sensor array (Figure 1.3)[71]. The system accurately identifies anions such as CN−, SCN−, S2−, and I− in aqueous solutions. The innovative design concept known as "competitive coordination control AIE mode” makes this sensor array unique, which enables the creation of anion-responsive gels using a single synthesized gelator.

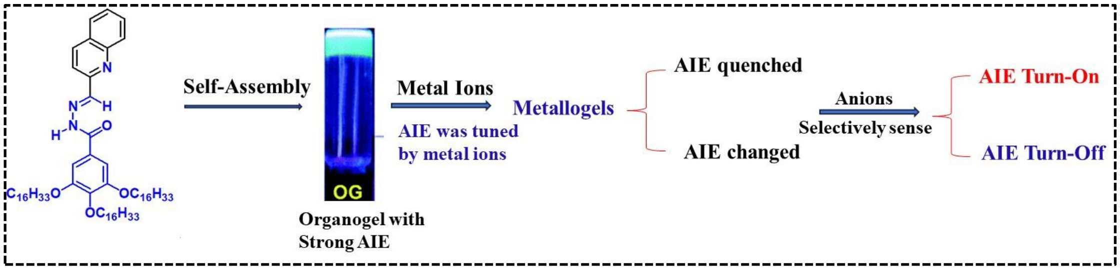

Figure 1.3 supramolecular metallogel-based high-resolution anion sensor array[71]

Alphy Sebastian and Edamana Prasad present a novel metallogel based on copper for detecting cyanide in water using fluorescence "turn-on" signaling (Figure 1.4)[72]. The metallogel was designed and synthesized, utilizing a dendrone derivative (G1) with terpyridine attachments on a poly (aryl ether) backbone. G1 formed a gel and exhibited Aggregation Induced Emission (AIE) properties. By introducing copper ions to the gel, a nonluminescent copper metallogel (CuG) was formed. The copper metallogel selectively detected cyanide in water by generating a fluorescence "turn-on" signal, attributed to the regeneration of the AIE active gel. The sensing mechanism was investigated, and the detection limit was determined to be as low as 1.09 µM. Furthermore, we prepared a thin film of CuG by casting the gel, which was utilized as a test strip for visually detecting cyanide in water.

The study conducted by Pal et al. involves the development of a redox-responsive Cu(I) metallogel for the removal of Cr(VI) ions from aqueous solutions[73]. The metallogel is designed to exhibit redox-responsive behavior, which is attributed to the presence of Cu(I) metal centers throughout its structural backbone. This property allows the metallogel to effectively interact with and remove Cr(VI) ions.

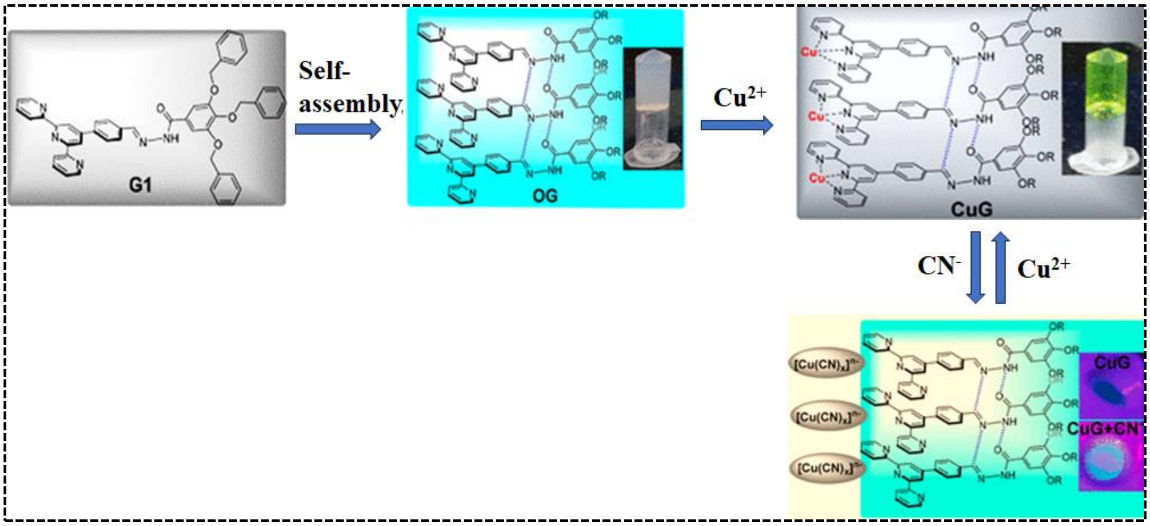

Figure 1.4 The copper metallogel selectively detected cyanide in water by generating a fluorescence "turn-on" signal

