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Synthesis of surfactant-modified ZIF-8 and its application in drug delivery and tumor therapy

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Abstract

Metal-Organic Frameworks (MOFs) represent a distinct category of crystalline substances consisting of metal ions or clusters interconnected by organic linker molecules. These materials exhibit remarkable porosity, surface area, and adjustable properties, rendering them highly versatile. Metal-organic frameworks (MOFs) are known for their diverse applications, encompassing gas storage, catalysis, drug delivery, and separation processes. This versatility stems from their inherent capability to selectively adsorb and desorb gases, liquids, and various other substances in a regulated fashion. Nevertheless, regulating the microstructure of Metal-Organic Frameworks (MOFs) remains a significant challenge in enhancing drug delivery efficiency and achieving sustained release behavior, thereby posing a hurdle for their clinical application. This paper presents a methodology for synthesizing Surfactant-Modified Zeolitic Imidazolate Framework-8 nanoparticles (SSM-ZIF-8) with varying microstructures. The method involves the utilization of different surfactants to alter the ZIF-8 material. The nanoparticles of ZIF-8 that have been modified with surfactants exhibit a greater Specific Surface Area (SSA) and larger Total Micropore Volumes (TMV) than the unmodified ZIF-8. This enhanced characteristic allows for a more efficient Doxorubicin (DOX) loading onto the drug carriers, leading to a regulated and long-lasting absorption of the drug. The exceptional degradation performance exhibited by ZIF-8 Nanoparticles (NP) enhances the metabolic processes of drug carriers. The porous architecture of the modified ZIF-8, in conjunction with surfactant improvements, facilitates effective drug encapsulation and regulated drug release in the field of drug delivery. This has the potential to enhance therapeutic efficacy while mitigating adverse effects. In the tumor therapy framework, utilizing this modified material holds the potential for targeted delivery of anticancer drugs to tumor sites, thereby optimizing the effectiveness of treatment and reducing systemic exposure. Consequently, this approach can potentially enhance the prospects for cancer therapy. The research comprises both the synthesis of the element and its possible utilization in the medical domain.

Key Words: metal-organic frameworks, ZIF-8, tumor therapy, surfactant, doxorubicin, drug delivery.

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INTRODUCTION

Significant progress has been made in the pharmaceutical sector over the recent decades. The increasing prevalence of diseases has led to the emergence of novel pharmaceuticals and diverse approaches to drug administration. Drug Delivery Systems (DDS) have developed as capable applications in human healthcare. A DDS encompasses either a mixture or a miniature device designed to contain the therapeutic substance intended for administration into the body to enhance security, efficiency, and reliability. These systems have given rise to the traditional notion of the "magic bullet" [1]. The primary research area of interest pertains to carrier-based DDS, which offers the added benefit of safeguarding drug molecules while enhancing the bioavailability of medications [2]. Nanoparticle-based treatments have garnered growing attention in recent years.

Nano-carriers have been found to enhance drug constancy and competence, reduce drug intake, lower harmfulness, and enable precise drug plasma levels [3]. The nano-carriers that have received the most attention in research are micelles, polymeric NPs, liposomes, hydrogels, Solid Lipid Nanoparticles (SLNs), micro sponges, dendrimers, nanosponges, cyclodextrins, and solids with pores like zeolites [4]. Every nanoscale drug carrier possesses both advantages and disadvantages. Persistent endeavors in this trajectory culminated in the emergence of a novel hybrid substance known as MOF, which holds the potential to overcome the challenges associated with current carrier systems.

MOFs are a class of crystalline porous solids that exhibit 2D or 3D structures. The structure of MOF is characterized by the presence of "nodes" and linkers, which are

organic ligands. The nodes function as connection points, while the linkers act as linking molecules, forming a two- or three-dimensional framework [5]. MOFs are alternatively referred to as porous coordination polymers or NCPs.

