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*REVIEW ARTICLE*

# APPLICATION OF $CO_3(BTC)_2{\cdot}12H_2O$ AS A METAL-ORGANIC FRAMEWORK IN CONTROLLED UPTAKE-RELEASE OF OXALIPLATIN

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**ABSTRACT**

Metal-organic frameworks (MOFs) are a class of Coordination polymer of metal ions or clusters coordinated to organic ligands to form which has nanometer cavities. In the category porous materials, they have domination of great surface areas, tunable pore size and structure, tunable combination and functionalized surface area and porous size, ability to encapsulate and post-synthesis which enables them unique advantages important point concerning the possible use of such functional materials for applications in adsorption and release of drug and active pharmaceutical ingredient. By examining the patterns resulting from N2 adsorption/desorption isotherms and thermogravimetric analysis, it is possible to understand the thermal stability and the appropriate volume of holes available to the molecule for the presence of guest molecules. In this report, we study uptake and release properties of Oxaliplatin from nano Co (II)-MOF in comparison with bulk Co(II)-MOF. To explore the absorption ability of the Co (II)-MOF to Oxaliplatin, a fresh sample of Co-BTC metal-organic framework was immersed in an aqueous solution of Oxaliplatin and was monitored in real-time with HPLC spectroscopy. Results show that the adsorbed quantity of Oxaliplatin over Nano Co-BTC metal-organic framework is much higher than those over a bulk Co-BTC. According to the Oxaliplatin adsorption times, the time is 22 h, the uptake of Oxaliplatin (99.94%) on Nano Co-BTC is remarkable and clarity exceeds that of bulk Co-BTC and activated carbon in the aqueous solution of Oxaliplatin. When these interactions disappear with increasing Oxaliplatin extrusion, labor in the second stage is largely governed by a free-diffusion process, and full Oxaliplatin release from Nano Co-BTC takes more than 5 days to reach equilibrium.

**KEYWORDS**

Anticancer, Oxaliplatin, Co-BTC, HPLC Spectroscopy

## 1. INTRODUCTION

Designing and selecting a practical source for the safe transfer of drugs to the target tissues for the effect of treatment and not damaging healthy tissues makes the need for targeted drug delivery more important than ever. Due to the biocompatibility and low toxicity of MOFs compounds, as well as the appropriate chemical and surface properties of these compounds, they have potential applications in targeted drug delivery (Amini et al., 2020; Cao et al., 2020; Jarai, 2020; Terzopoulou, 2020; Wang, 2020). Metal-organic frameworks (MOFs) are types of inorganic polymer consisting of metal ions by complex formation coordinated to organic ligands to form 2D & 3D dimensional structures (Jamal and Alavinia, 2022; Xia, 2021). Metal Organic Frameworks & Covalent Organic Frameworks are a subset of three-dimensional porous polymers, the existence of holes with high access level and excellent stability are the unique features of this group of compounds (Kouhdareh, 2022; Cao, 2021; Zhou, 2021). Generally, MOFs is a coordination complex reticular consist of an organic ligand that has nanometer-sized holes. By changing the type of ligand, it is possible to control the size of these holes (Babapour, 2022; Wang, 2022; Wu, 2022).

Metal-Organic Frameworks (MOFs) are very important because, among other reasons, that is, The presence of nanometer cavities in them has led to applications such as adsorbents, Nanoreactors, targeted drug delivery, Nano catalysts, as well as many other applications (Ricco, 2013; Li, 1999; Yoon et al., 2012; Suh, 2012; Britt, 2008; Lee, 2009; Li et al., 2012; Torchilin, 2005; Yaghi et al., 1995). Therefore, due to having a very wide active surface and also the ability to connect different functional groups to MOFs, targeting the target tissue and ease of drug transfer are some of the benefits of using MOFs in the targeted drug delivery field (Gautam, 2022; Safdar et al., 2022). Drug delivery systems refer to approaches that are one of the most promising applications for storage systems, and technologies involved in areas of human safety and health care and represent an ever-evolving field for transporting a pharmaceutical compound at specific points in human tissues.