To synthesize the metallogel, the researchers utilized 2-mercaptobenzimidazole and $CuCl_2$ as the key components. The synthesis process took place in an ethanolic medium. The specific details of the synthesis procedure, including reaction conditions, concentration, and purification methods, may be outlined in the original research paper by Pal et al.

1.4.4. Drug delivery

Some potential applications of metallogels in drug delivery:

Controlled release of drugs: Metallogels can act as carriers for drugs, allowing controlled release over time. The gel matrix can encapsulate the drug molecules, protecting them from degradation and enabling sustained release. The release rate can be tuned by adjusting the properties of the gel, such as its composition, porosity, and cross-linking density.

Targeted drug delivery: Metallogels can be functionalized with ligands or antibodies that specifically bind to receptors or markers on target cells. This targeting ability enhances the delivery of drugs to specific tissues or cells, reducing off-target effects and improving therapeutic efficacy.

Combination therapy: Metallogels can be designed to deliver multiple drugs simultaneously, enabling combination therapy. Different drugs with complementary mechanisms of action can be loaded into the gel matrix and released in a controlled manner, providing synergistic effects and enhancing therapeutic outcomes.

Imaging-guided drug delivery: Metallogels can incorporate imaging agents, such as fluorescent dyes or contrast agents, to enable real-time monitoring and tracking of drug delivery. This allows visualization of the gel distribution, drug release, and interaction with the target site, providing valuable feedback for optimizing drug delivery strategies.

Theranostics: Metallogels can combine therapeutic and diagnostic functionalities into a single system, known as theranostic agents. By integrating therapeutic agents, imaging agents, and targeting ligands, metallogels can deliver drugs while simultaneously providing diagnostic information, enabling personalized medicine and precise treatment monitoring.

Localized drug delivery: Metallogels can be used for localized drug delivery in specific anatomical sites or tissues. For example, in the field of regenerative medicine, metallogels can serve as scaffolds to deliver growth factors or stem cells to promote tissue regeneration in a targeted manner.

K. Sarkar and P. Dastidar report a series of metallogels was designed based on a newly synthesized dicarboxylate ligand called 5-(3-(pyridin-3-yl) ureido isophthalic acid (PUIA)[74]. This ligand formed metallogels when reacted with different metal salts (Cu(II), Zn(II), Co(II), Cd(II), and Ni(II) salts) at room temperature. The gels were characterized using dynamic rheology and transmission electron microscopy (TEM). The presence of coordination bonds in the gel state was confirmed through FT-IR and 1H NMR spectroscopy in the case of a Zn(II) metallogel (MG2). MG2 exhibited anti-inflammatory properties, as demonstrated by the PGE2 assay conducted on a macrophage cell line (RAW 264.7). It also showed anti-cancer activity in a highly aggressive human breast cancer cell line (MDA-MB-231) based on a cell migration assay. The metallogel matrix of MG2 was able to load and release the anti-cancer drug Doxorubicin in a pH-responsive manner. Fluorescence imaging of MDA-MB-231 cells treated with MG2 confirmed successful internalization of the metallogel.

1.4.5. Catalytic Material

Metallogels can exhibit catalytic activity by incorporating catalytically active metal nanoparticles or complexes within the gel matrix. The gel matrix provides a stable and confined environment for the catalytic species, allowing for enhanced catalytic performance and selectivity.