MOFs exhibit an extremely high porosity level, up to 90% of their volume being free space. The pore windows of these materials range from 5 to 25 Å. MOFs also possess remarkably large surface extents extending beyond 6200 m2/g [6]. These materials exhibit reduced density and demonstrate significant physical and thermal resilience. Therefore, MOFs have emerged as promising contenders for applications in diverse fields, including detection, ion-conducting pores, DDS, catalytic processes, gas storage, and chemical-based separations [7]. MOFs have garnered significant interest in DDS due to their exceptional properties and potential applications. For an MOF to serve as an effective drug delivery system, the MOF must exhibit biocompatibility, non-toxicity, stability, and a specific level of chemical instability. The Materials of Institut Lavoisier (MIL) were the initial group of MOFs identified as promising for DDS use [8].

This study entails the development of a modified variant of ZIF-8, a crystalline material, via a synthesis procedure that integrates surfactants. The present study investigates the potential applications of the modified ZIF-8 material in DDS and tumor therapy. The porous architecture of the modified ZIF-8, in conjunction with surfactant enhancements, facilitates effective drug encapsulation and regulated drug release in the field of drug delivery. This capability has the potential to enhance therapeutic efficacy and mitigate adverse effects. In the context of tumor therapy, utilizing this modified material holds the potential for targeted delivery of anticancer drugs to tumor sites, thereby optimizing the effectiveness of treatment and reducing systemic exposure. Consequently, this approach can potentially improve the outlook for cancer therapy. This study encompasses both the synthesis of the material and its potential applications within the medical domain.

RELATED WORKS

Recently, materials science has made advances in biomedical materials. MOFs are popular due to their porosity, customization, and adaptability. ZIF-8 receives attention in MOFs for its crystalline structure and versatility, making it a promising candidate for various applications. This literature review discusses surfactant-modified ZIF-8 synthesis, drug delivery, and tumor therapy applications. Surfactants can be used to customize ZIF-8 and improve its efficacy in regulated therapeutic agent delivery. In light of the importance of accurate drug administration and focused, therapeutic interventions in modern medicine, this survey aims to provide insight into current research in this dynamic domain.

Pourmadadi, Aslani, and Abdouss (2023) propose synthesizing and characterizing a carboxymethylcellulose-gelatin hydrogel with ZIF-8 MOFs. This hydrogel delivers anticancer drugs continuously [9]. The hydrogel was synthesized, and the drug was added to the ZIF-8 system. Hydrogel structural properties and drug release kinetics were characterized during implementation. The output showed drug release kinetics and encapsulation efficiency. This method allows sustained drug release, which may improve cancer treatment. However, drawbacks must be considered. These may include complex synthesis and DDS compatibility issues with biological entities.

Abdelhamid (2021) studied the biointerface formed by ZIF-8 and biomolecules and its potential applications. This study examines ZIF-8's interactions with various biomolecules [10]. Implementation included experimental studies and interaction characterization. The values revealed biointerface properties and applications. Studying ZIF-8 in biointeractions may improve understanding. This research may have different applications and practical implications in different contexts.

In 2023, Liu et al. developed a DDS using ZIF-8 modified with Dioscorea opposite Thunb Polysaccharide for glucose-triggered release. As described in [11], this study prepared modified ZIF-8 and examined its glucose response. The study measured trans-N-p-Coumaroyltyramine liberation in vitro and in vivo. The observed values correlated with release patterns and therapeutic effects. Glucose-triggered drug delivery targets specific body parts. Drawbacks include meticulous glucose regulation and the need to consider patient factors.

Gao et al. (2020) created folic acid-decorated hollow ZIF-8/Au/CuS nanocomposites to improve and selectively target anticancer therapy [12]. The method included nanocomposite synthesis and characterization. The interventions were tested in vitro and in vivo for anticancer effects. The output values included therapeutic effects and targeting specificity. Benefits include improved anticancer therapy selectivity, while drawbacks may include nanocomposites' complexity.

Abdelhamid (2021) analyzed ZIF-8, focusing on its potential applications in biomedicine [13]. This study reviewed the literature and examined ZIF-8's biomedical applications. The implementation phase involved summarizing and synthesizing existing knowledge. The output values illuminated ZIF-8's many biomedical uses. This method gives a thorough overview of the topic. Be aware that the review may lack empirical evidence.