This report addresses MOFs as a new way to deliver drugs and demonstrates their ability to deliver a variety of therapeutic drugs, including generic and anticancer drugs (Alves, 2021; Cui, 2021; Kiadeh, 2021; Lawson et al., 2021; Liu, 2021; Osterrieth and Fairen-Jimenez, 2021; Yo and Lee, 2006). It also provides the details required for MOFs for stability, particle size, and surface modification applications. Taking advantage of the unique advantages of MOF materials, efficient delivery of various drugs in this category of mesoporous materials has been achieved

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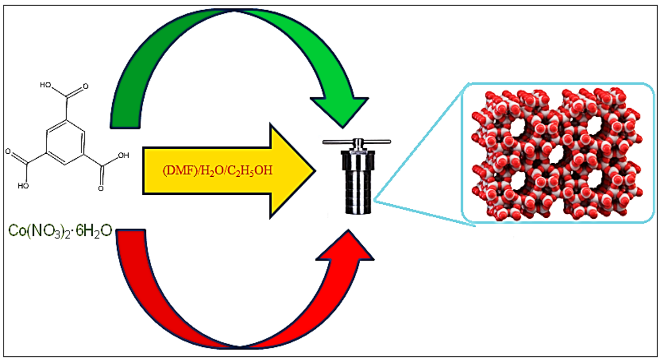
(Wang, 2021; Yang, 2021; Zhang, 2021; Zhu et al., 2021; Gupta et al., 2012). One of the anti-cancer drugs that for certain physiological reasons confirms the need to use drug carriers for is Oxaliplatin. Oxaliplatin is a platinum-based chemotherapy drug in the same family as cisplatin and carboplatin it is Often it is used together with other anticancer drugs (fluorouracil and leucovorin). It is given by injection into a vein (Grothey and Goldberg, 2004; Arbaeen, 2019).

Oxaliplatin is the research in this project produced by Arasto Pharmaceutical Chemicals company and all the steps of synthesis and identification of this product have been done in this company. Co-BTC MOF in comparison with activated carbon. Co-BTC is neutral coordination with a lot of cavities and voids, which makes this material have great potential for many practical applications related to the structure. To identify and measure the purity of Oxaliplatin in the environment in the uptake and release process, Pharmacopeia USP 43 monograph was used using an HPLC device (USP, 2008). Compared to organic porous materials (e.g., nano-carbon hybrid materials) or similar inorganic compounds (e.g., zeolites, silica), MOFs as potential drug bearers have the appendix unique advantages. Therefore, in this study, we investigated an effective strategy for the uptake and delivery of Oxaliplatin anti-cancer drug in Co-BTC and safe, effective, and efficient release in a simulation environment. The potential of these compounds in targeted drug delivery, confirms the need to use these compounds as a suitable drug carrier.

## 2. Experiments

### 2.1 Materials Preparation

BTC (1,3,5-benzenetricarboxylic acid) (0.005 mol,1.05 g) and Co $(NO_3)_2{\cdot}6H_2O$ (0.01 mol, 2.908 g) were dissolved in 60 mL solvent (N, N-dimethylformamide (DMF): $H_2O$:$C_2H_5OH$=1:1:1, ν/ν). Stir the solution for about 15 minutes at room temperature until completely dissolved and placed in a stainless-steel vessel, which was sealed and placed in a programmable furnace. The mixture was heated to 95 °C at 5 °C/min and held at that temperature for 24 h, Then slowly lowering the temperature and reaching room temperature. The resulting crystals were washed twice with 30 ml of distilled water and then 30 ml of ethanol and then air-dried to give 1.33 g of nano [$Co_3(BTC)_2{\cdot}12H_2O$] and finally the samples were calcined at 300 °C for 4 h (Tan, 2015).




**Scheme 1:** Design Synthesis Nano $Co_3(BTC)_2{\cdot}12H_2O$ metal-organic framework

### 2.2 Synthesis of Bulk Co-BTC MOF

BTC (1,3,5-benzenetricarboxylic acid) (0.005 mol,1.05 g) and Co $(NO_3)_2{\bullet}6H_2O$ (0.01 mol, 2.908 g) were dissolved in 60 mL solvent (N, N-dimethylformamide (DMF): $H_2O$:$C_2H_5OH$=1:1:1, ν/ν). The mixture of the solution was sonicated for 5 minutes and the mixture then under continuous stirring was heated at a fixed temperature of 100 ° C for 24 h. The bulk product was obtained in a 73.4% yield.

### 2.3 Materials Characterization

To investigate the particle morphology, size, and elemental mapping of this sample, a scanning electron microscope (SEM) (SEM FEI MIRA3 TESCAN scanning microscope), equipped with the energy-dispersive X-ray spectroscopy (EDS) was performed. The IR spectra were recorded on a Thermo model Bruker spectrometer (KBr). X-ray diffraction (XRD) images were obtained from a Bruker XRD D8 Advance instrument with Cu Kα radiation (l = 0.15418 nm). To study the absorption and release of the drug from the High-performance liquid chromatography (HPLC) of the brand Knauer Azura model D-14163 was used. The was used to investigate the electrochemical properties of the Metrohm Autolab device model PGSTAT302N.

