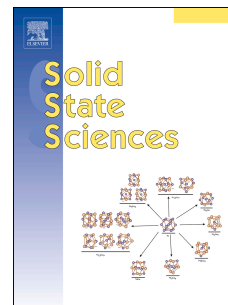


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# Journal Pre-proof

A facile and efficient approach to increase the magnetic property of MOF-5

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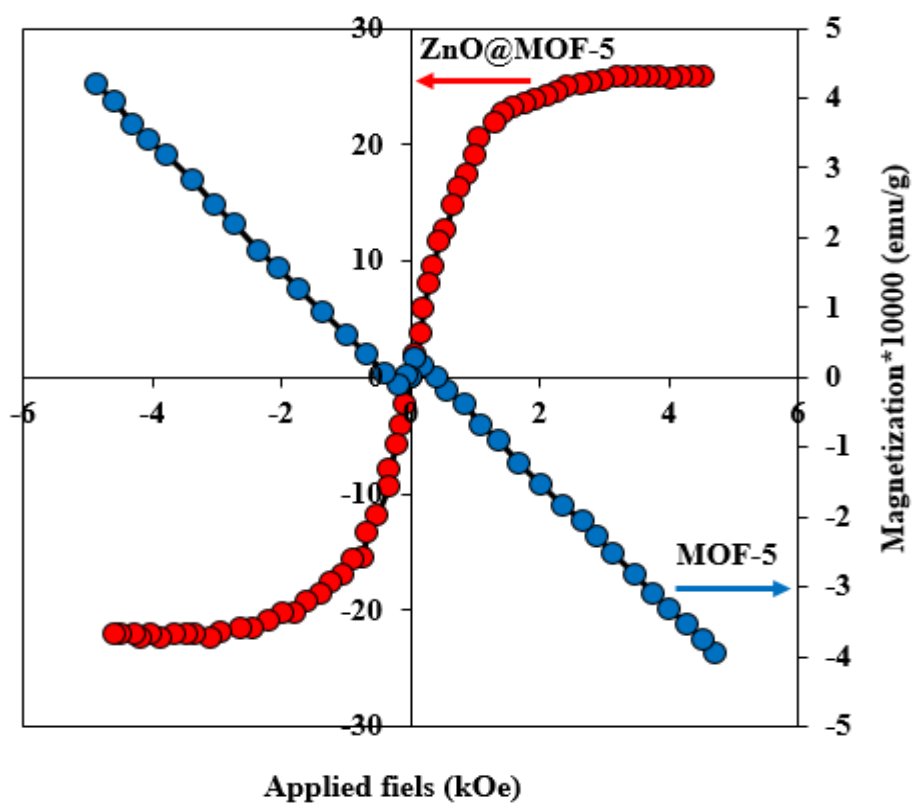
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## Graphical Abstract



## **A facile and efficient approach to increase the magnetic property of MOF-5**

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**Abstract**

In this study, a facile and efficient approach to increase the magnetic property of metal-organic framework-5 (MOF-5) has been investigated. The basis of this approach is the encapsulation of cluster-oxygen composition (i.e. ZnO in MOF-5) during the synthesis process of MOF-5 to form ZnO@MOF-5 nanocrystals. Both MOF-5 and ZnO@MOF-5 were synthesized for comparison purposes, considering their magnetic property. The physicochemical properties of MOF-5 and ZnO@MOF-5 were characterized by XRD, FTIR, TGA, DLS, FESEM, and Magnetization measurements. The FTIR spectra confirmed the presence of additional ZnO molecules in the ZnO@MOF-5 structure. Results from the XRD showed that the presence of additional ZnO molecules in the ZnO@MOF-5 altered the structure of MOF-5. The TGA analysis also confirmed the presence of additional ZnO molecules in the ZnO@MOF-5 structure, indicating that the ZnO@MOF-5 contains 15.23 wt% ZnO more than MOF-5. The FESEM and DLS results showed that the average sizes of MOF-5 and ZnO@MOF-5 nanocrystals are below 100 nm, with no defined morphology. Finally, the magnetization measurements showed that the MOF-5 nanocrystals have diamagnetic properties. For ZnO@MOF-5 nanocrystals, a ferromagnetic-like character was observed in the scanned field range and the saturation value of about  $2.59 \times 10^{-3}$  emu/g was obtained. The success of this facile and hassle-free approach can be an important step towards enhancing the magnetic properties of MOFs.