Zeyni, Karimi, and Namazi (2023) used magnetic hydroxyapatite to surface the PEGylate ZIF-8 metal-organic framework to improve its anticancer drug delivery potential. The study modified ZIF-8 and examined its drug delivery [14]. In vitro and drug release kinetics studies were conducted. The output included magnetic targeting and drug release profiles. This method improves drug delivery regulation. However, the complexity of the modification procedure may have drawbacks.

Xu et al. (2020) synthesized surfactant-modified ZIF-8 materials to control microstructures. They then examined drug loading and sustained release. This study synthesized modified ZIF-8 and assessed its drug encapsulation and release [15]. The execution included empirical studies on drug loading and release kinetics. The output values were drug loading and release data. Adjusting drug release rates is a benefit of this method. However, regulating microstructure complexity may be difficult.

Wang et al. (2020) synthesized and modified ZIF-8 for DDS and tumor therapy [16]. The study synthesized and tested modified ZIF-8 for drug delivery. The study evaluated therapeutic efficacy in vitro and in vivo. Drug delivery efficacy and tumor therapy outcomes were output values. Improved drug administration and therapeutic efficacy are the benefits, but translating these discoveries into clinical practice may be difficult.

In conclusion, surfactant-modified ZIF-8 research is exciting and promising for biomedical science. ZIF-8's multifunctionality and surfactant modification offer new drug delivery and tumor therapy options. Several studies have explained the synthesis methods and examined the properties of modified materials. However, many challenges and opportunities remain. The challenges include optimizing drug loading capacity, improving drug release kinetics, and creating multifunctional systems to improve therapeutic outcomes. This research domain promises improved cancer treatment and spans focused medication administration, reshaping pharmaceutical technology.

MATERIALS AND METHODS

The synthesis methods commonly employed for producing MOF materials encompass the traditional solution reaction method, hydrothermal (solvothermal) method (including microwave-assisted heating), diffusion method, and mechanical grinding. The traditional reaction of the solution method combines a metal salt and an organic bridging ligand in a designated solvent, such as water or an organic solvent. The pH value may be adjusted if required, and the mixture is then subjected to stirring or allowed to stand in an open system. The temperature is typically maintained below 100°C, and as the reaction progresses, the reaction products are precipitated either through a decrease in temperature or the evaporation of the solvent.

The hydrothermal method, also known as the solvothermal method, involves directly combining a metal salt and an organic co-ligand in a designated solvent, such as water or an organic solvent. This mixture is subsequently placed within a sealed reactor. The reactants undergo a chemical reaction within the system due to the internal pressure generated by the system due to heating. The diffusion method involves the dissolution of two reactants in either the similar or dissimilar solvents. The objective is to carefully manipulate the reaction conditions to facilitate the contact between the two fluid phases containing the reactants. This contact occurs through diffusion at the boundary or within a designated medium, ultimately forming the desired product. The term "mechanical grinding" pertains to combining and

grinding two compact stages of metal composites and organic bridging ligands using a ball mill to initiate a reaction that yields the desired end product. In certain instances, it becomes imperative to modify synthesized MOFs to incorporate essential functional groups that confer specific properties upon them. In a precise sense, the modification of MOFs pertains to the chemical alteration of the metal center and the organic bridging ligand after eliminating the easily detachable end-capping ligand from the metal center. This modification can involve processes such as oxidation following synthesis, substitution of the easily detachable end-capping ligand with ligands possessing robust coordination capabilities, or the immersion of MOF crystals to facilitate the exchange of metal ions or organic ligands.

SSM-ZIF-8

Recent research has revealed that ZIF-8 exhibits a substantial surface area and TPV, leading to a significant adsorption capacity and facilitating the adsorption of small molecular drugs. The structural composition of ZIF-8 consists entirely of ZnN4 tetrahedra. The truncated octahedral structure of the methylimidazolium ester is created through the amalgamation of Zn ions and N atoms, forming the structural unit. ZIF-8 exhibits a BET-SSA of 1400 m2/g and high thermal stability with a temperature threshold of 420°C. In contrast to conventional zeolite, ZIF-8 displays extraordinary thermal and hydrothermal solidity. ZIF-8 demonstrates the ability to regulate drug release and augment the cytotoxicity of drugs towards tumor tissue. Furthermore, it should be noted that ZIF-8 exhibits favorable biological compatibility and decomposition properties. These outstanding characteristics produce ZIF-8 a perfect choice in MOF's DDS.