### 2.4 $Co_3(BTC)_2{\cdot}12H_2O$ Metal-Organic Framework Discussion

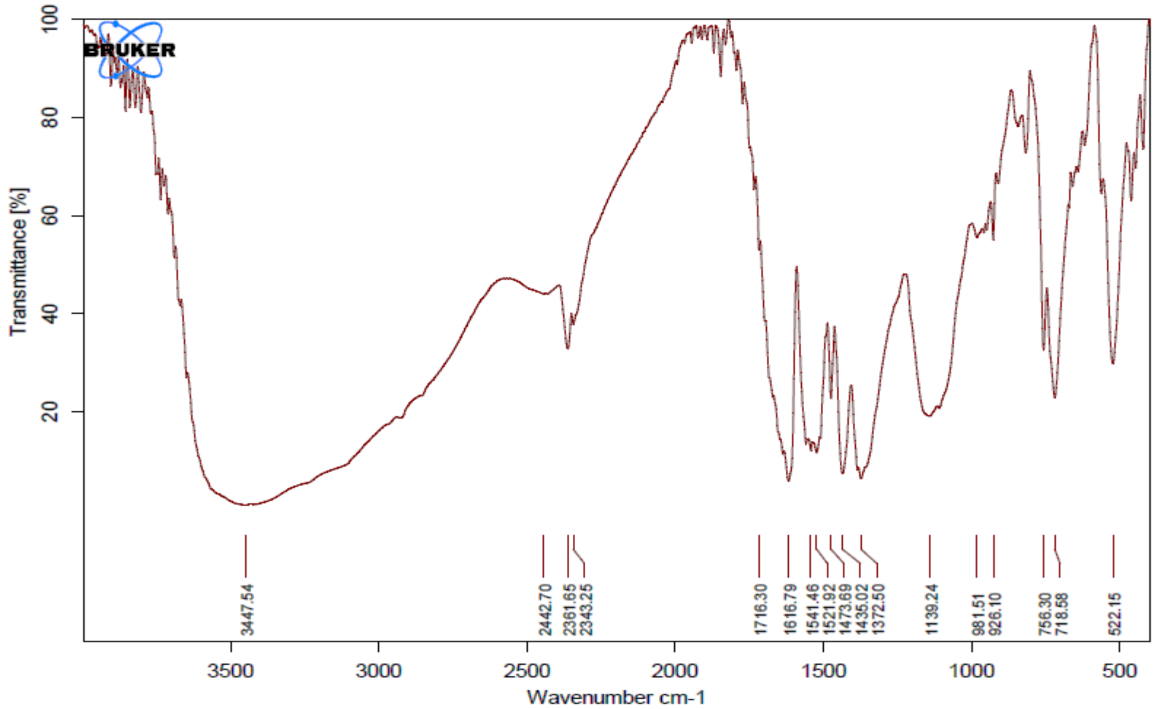



**Figure 1:** FTIR spectra of Nano Co-BTC

The successful synthesis of the $Co_3(BTC)_2{\cdot}12H_2O$ metal-organic framework can be inferred from FT-IR techniques (Figure 1).

The morphology, shape, and size of the prepared Co-BTC MOF were identified by Field emission scanning electron microscopy (SEM) of Co-BTC MOF (Figure 2) confirmed the successful preparation of the Co-BTC MOF Which were synthesized by sonicated and thermal methods. The structural and crystalline patterns of Nano Co-BTC synthesized by thermal methods are more regular and have larger particle sizes.

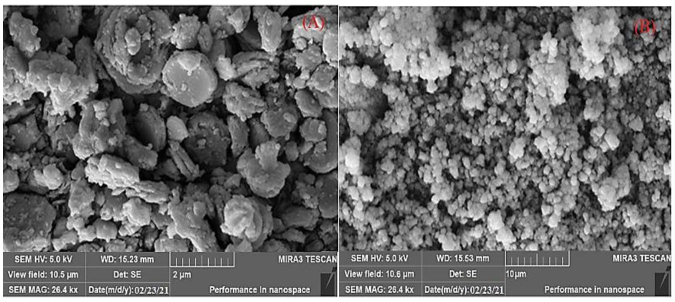



**Figure 2:** Scanning Electron Microscopy (SEM) for(A) Nano Co-BTC, (B) bulk Co-BTC

In Figure 3, which is the result of the XRD analysis, the Co-BTC crystalline phase can be seen. The overall XRD pattern obtained with characteristic peaks (005), (013), (018), (024), (232), (118) and (239) was in good agreement with the researches of previous research teams, and confirmed It corrects the Co-BTC synthesized crystal structure (Peng, 2017; Tian, 2019; Wu, 2019). In the phase synthesized by the crystal method, the observed patterns have more order than the bulk state.