Metallogels, being intelligent materials, have found applications in catalyzing significant transformations of organic molecules. Some notable findings in this area are summarized below. In their study, S. Dhibar et al. introduced a CoMEA metallohydrogel that demonstrates heterogeneous catalysis for C-S coupling reactions[75], leading to the formation of aryl-sulfur bonds. Remarkably, the CoMEA metallohydrogel facilitates rapid single-pot C-S coupling reactions at room temperature. An intriguing aspect is that the catalytic process with the CoMEA metallohydrogel does not necessitate column-chromatographic purification methods to obtain pure aryl-thioether.

1.4.6. Charge Transfer Behaviour

Charge transfer processes occurring at the supramolecular level, particularly in the gel state, have introduced new dimensions to these materials by harnessing their fascinating photophysical properties. As a result, they have become valuable components in devices such as bulk-heterojunction solar cells and organic transistors. Charge transfer can be categorized into two types based on the presence of donor (D) and acceptor (A) units within a single molecule or different molecules: intermolecular charge transfer and intramolecular charge transfer (ICT). Significant research efforts have been dedicated to synthesizing intermolecular charge transfer gels in the past few decades, primarily due to their outstanding performance in optoelectronic applications. Charge transfer interactions typically involve strong interactions between a donor and an acceptor molecule, leading to their orderly stacking in an alternating fashion, ultimately resulting in the formation of charge transfer gels.

Nalluri et al. conducted a study where they developed a chiral supramolecular organic gel through the enzymatic condensation of a NDI-functionalized tyrosine (NDI-Y) and phenylalanine-amide (FNH2). Additionally, these building blocks, when mixed with hydroxyl or alkoxy donor compounds such as 1,5-dialkoxy naphthalene (1,5-DAN), 2,6-dialkoxy naphthalene (2,6-DAN), 1,5-dihydroxy naphthalene (1,5-DHN), and 2,6-dihydroxy naphthalene (2,6-DHN), generate a charge transfer interaction. The incorporation of these donor molecules leads to the formation of one-dimensional (1D) donor-acceptor (D-A) stacks, thereby facilitating charge transfer interactions[76].

1.4.7. Stimuli Responsive Materials

The responsiveness of metallogels refers to their ability to undergo reversible changes in their structure, properties, or behavior in response to specific stimuli. These stimuli can include changes in temperature, pH, light, electric or magnetic fields, or the presence of certain chemicals or ions[77]. These external stimuli trigger a response in the metallogel, leading to changes in its physical or chemical properties.

Dillip K. Chand et.al have successfully created a self-assembled binuclear coordination cage with the Pd_2L_4 composition. This was achieved by combining $Pd(NO_3)_2$ with N,N'-bis(3-pyridylmethyl)-naphthalenediimide (L). Microscopy studies revealed the formation of rare nanoscale metal-organic particles within the metallogel's structure[78]. Notably, the gel exhibited thixotropic (mechanoresponsive) properties and showed reversible responsiveness to chemical stimuli. The presence of naphthalenediimide units in the cage's backbone and the positively charged cavity of the cage offer potential for studying the functional aspects of the gel. The porous gel demonstrated the ability to absorb pyrene as a guest molecule and selectively remove anionic dyes from aqueous solutions. Specifically, the gel successfully bound anionic dyes like "acid blue 93" and "methyl orange" in the presence or absence of certain cationic dyes, making it suitable for selective dye removal applications.

1.4.8. Chiral Metallogels

By introducing chirality into the metallogelator, fascinating alterations can occur in its structure. One of the notable changes is the introduction of twisting or helical patterns into the fibers of the metallogel. Chirality imparts a handedness or asymmetry to the molecular arrangement, leading to these intriguing three-dimensional shapes. Additionally, the optical properties of the metallogel can be significantly affected by chirality. Chiral molecules have distinct interactions with polarized light, resulting in different responses to left-handed circularly polarized light (LCP) and right-handed circularly polarized light (RCP). This phenomenon, known as circular dichroism (CD), allows chiral metallogels to selectively absorb or reflect light of a particular handedness. The combination of these structural and optical changes makes chiral metallogels a subject of great interest in materials science and nanotechnology, offering new possibilities for designing advanced materials with unique properties and functionalities.