**Keywords:** MOF-5, ZnO, magnetic, organic ligand.

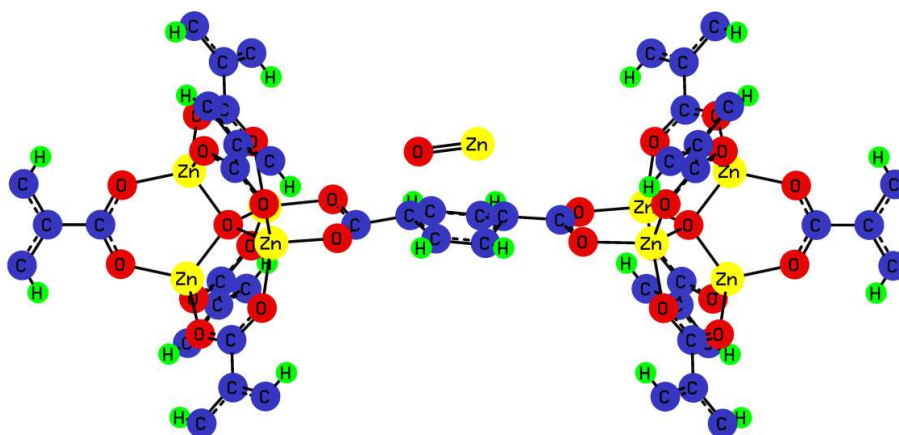
## 1 Introduction

Metal-organic frameworks (MOFs) are crystalline materials in which an inorganic metal cluster is bonded to an organic ligand to form an infinitely extended network-like structure [1-5]. Because of its large surface area and tunable pores, MOF show great potential for adsorption process [6-12]. Moreover, magnetic MOF inherit the good performance of MOF such as the rapid separation and recovery of magnetic species [13-15]. Among the various MOF adsorbents, MOF-5 is one of the most attractive adsorbents, which is dated back to 1999 [16]. This crystalline structure consists of  $Zn_4O$  as inorganic metal clusters bonded by 1,4-benzenedicarboxylate (BDC) as organic linkers to form a porous  $Zn_4O(BDC)_3$  framework, named pure or activated MOF-5 [16]. However, a pure MOF-5 does not have the desired magnetic properties [17-20], although several methods have been attempted to increase its magnetic properties.

Zhang et al., [17] synthesized a magnetic MOF-5 through a multiple step process using  $NiFe_2O_4$  as a magnetic core ( $NiFe_2O_4@MOF-5$ ). In the  $NiFe_2O_4@MOF-5$ , the MOF shells were formed on the surfaces of Fe containing magnetic nanoparticles (MNPs) and the surface area has been significantly decreased comparing with neat MOF-5. Moreover, the obtained magnetic saturation (MS) values of  $NiFe_2O_4@MOF-5$  (19.6 emu/g) were much lower than that of neat  $NiFe_2O_4$  MNPs (51.2 emu/g) due to the presence of MOF-5. In another study, Hu et al., [18] fabricated a hybrid magnetic MOF-5 with amino functionalized  $Fe_3O_4$  MNPs to form covalent binding with organic linkers on the MOF-5 structure. The obtained MS value for this hybrid MOF-5 was 0.22 emu/g. Adding  $Fe_3O_4$  to the MOF-5 did not affect the crystalline structure. Li et al., [19] synthesized a composite MNP using  $SiO_2@Fe_3O_4$  nanocomposite with MOF-5 structure grown on the surface of magnetic  $SiO_2@Fe_3O_4$ . In a similar way,  $MOF-5@Fe@SiO_2$  MNP was synthesized by encapsulation of  $Fe@SiO_2$  in the MOF-5 structure [20]. Despite the success in increasing the magnetic properties of MOF-5,

the proposed multi-stage methods are complex and there is a need to develop an efficient method that is able to shorten the path to achieve magnetic MOFs.