In this study, it has been investigated the impact of surfactants on the production of ZIF-8 NPs, examining their ability to modify the microstructures of the particles. Additionally, we analyzed the behavior of drug loading and release in these modified nanoparticles. The nanoparticles of ZIF-8, which were modified with surfactants, were synthesized using the liquid-phase diffusion method. To modify the microstructure of ZIF-8, the synthesis procedure involved the addition of three types of surfactants: an amphoteric surfactant known as 2-(dodecyldimethylammonio)acetate (DA), a cationic surfactant called exadecyltrimethylammonium bromide (CTAB), and an anionic surfactant referred to as sodium dodecylbenzenesulfonate (SDBS). The materials were denoted as ZIF-8(DA), ZIF-8(C, S), and ZIF-8(CTAB) when incorporating DA, CTAB + SDBS, and CTAB, respectively.

Additionally, a relative ZIF-8 sample, denoted as ZIF-8(O), has been synthesized in the absence of surfactant. This was done to demonstrate the impact of surfactant addition on the material's structure and performance. It was observed that all three SSM-ZIF-8 variants exhibited higher SSA and TMV when compared to ZIF-8(O).

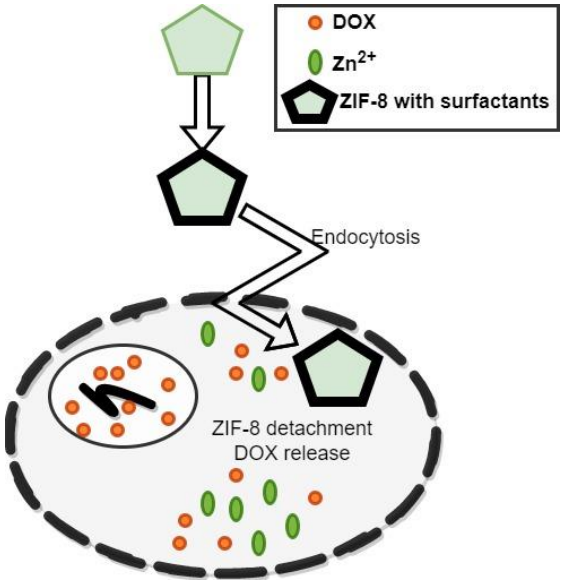


Fig.1. Schematic illustration depicting SSM-ZIF-8.

Among the various options considered, it is noteworthy that ZIF-8(C, S) exhibits the most significant SSA, surpassing 2500 m2/g. Additionally, this material showcases the largest total mesoporous volume, measuring 0.945 cm3/g, and the greatest overall microporous volume, measuring 0.74 cm3/g. The DOX- lading rate and prolonged release behavior of three types of surfactants were investigated in our study. The rate of DOX loading in SSM-ZIF-8 is significantly higher than ZIF-8(O), and the SSM-ZIF-8/DOX also exhibits a more favorable pH stimuli-response release behavior. The ZIF-8/DOX nano drugs can issue DOX in response to changes in pH levels. This release mechanism enables the effective targeting and destruction of cancer cells. The

internalization of these nano drugs into the cells occurs through endocytosis, as depicted in Fig. 1.

A solution of Zn(NO3)2.6H2O (0.00090 mol) was fully liquified in a volume of 12 ml of methanol. Subsequently, dropwise was added to a solution containing 2-methylimidazole (0.00690 mol) and DA (0.00041 mol) in a volume of 13 ml of methanol. The mix was agitated at ambient heat level for one day. Subsequently, the product underwent centrifugation for 15 minutes at a rotational speed of 15,000 revolutions per minute (rpm), followed by three successive washes with methanol. Subsequently, the product was subjected to overnight drying at a temperature of 65°C. The obtained specimen was documented as ZIF-8(DA). The synthetic procedures employed for the production of ZIF-8(C, S) and ZIF-8(CTAB) are analogous to those utilized for the synthesis of ZIF-8(DA), with the exception that 0.00003 mol SDBS replaces the substitution of 0.00036 mol CTAB in the former case, and 0.00036 mol DA substitutes 0.00036 mol CTAB in the latter case.