In a recent study by their research, Sun et al. utilized an alanine-based chiral ligand that, upon binding with metal ions, formed chiral organoplatinum(II) metallacycles with specific shapes, including rhomboids and hexagons[79]. Further, under appropriate conditions, these metallacycles underwent additional assembly, forming fine nanospheres. The nanospheres then experienced additional aggregation through interactions such as H-bonding and π-π stacking, eventually leading to the creation of chiral metallogels. What makes these metallogels particularly interesting is that, upon examination using TEM analysis, they displayed a helical morphology. This finding highlights the unique self-assembly behavior of the chiral metallogels, where the helical structures emerge as a result of the specific interactions and arrangements of the metallogel components. Such helical metallogels have potential applications in various fields, such as materials science, nanotechnology, and even in biomedical settings, due to their distinctive properties and structures.

In a study by Tu et al., they discovered a fascinating chiral metallogel with the unique ability to distinguish between (R)- and (S)-binap, which are enantiomers of a chiral compound. The metallogel displayed a distinct response when exposed to each enantiomer. In the presence of (R)-binap, the metallogel underwent a collapse or structural change, indicating a strong interaction or recognition with this specific enantiomer. On the other hand, when (S)-binap was introduced, the metallogel's architecture remained unchanged, suggesting that it did not interact significantly with this enantiomer. This selective and differential response of the chiral metallogel towards (R)- and (S)-binap demonstrates its potential as a useful tool for chiral recognition and discrimination.

1.4.9. Fluorescent Metallogels

Metallogels with fluorescent properties have gained significant attention in recent years due to their unique combination of structural integrity and luminescent behavior[80]. The integration of metallogels with fluorescence opens up several exciting applications across different fields. Some potential applications of fluorescent metallogels include:

Sensing and Detection: Fluorescent metallogels can be used as highly sensitive and selective sensors for various analytes. By incorporating specific ligands or functional groups within the metallogel network, they can selectively respond to particular chemicals or environmental changes, emitting fluorescence when a target molecule is present. This makes them valuable tools for environmental monitoring, medical diagnostics, and food safety testing.

Imaging: The fluorescent properties of metallogels make them excellent candidates for bioimaging applications. When combined with biocompatible components, they can be used as imaging probes to visualize cells, tissues, and even specific biomolecules. This has potential applications in medical imaging, bioanalytical techniques, and cellular research.

Light-Emitting Devices: Fluorescent metallogels can serve as the active component in light-emitting devices. By incorporating suitable metal complexes or quantum dots into the gel network, they can emit light of specific colors, making them useful for developing efficient and tunable LEDs or other optoelectronic devices.

Photocatalysis: Some fluorescent metallogels can exhibit photocatalytic behavior, utilizing light energy to drive chemical reactions. By incorporating specific metal complexes with photocatalytic activity into the gel, these materials can be employed in areas like water purification, pollutant degradation, and organic synthesis.

Data Storage and Security: The fluorescent properties of metallogels can be harnessed in data storage and security applications. They can be used as fluorescent markers or tags for encoding and decoding information, and their unique properties can help improve the security of encrypted data.

Soft Robotics: The mechanical properties of metallogels, combined with their fluorescence, can be utilized in soft robotics. These materials can serve as actuators or sensors that respond to external stimuli such as light, enabling more advanced and adaptable soft robotic systems. By strategically incorporating AIE (Aggregation-Induced Emission) phenomena into a self-assembled gel network, X. Ma and colleagues achieved the synthesis of a straightforward yet intelligent fluorine-gelator based on multiple pseudoamide units. Through non-covalent interactions, the deliberate inhibition of intermolecular rotation and vibration facilitated the increased radiative decay of excited state species[81]. The main driving force for the formation of the gel was the presence of multiple functionalities capable of hydrogen bonding and metal coordination sites. The distinguishing feature of this ligand was its ability to intelligently sense the presence of Al^{3+} ions amidst other cations, as indicated by significant luminescence enhancement. Building upon this intriguing outcome, the researchers proposed a novel writable gel material that could be easily erased by an F^- ion solution. This erasure was attributed to the precipitation of AlF_3, which promptly quenched the fluorescence through the recovery of PET (Photoinduced Electron Transfer) phenomenon.