As a matter of fact, the presence of any extra species in the MOF-5 pores, which are generated during the synthesis process, changes the MOF-5 structure from activated to non-activated [5,11,21-24]. One of the most important excess molecules, which are present in the non-activated MOF pores is the cluster-oxygen composition (i.e. ZnO in MOF-5) that is removed during the activation process [21,23]. Investigations on synthesis of MOF-5 have shown that if  $H_2O_2$  were added into the solution of zinc nitrate and  $H_2BDC$ , ZnO species generate in MOF-5 pores due to the reaction between  $H_2O_2$  and zinc nitrate [5,21,23]. Although it was proved that at room temperature the ZnO species exhibit diamagnetism, Garcia et al. have shown that the electronic structure of ZnO becomes modified after the interaction between ZnO species and organic molecules, and the species exhibit a strong ferromagnetic-like behavior [23,25]. Fig. 1 shows the simulated primitive structure of ZnO@MOF-5 obtained by computational density functional theory (DFT) methods [23]. As shown in Fig. 1, the extra ZnO species in MOF-5 interacted with organic ligand [23,26]. Therefore, there is a possibility to use extra ZnO species as magnetic agents in the MOF-5 pores. In this point of view, it can be expected that the ZnO@MOF-5 particles show magnetic properties.



**Fig. 1** Simulated primitive cell of ZnO@MOF-5 (Red spheres, O; green, H; blue, C; yellow, Zn) [23].

The present study seeks to answer the question of whether extra ZnO particles in MOF-5 pores could be used as an agent to enhance the magnetic properties of MOF-5. Accordingly, MOF-5 and ZnO@MOF-5 are directly compared, considering their magnetic properties for the first time. The physicochemical properties of MOF-5 and ZnO@MOF-5 were characterized by X-ray diffraction (XRD), Fourier-transform infrared spectroscopy (FTIR), Thermal gravimetric analysis (TGA), Dynamic light scattering (DLS), Field emission scanning electron microscopy (FESEM), and Magnetization measurements (M). The success of this simple and hassle-free method can be an important step towards enhancing the magnetic properties of MOF, which has been investigated for the first time in this study.

## 2 Experimental

### 2.1 Materials

Benzene-1,4-dicarboxylic acid ( $\text{H}_2\text{BDC}$ , >99% purity), Zinc nitrate hexahydrate ( $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ , >99% purity), dichloromethane ( $\text{CH}_2\text{Cl}_2$ , 99.8% purity), N,N-dimethylformamide (DMF, 99.8% purity), triethylamine (TEA), and hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) were purchased from Sigma Aldrich and used as received.  $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  was stored in a nitrogen environment to reduce the effect of moisture.

### 2.2 Synthesis of MOF-5

For the synthesis of MOF-5, 0.166 g of  $\text{H}_2\text{BDC}$  and 0.9 g of  $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  were dissolved in a 200 ml bottle containing 98 ml of DMF and 2 ml of  $\text{H}_2\text{O}$ . The solution was maintained at 75 °C and vigorously stirred. The solution was heated for 30 min and it was placed in an oven at 110 °C for 10 h. Then, the reaction vessel was cooled to ambient temperature and solvent was removed. After removing the solvent, white powder was washed five times with 100 ml of anhydrous DMF and five times with 100 ml of anhydrous  $\text{CH}_2\text{Cl}_2$ . Finally, the MOF-5 crystals were dried for 20 h in a vacuum oven at 115 °C.