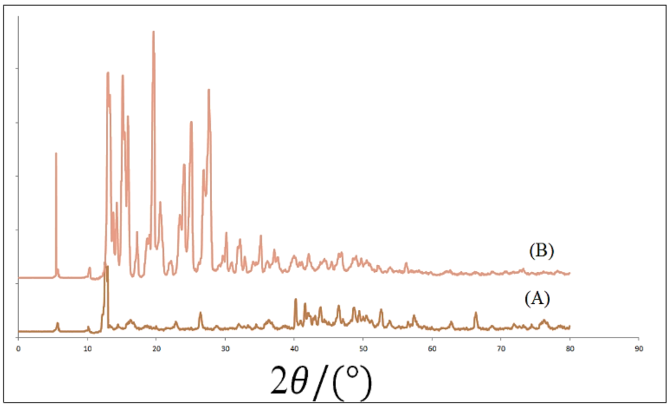



**Figure 3:** Simulated pattern based on single-crystal data of XRPD pattern of (A) bulk Co-BTC (B) Nano Co-BTC

Figure 4 illustrates the TGA of the prepared bulk Co-BTC (A), and Nano Co-BTC (B),. According to Figure 4, a considerable weight loss occurs at 200-400°C for bulk Co-BTC (A), and Nano Co-BTC (B). This weight loss is attributed to the loss of organic moieties. As shown in Figure 4, the thermal stability of both types is similar because the molecular structure and functional groups forming the complex are the same and they differ only in the crystal structure, which has little effect on determining the thermal stability.

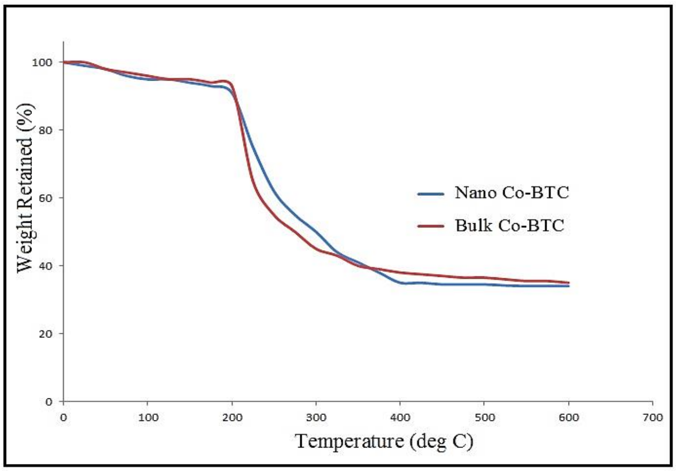
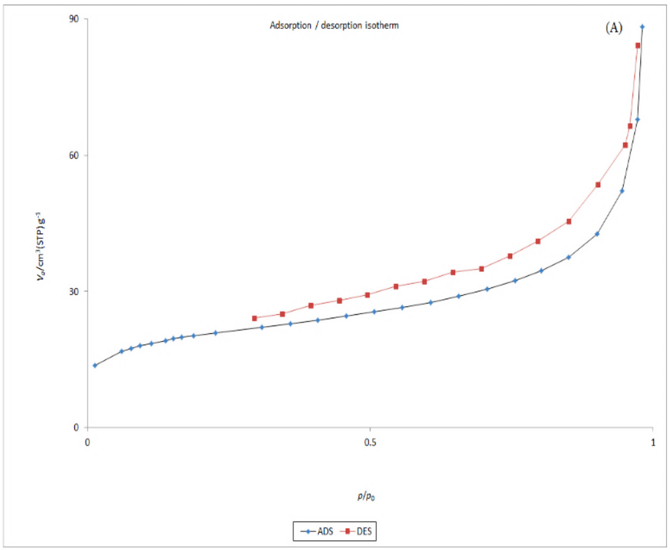



**Figure 4:** TGA plots of (A) bulk Co-BTC (B) Nano Co-BTC

Figure 5 the bulk Co-BTC (A), and Nano Co-BTC (B) porosity was determined via the N2 adsorption/desorption isotherms at 77 K. The results showed a type-H3 hysteresis loop (defined by IUPAC) and a type-IV isotherm (due to its mesoporous substances). According, to the resulting graphs, it can be concluded that the specific surface area of available holes in the crystalline state is more than the bulk type and has a more stable.