Encapsulation of drugs and the subsequent controlled release of their active components

Drug encapsulation and controlled release are among the key applications of surfactant-modified ZIF-8. The porous nature of ZIF-8 enables the encapsulation of various therapeutic agents, such as small molecules, proteins, and nucleic acids. The process of surfactant modification improves the loading capacity and offers possibilities for surface functionalization, thereby allowing for precise regulation of drug release. Controlled drug release has been accomplished by manipulating the hydrophobicity of the surfactant. This alteration subsequently affects the interaction between the carrier ZIF-8 and the drug encapsulated. This facilitates the customization of drug delivery profiles, encompassing sustained and triggered release in response to particular stimuli.

Tumor Targeting and Therapy

The utilization of surfactant-modified ZIF-8 has demonstrated significant promise in enhancing the efficacy of tumor treatment. The modified ZIF-8 can be intentionally designed to demonstrate pH-responsive characteristics, capitalizing on the acidic conditions in tumor tissues. Consequently, the ZIF-8 material loaded with drugs exhibits a selective release of therapeutic agents specifically in the tumor's proximity, thereby reducing off-target effects. Additionally, introducing targeting ligands, such as folic acid or antibodies, through surface modification augments the selectivity of drug delivery to cancer cells, thereby minimizing the potential adverse effects on normal tissues.

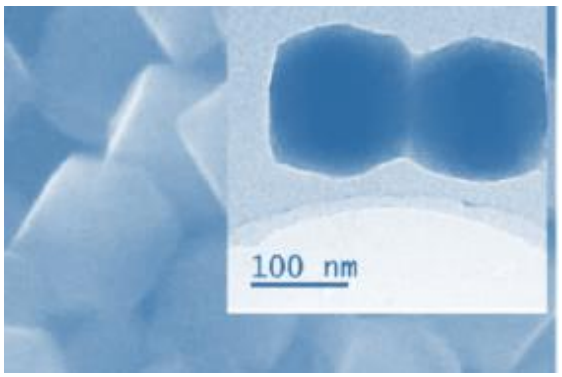
The efficacy of surfactant-modified ZIF-8 in augmenting the therapeutic efficacy of different anticancer agents has been demonstrated through in vivo studies. The materials mentioned above have been utilized for administering chemotherapeutic drugs, photodynamic therapy agents, and gene therapies. Surfactant-modified ZIF-8 exhibits favorable attributes such as sustained release and enhanced targeting capabilities, rendering it a highly promising contender for personalized cancer therapy.

Although surfactant-modified ZIF-8 exhibits significant potential, various obstacles still need to be addressed. Additional research and development are necessary to facilitate the expansion of production and the application of these materials in clinical environments. Furthermore, it is imperative to thoroughly investigate the long-term biocompatibility and potential toxicity concerns to guarantee patient safety. In summary, SSM-ZIF-8 demonstrates a noteworthy drug delivery and tumor therapy progression. The versatility, tunability, and selectivity attributes render it an invaluable resource in pursuing enhanced and focused cancer therapies. As the study in this field grows, it is expected that there will be added advancements in synthesizing and utilizing surfactant-modified ZIF-8, leading to enhanced patient outcomes in cancer therapy.

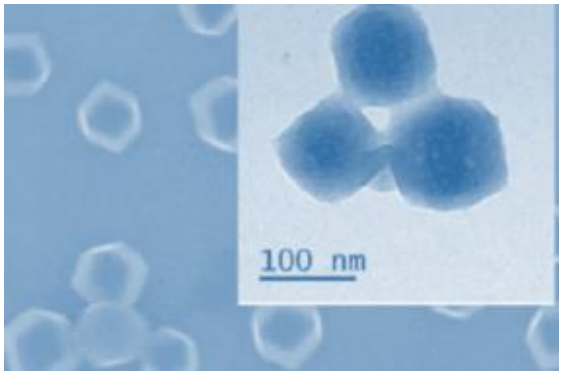
RESULTS AND DISCUSSION

The FEI Nova NanoSEM 450 microscope was utilized to conduct Scanning Electron Microscopy (SEM) analyses. The acquisition of Transmission Electron Microscope (TEM) images was performed using an FEI Tecnai G2 F30 TEM microscope with an operating voltage of 200 kV. The X-ray Diffraction (XRD) pattern was observed using the SHIMADZU XRD-7500S diffractometer, with a scanning speed of 0.03° s-1. The