### 2.3 Synthesis of ZnO@MOF-5

For the synthesis of the MOF-5 samples containing extra ZnO species (ZnO@MOF-5), 0.68 g of H<sub>2</sub>BDC and 3 g of Zn(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O were mixed in 100 ml of DMF. Afterward, ten drops of H<sub>2</sub>O<sub>2</sub> solution were added under continuous stirring. After six minutes of continuous stirring, 5 ml of TEA was added into the solution. The H<sub>2</sub>O<sub>2</sub> acted as an oxygen precursor and caused the formation of extra ZnO species. In addition, the TEA was used for the deprotonation of H<sub>2</sub>BDC through reaction with Zn<sup>2+</sup> ions. After the continuous stirring of the solution at 60 °C for 3 h, the solution was placed in an oven at 110 °C for 10 h. Then, the reaction vessel was cooled to room temperature. The white product was soaked in anhydrous DMF for 10 h. Finally, the ZnO@MOF-5 particles were dried for 24 h in a vacuum oven at 115 °C.

### 2.4 Characterization

FTIR patterns of the MOF was recorded at 4000-400 cm<sup>-1</sup> (Thermo Nicolet Avatar 370). The thermal stability of samples was measured by TGA at a heating rate of 10 °C/min (TGA-50, Shimadzu). The X-ray diffraction patterns of the synthesized MOF was measured by XRD (PW3710, Philips). In order to measure the size of MOF, the DLS analysis was performed (Shimadzu sald-2101). The morphology of the synthesized MOF was observed with FESEM images (TM3000, Hitachi). The magnetic measurements of the MOF-5 and ZnO@MOF-5 were carried out using a vibrating sample magnetometer (VSM, Lake Shore 7307, USA).

## 3 Results and discussion

The XRD patterns of MOF-5 and ZnO@MOF-5 are shown in Fig. 2. The obtained results were similar to the reported XRD patterns [2,21,22]. Both MOF-5 and ZnO@MOF-5 show structural changes via the increasing and decreasing of two main peak. If the structure of MOF-5 changed from the neat MOF-5 to the ZnO@MOF-5, the intensity at  $2\theta=7.0$  becomes

lower than that at  $2\theta=9.8$ , and vice versa, as shown in Fig. 2. The differences in XRD patterns are attributed to the presence of additional ZnO molecules in the ZnO@MOF-5 structure [5,21-24].

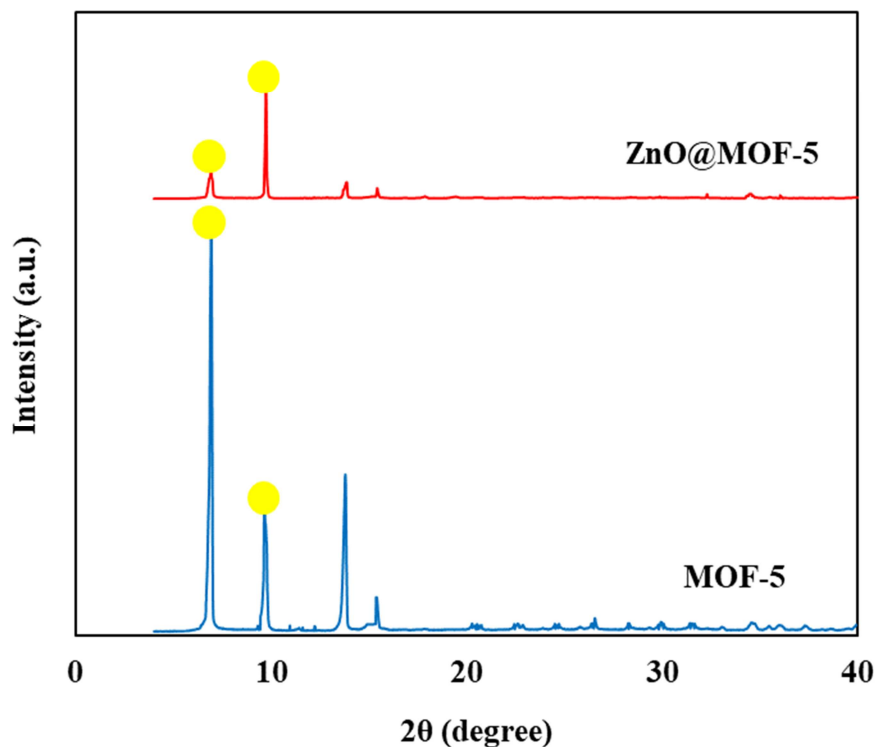


Fig. 2 XRD patterns of MOF-5 and ZnO@MOF-5 nanocrystals.