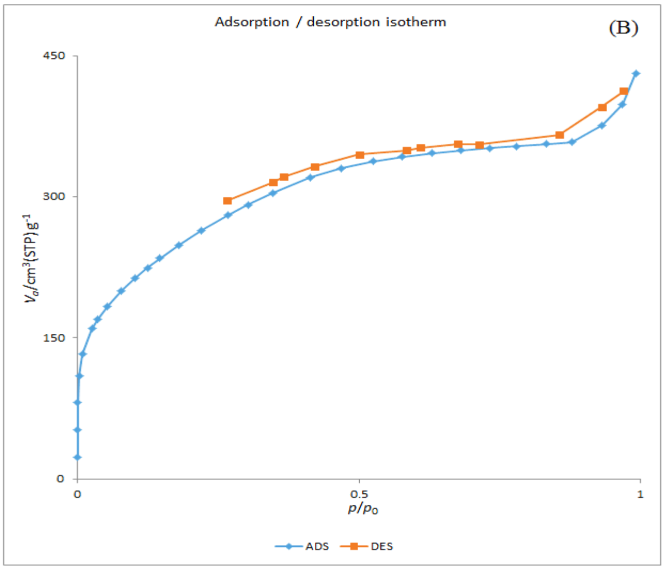
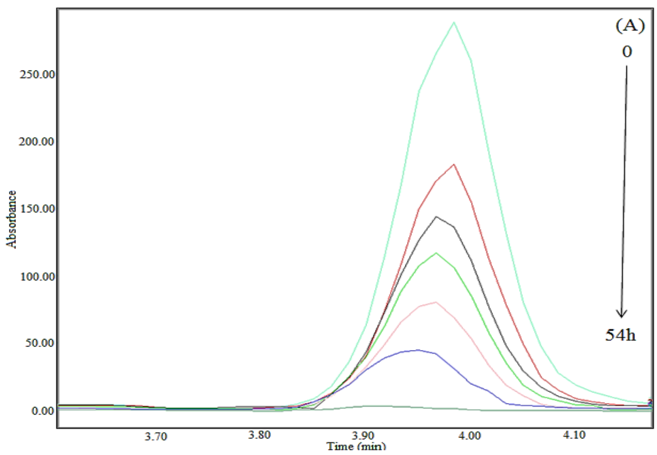




**Figure 5:** $N_2$ adsorption-desorption isotherms A) bulk BTC B) NANO BTC

## 2.5 Adsorption Affinity

For augment of porosity in Co-BTC, we successfully tested its porosity with Oxaliplatin (guest molecules) by suspending it in an aqueous solution of Oxaliplatin. The guest content was estimated by HPLC spectroscopy (Figure 6). The change of intensity and width indicates that the resulting solid Co-BTC retains the host framework crystallinity as Oxaliplatin molecules diffused in (Lee, 2009). To explore the absorption ability of the Co-BTC to Oxaliplatin, a fresh sample of Co-BTC (50.0 mg) was immersed in an aqueous (50.0 mL) solution of Oxaliplatin (0.5 mmol) and were monitored in real-time with HPLC spectroscopy and at the same time, a qualitative study was decreasing trend of Oxaliplatin concentration in solution.