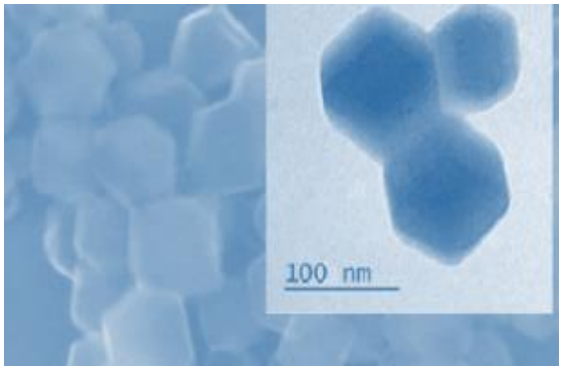
range of 2θ values recorded spanned from 1° to 50°.



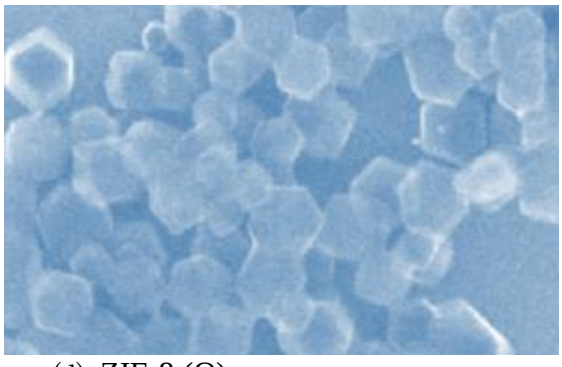
(a) ZIF-8 (DA)



(b) ZIF-8 (C,S)



(c) ZIF-8 (CTAB)



(d) ZIF-8 (O)

Fig.2. SEM images and the TEM images (insets) of the proposed SSM-ZIF-8.

Fig. 2 displays the SEM images of the proposed SSM-ZIF-8, with three surfactant modified samples. The geomorphology of the three SSM samples exhibits a consistent rhombic dodecahedron shape, which is identical to that of the initial ZIF-8(O) as depicted in Fig. 2d. The particle sizes of three surfactant modified samples are approximately 140-190 nm, 80-110 nm, and 100-140 nm. Various types of surfactants exhibit a discernible influence on the particle size of ZIF-8. The insets depicted in Fig. 2 showcase TEM images of the three distinct types of SSM-ZIF-8. The SSM-ZIF-8 samples demonstrate the presence of rhombic dodecahedron crystals, as observed in the SEM images.

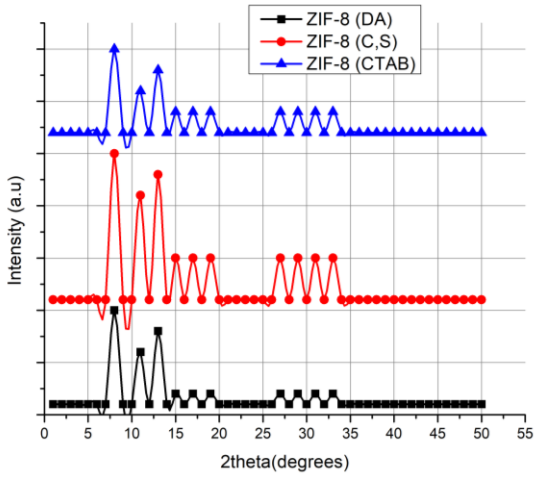


Fig.3. XRD results for the proposed SSM-ZIF-8.