The FTIR spectra of MOF-5 and ZnO@MOF-5 are shown in Fig. 3. A detailed discussion of the FTIR spectrum of MOF-5 can be found in the literature [3,27-30]. As shown in Fig. 3, the obtained results in this study are similar to those reported in the literature. According to obtained results, all main peaks in both MOF-5 and ZnO@MOF-5 are quite similar. However, the most significant difference between these two MOFs is a band appeared around  $530\text{-}400\text{ cm}^{-1}$  for ZnO@MOF-5 that is attributed to the presence of additional ZnO molecules in the ZnO@MOF-5 structure [3,5,23,27,28,30].

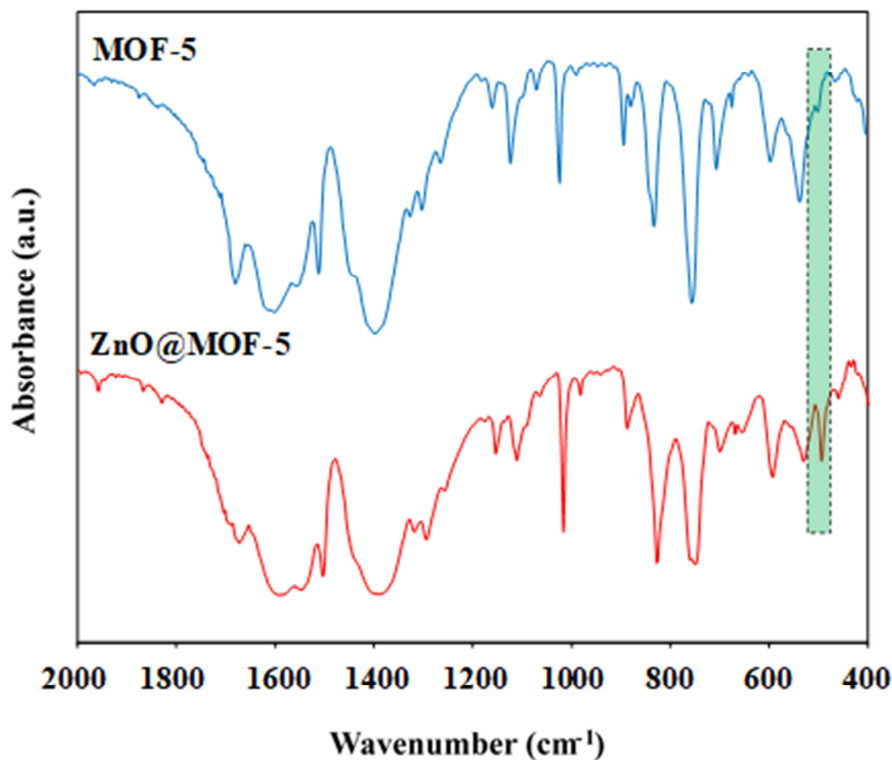
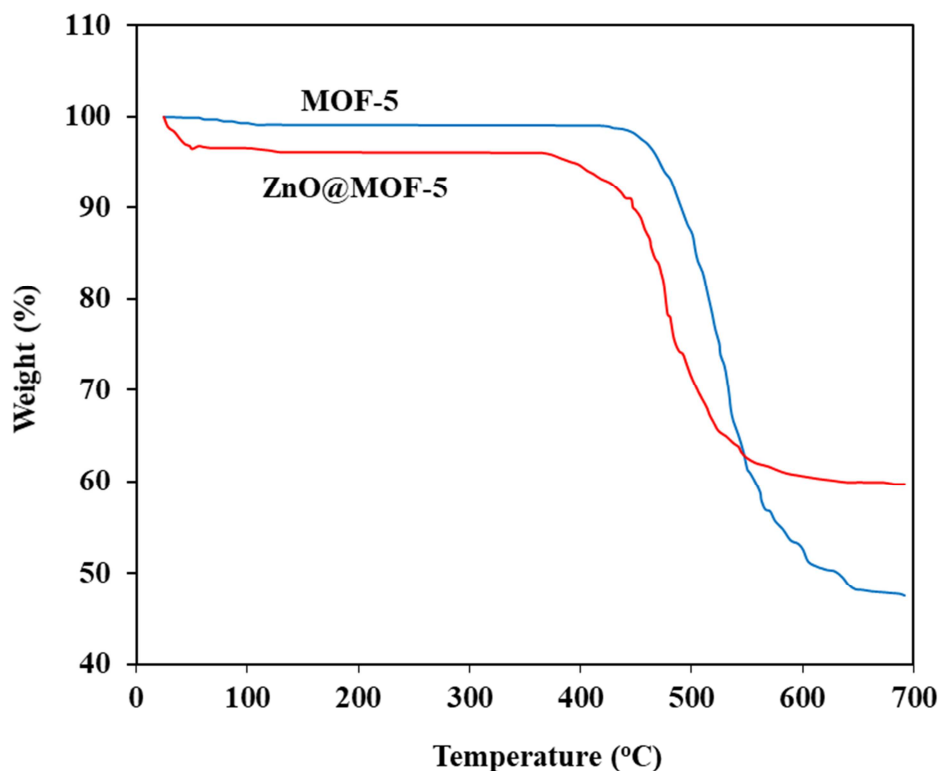