1.5. Findings of the Literature

Metallogels are a subclass of supramolecular gels, where metal ions play a significant role in the gelation process. These gels are formed through the self-assembly of metal-containing molecules or coordination complexes. Here are some general findings from the literature are listed in the following sections:

1.5.1. Gelation Mechanisms: The literature on metallogels often discusses the various mechanisms involved in the gelation process. Coordination bonds between metal ions and ligands are typically the primary driving force for gel formation. Factors such as the nature of metal ions, ligand structure, concentration, solvent, and temperature all influence the gelation process.

1.5.2. Tuning Properties: The properties of metallogels can be tuned by altering the metal-ligand coordination, introducing additional functional groups in the ligands, or using different metal ions. This tunability allows researchers to design gels with specific properties for targeted applications.

1.5.3. Rheological Properties: Metallogels exhibit fascinating rheological properties. The rigidity and viscoelasticity of the gels can be tuned by adjusting the gelator concentration or altering the metal-ligand coordination. This makes them attractive candidates for various applications, including in the field of soft materials and tissue engineering.

1.5.4. Characterization Techniques: Researchers have used various techniques to characterize metallogels, including rheology, microscopy (such as SEM and TEM), X-ray diffraction, spectroscopy (NMR, IR, UV-Vis), and computational modeling.

1.5.5. Metallogel as a Semiconductor Substitute: In traditional Schottky diodes, a semiconductor material (usually a doped silicon or gallium arsenide) is used. However, metallogels can act as potential substitutes for traditional semiconductors. Their electronic and optoelectronic properties can be tuned by varying the metal-ligand coordination, allowing for the design of diodes with specific characteristics.

1.5.6. Band Gap: The band gap of a semiconductor material is a critical parameter that determines its electronic behavior. Metallogels can offer the advantage of a tunable band gap, allowing the design of diodes optimized for specific applications, such as photodetectors , light-emitting diodes (LEDs) and Schottky diode.

Schottky diodes are semiconductor devices that use metal-semiconductor junctions to control the flow of electric current. When metallogels are incorporated into these diodes, they can potentially offer unique properties and functionalities. Here are some of the findings and potential applications of metallogel-based Schottky diodes.

1.5.7. Barrier Height Modification: The metallogel interface with the semiconductor can influence the Schottky barrier height, which affects the diode's rectification behavior. Researchers have investigated how different metallogels can modify the Schottky barrier height and impact the diode's electrical characteristics.

1.5.8. Improved Charge Transport: Metallogels' self-healing and gel-like nature might facilitate improved charge transport within the Schottky diode structure, potentially reducing resistive losses and enhancing device performance.

1.5.9. Environmental Stability: Some metallogels have shown good environmental stability and resistance to oxidation and degradation. This stability is essential for the long-term performance and reliability of electronic devices.

It is essential to note that the research on metallogel-based Schottky diodes was still in its early stages, and the number of published studies might have been limited. Since technology and research continue to evolve, there may have been further advancements and new findings in this field since my last update.

1.6. Research Objectives

Based on the comprehensive review of existing literature and the details mentioned earlier, the objective of this research is to create and investigate straightforward methods for fabricating new metallogels and gelation mechanisms, and their potential applications particularly in electrical applications. Additionally, the research also seeks to establish novel materials using a simple synthesis approach that is economically viable.

In this regard, the main objectives of this Ph.D. thesis are as follows:

- An appropriate ligand species is synthesized with a functional group that binds to metal
- As-synthesized gelator is directly gelated into a variety of metal cations under varying solvent conditions
- Exploring the tunability of metallogels by varying the metal ions, gelator molecules, and solvent conditions. This enables researchers to design metallogels with specific properties, such as mechanical strength, thermal stability, and responsiveness to external stimuli.
- Optimizations of synthesis of metallogel and their characterizations.
- Analyzing the structure and morphology of the metallogels using techniques such as X-ray diffraction, scanning electron microscopy, transmission electron microscopy, and nuclear magnetic resonance spectroscopy.
- Investigate the potential of metallogels as semiconducting materials for electronic devices.
- Explore the use of metallogels particularly in active electronic devices.
- Assess the environmental impact and sustainability of metallogels used in active electronic.

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