Fig. 3 displays the XRD patterns for the proposed SSM-ZIF-8. All samples of ZIF-8 demonstrate distinct diffraction peaks that align with those documented in previous studies. The intensity of the diffraction peak in ZIF-8(C, S) and ZIF-8(CTAB) is observed to be higher compared to that of ZIF-8(DA), indicating a higher degree of crystallinity in ZIF-8(C, S) and ZIF-8(CTAB) as compared to ZIF-8(DA). The XRD data presented in Fig. 3 illustrate the peak intensities observed at different 2θ (degrees) values for three distinct samples of SSM-ZIF-8. As mentioned above, the peak values serve as indicators of the diffraction pattern observed in each respective sample. It is worth mentioning that in all three samples, there is a consistent observation of peaks at 2θ degrees of 1, 11, and 27. This indicates the existence of comparable crystallographic structures in all three variants of ZIF-8. Nonetheless, discernible discrepancies in the maximum intensity and locations at

elevated 2θ values (8, 10, 25, and 35 degrees) exist among the specimens, potentially indicating disparities in crystallinity or structural alterations within the ZIF-8 substances. Additional examination and juxtaposition of these maximum values can offer valuable insights into the structural discrepancies and the caliber of the synthesized ZIF-8 specimens, thereby facilitating their characterization and potential utilization.

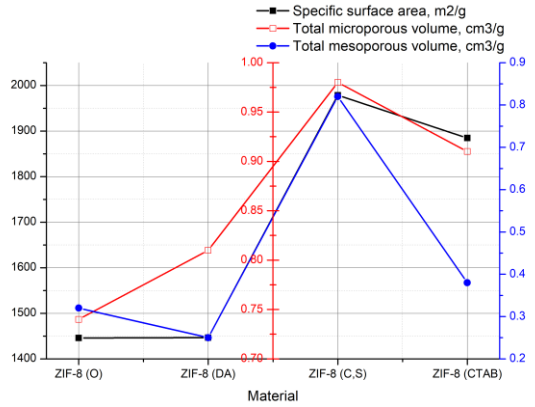


Fig.4. SSA and TPV of the proposed SSM-ZIF-8.

Fig. 4 provides data on the SSA and TPV of three SSM-ZIF-8 specimens. The values play a crucial role in determining the porosity and adsorption properties of the materials. ZIF-8 (O) and ZIF-8 (DA) demonstrate comparable SSA, measuring 1445.8 m2/g and 1446.5 m2/g, respectively. In contrast, ZIF-8 (DA) exhibits a marginally greater overall microporous volume (0.81 cm3/g) in comparison to ZIF-8 (O) (0.74 cm3/g). On the other hand, ZIF-8 (C, S) and ZIF-8 (CTAB) exhibit considerably elevated SSA of 1978.6 m2/g and 1884.6 m2/g, respectively, thereby implying a greater extent of available surface areas for adsorption. In addition, it is worth noting that ZIF-8 (C, S) demonstrates a significantly increased total mesoporous volume of 0.82 cm3/g, indicating the existence of larger pores in the material. The observed values indicate disparities in porosity and adsorption capacities across the various ZIF-8 materials. These are crucial considerations in assessing their potential applications, including catalysis, and drug delivery for cancer therapy.

CONCLUSION

This study introduces a new methodology for producing Surfactant-Modified Zeolitic Imidazolate Framework-8 nanoparticles (SSM-ZIF-8) featuring diverse microstructures. The methodology entails the application of various surfactants for the modification of the ZIF-8 substance. The ZIF-8 nanoparticles, which have undergone surfactant modification, demonstrate enhanced SSA and TMV compared to the unmodified ZIF-8 nanoparticles. This augmented attribute facilitates a more effective loading of DOX onto the drug carriers, resulting in a controlled and enduring DDS. Within the context of tumor therapy, the application of this modified substance exhibits promise for the precise transportation of anticancer medications to specific tumor locations, consequently enhancing the efficacy of treatment and minimizing overall systemic exposure. The XRD data presented in this study depict the intensities of peaks observed at various 2θ (degrees) values for three distinct samples of ZIF-8. ZIF-8 (O) and ZIF-8 (DA) exhibit similar SSAs, with measurements of 1445.8 m2/g and 1446.5 m2/g, respectively. On the other hand, ZIF-8 (DA) demonstrates a slightly higher TMV (0.81 cm3/g) when compared to ZIF-8 (O) (0.74 cm3/g). In contrast, ZIF-8 (C, S) and ZIF-8 (CTAB) demonstrate significantly increased SSAs of 1978.6 m2/g and 1884.6 m2/g, respectively. This suggests a larger amount of surface area accessible for the adsorption process.

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