Fig. 3. FTIR analysis of MOF-5 and ZnO@MOF-5 nanocrystals.

The TGA curves of MOF-5 and ZnO@MOF-5 are shown in Fig. 4. According to the obtained results, two weight loss stages occurred during the decomposition of both MOF-5 and ZnO@MOF-5: (I) desorption of H<sub>2</sub>O in the range of 30-300 °C (MOF-5=1% and ZnO@MOF-5=4%), and (II) decomposition of main structure above 350 °C that releases CO<sub>2</sub> and benzene (MOF-5=51% and ZnO@MOF-5=36%). No solvent was observed in both MOF-5 and ZnO@MOF-5. The final products from the decomposition of both MOF consist of carbon and ZnO [21-23]. As shown in Fig. 4, the percentage of remaining mass for MOF-5 and ZnO@MOF-5 is 48.43 and 63.67, respectively. Theoretically, the ZnO percentage in neat MOF-5 is 42% [21]. Assuming the same amount of carbon in the remaining mass of both MOF-5 and ZnO@MOF-5 [21], the remaining mass for MOF-5 (48.39%) and ZnO@MOF-5 (63.62%) showed that the final products of the ZnO@MOF-5 contain 15.23 wt% ZnO more than MOF-5.



**Fig. 4.** TGA analysis of MOF-5 and ZnO@MOF-5 nanocrystals.

Both MOF were compared in terms of appearance and size, and the results are shown in Figs. 5 and 6. As illustrated in Fig. 5, the crystalline structure of MOF-5 and ZnO@MOF-5 samples are evident, with no defined morphology. Similar morphologies were obtained in other studies that showed no defined crystal morphology with aggregates similar in size to the nanocrystals synthesized in this study<sup>2</sup>. However, high-magnified FESEM images illustrate the differences between MOF-5 and ZnO@MOF-5. Fig. 5 shows that the ZnO@MOF-5 crystals are relatively smooth and are surrounded by ZnO particles having uniform rod-like structures. However, in the MOF-5 sample, no traces of rod-like ZnO particles were observed. It is also clear that both MOF-5 and ZnO@MOF-5 crystals have dimensions less than 100 nm. The results of the DLS analysis shown in Fig. 6 agree with the results of Fig. 5. Furthermore, as shown in Fig. 6, the ZnO@MOF-5 nanoparticles are slightly larger than MOF-5 nanoparticles.