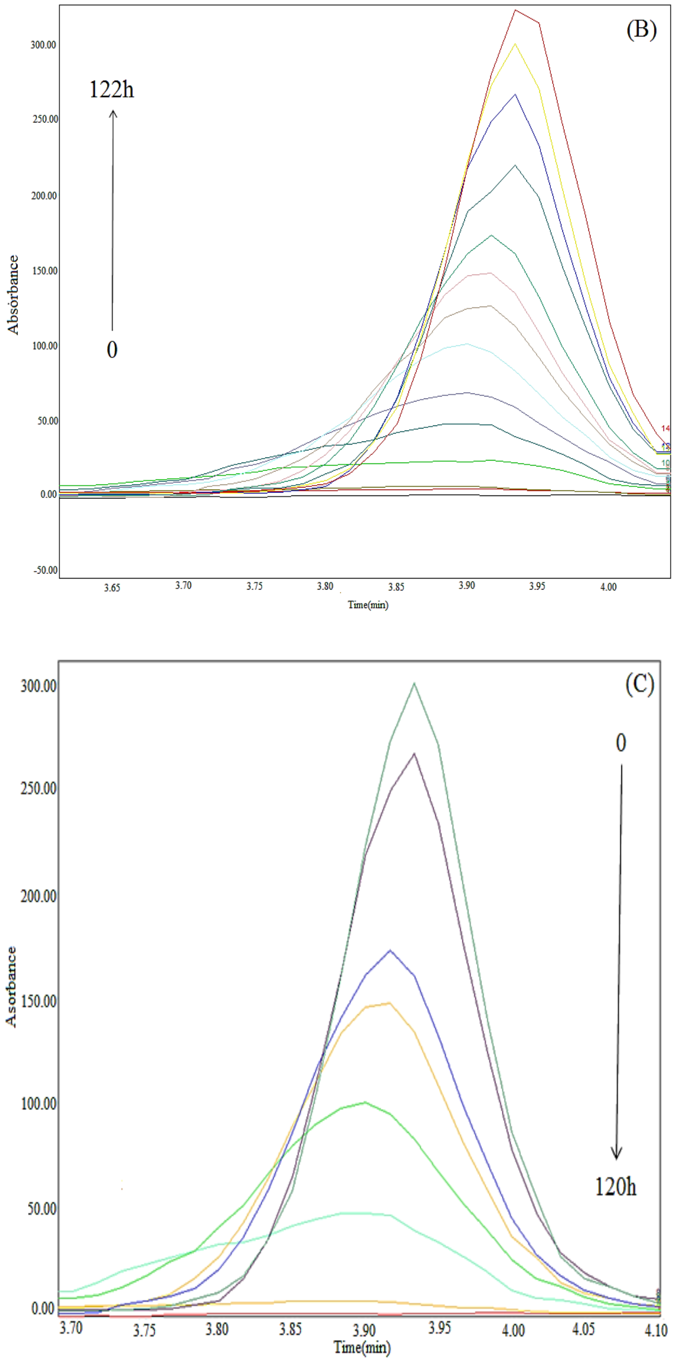
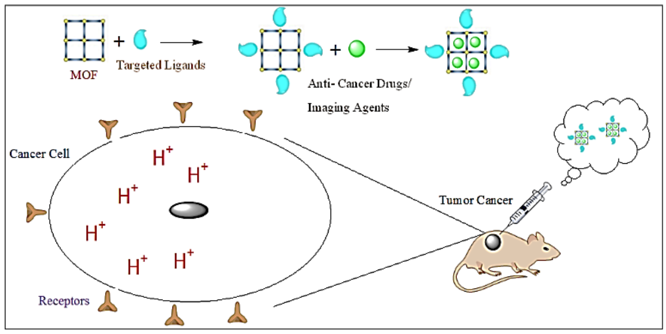



**Figure 6:** Temporal evolution of HPLC absorption spectra for the loading of Oxaliplatin from bulk Co-BTC (A), Nano Co-BTC (B), and activated carbon (C).

Adsorption of Oxaliplatin molecules was a spontaneous and endothermic process that increases the entropy of the guest molecule in the host nanostructure. Increasing the entropy means that the process is desirable. The adsorption of Oxaliplatin by Nano Co-BTC is It is possible that by hydrogen bonding to the structure Co-BTC, and as well as to other Oxaliplatin molecules, as there is no variation in the MOF due to covalent bonding with Oxaliplatin. As the time remained increases, the adsorption capacity of the Co-BTC usually increases. As shown in Figure 5, the adsorbed quantity of Oxaliplatin over Nano Co-BTC is much higher than those over a bulk Co-BTC and activated carbon.

According to the Oxaliplatin adsorption times, the time is 22 h, the uptake of Oxaliplatin (99.94%) on Nano Co-BTC is remarkable and clearly exceeds that of bulk Co-BTC and activated carbon in the aqueous solution of Oxaliplatin. The adsorption of Oxaliplatin in the solution increases linearly over time, a process that is controlled by the adsorption of Oxaliplatin in the host-guest interaction mode. By absorbing anticancer drugs in host nanostructures and their targeted transfer into cancerous tumors after drug release, they cause cancer cell destruction, which is targeted to increase the efficiency of anticancer drugs in cancer-damaged tissues and further protect cells. Healthy takes place nearby.




**Scheme 2:** The process of adsorption and desorption of anti-cancer drugs by Metal-Organic Framework (MOF) for targeted drug delivery

## 2.6 Release Assays

Guest drug molecules adsorbed in MOFs cavities could be quickly and easily removed from the lattices on plunging of guest@ MOFs in organic solvents. To investigate the release kinetics of the drug in the Metal-Organic Framework (MOF) by immersion in ethanol, the release process was recorded and controlled by HPLC spectroscopy. The evolutionary release of Oxaliplatin in ethanol solution was observed by HPLC spectroscopy, which indicates an increase in Oxaliplatin in ethanol-immersed solution over time, and this is influenced by the interaction of host and guest. When these interactions disappear with increasing Oxaliplatin extrusion, labor In the next step is largely governed by a free-diffusion process, and full Oxaliplatin release from Nano Co-BTC takes more than 5 days to reach equilibrium. Two parameters, maximum Oxaliplatin content and its amount indicate that it may be well defined due to a free diffusion flow in the lack of host-guest interactions. seems to be in Nano Co-BTC the length of channel become smaller and the Oxaliplatin molecules are more stable than bulk Co-BTC. Thereupon, the delivery of Oxaliplatin from bulk Co-BTC can be faster than Nano Co-BTC.

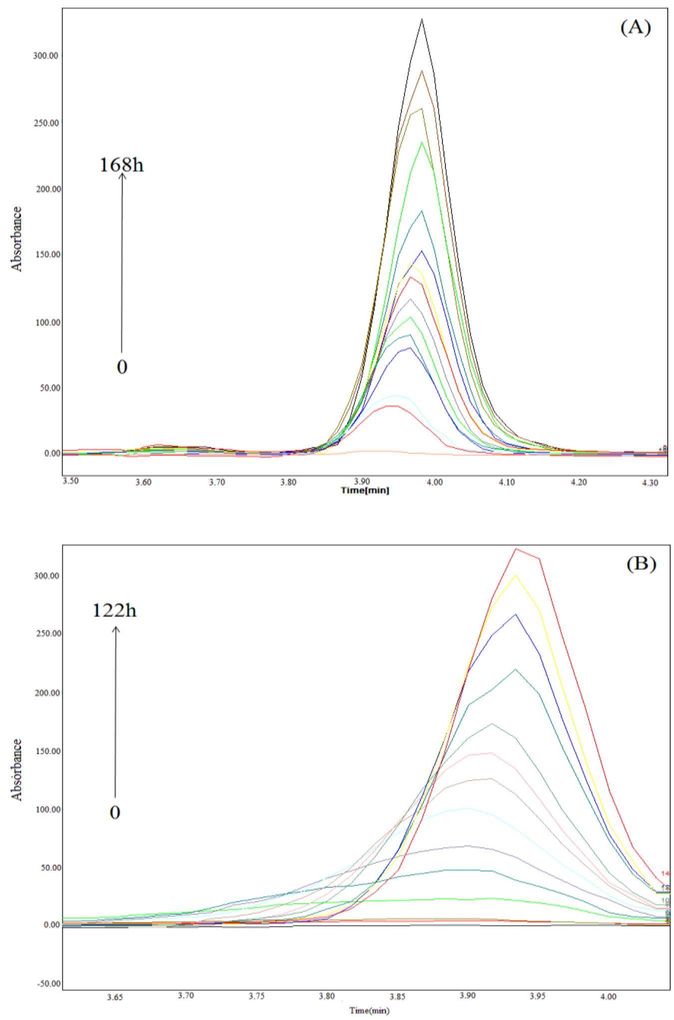
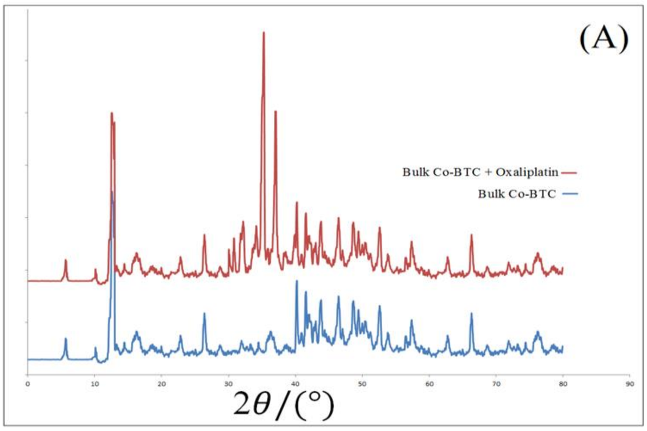



**Figure 7:** Temporal evolution of HPLC spectroscopy absorption spectra for the delivery of Oxaliplatin from compounds Nano Co-BTC (A), and bulk Co-BTC (B).

### 2.7 X-Ray Powder Diffraction (XRPD) and X-Ray Spectroscopy (EDX)

In Figure 8, after the guest molecule (Oxaliplatin) enters, by examining the X-ray powder diffraction (XRPD) spectrum patterns, the presence of the guest molecule can be seen in the structure, which is a proof of the potential power of these nano polymers in absorbing guest molecules.



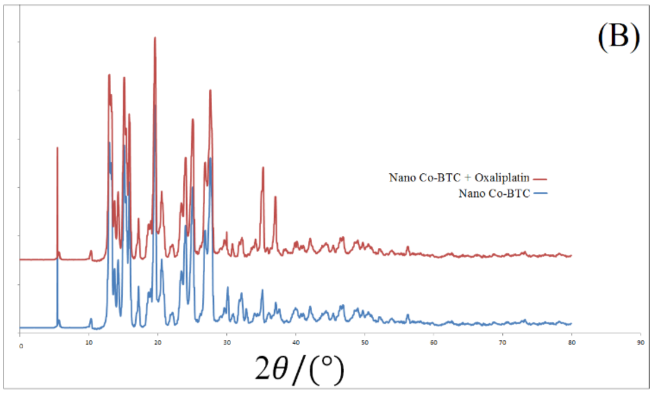




**Figure 8:** Simulated pattern based on single-crystal data of XRPD pattern of Bulk Co-BTC (A-blue), Oxaliplatin enclosed Bulk Co-BTC (A-red), Nano Co-BTC (B-blue), Oxaliplatin enclosed Nano Co-BTC(B-red)

Comparison study of the energy-dispersive X-ray spectroscopy (EDX) images of Co-BTC MOF (Figure 9) confirmed the successful preparation of the Co-BTC MOF, and at the stage of drug absorption, the existence of Oxaliplatin particles pervading a good combination on the Co-BTC MOF Inside cavities. As shown in the EDX spectrum, the presence of oxaliplatin authenticated the EDX analysis for the stabilization of Oxaliplatin on the Co-BTC MOF. The presence of the element gold is due to the coating during identification.

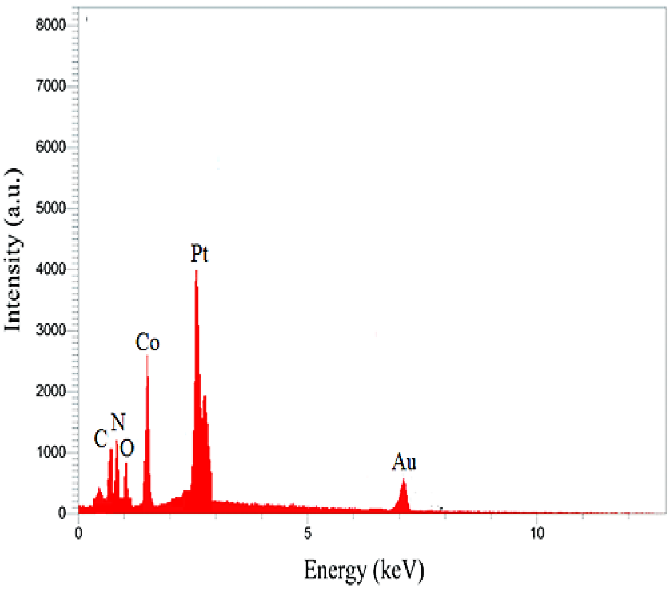



**Figure 9:** Energy-Dispersive X-ray spectroscopy (EDX) for Oxaliplatin enclosed Nano Co-BTC

## 3. Conclusion

Co-BTC MOF was synthesized by sonicated and thermal methods. The hollow cavities of the frames have an absorption for oxaliplatin and other anticancer drugs. The results of the drug adsorption pattern for Nano Co-BTC show a faster rate of oxaliplatin adsorption than the two samples of bulk Co-BTC and activated carbon, which is slower than them, which is a reason for its more regular crystalline pattern, which is confirmed by examining the XRPD patterns. Metal-organic frameworks (MOFs) can potentially be widely used in targeted drug delivery to enhance community health improvement methods due to their nature and have successful perspective in modern medicine.

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