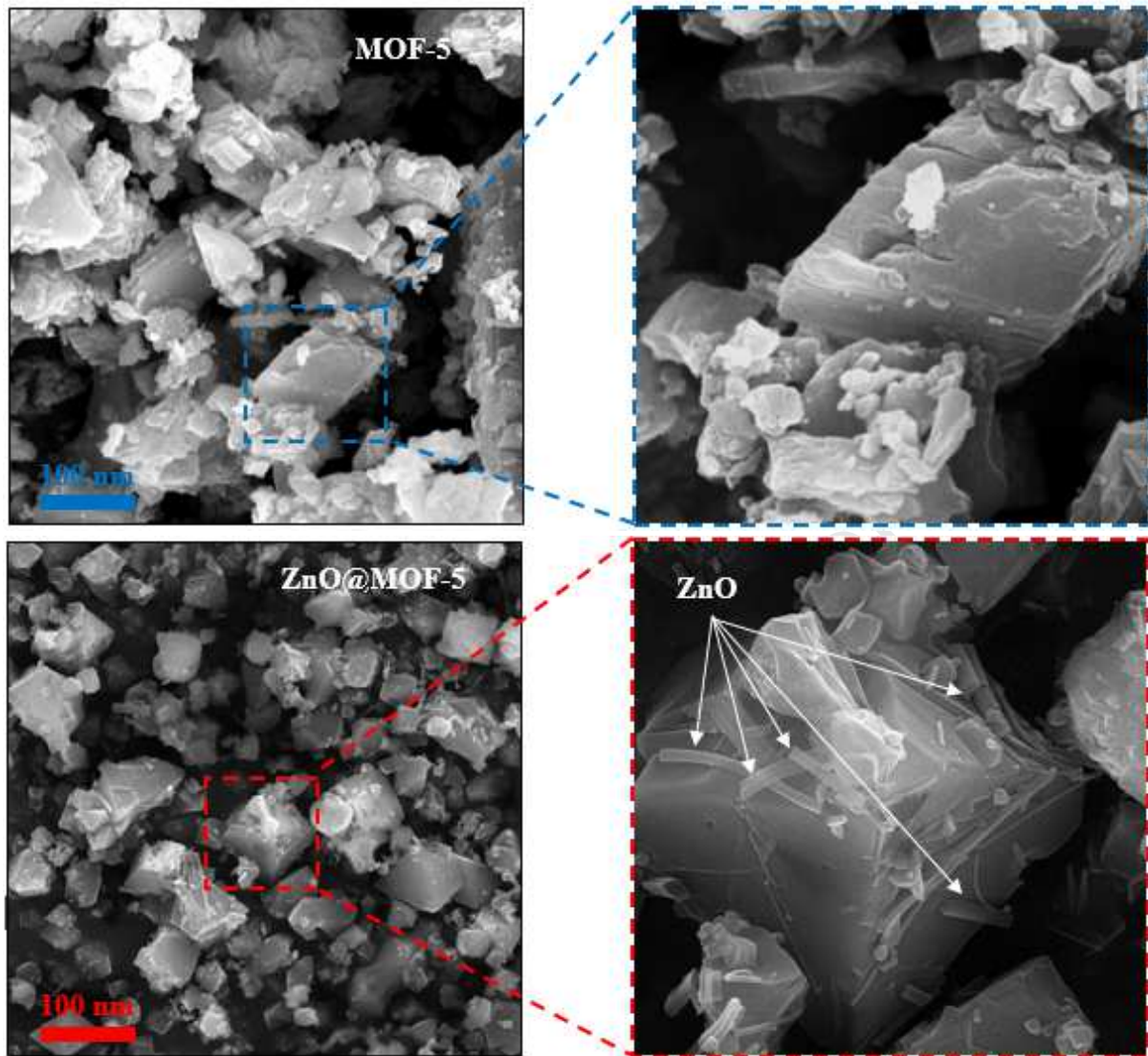


Fig. 5 FESEM images of MOF-5 and ZnO@MOF-5 nanocrystals.

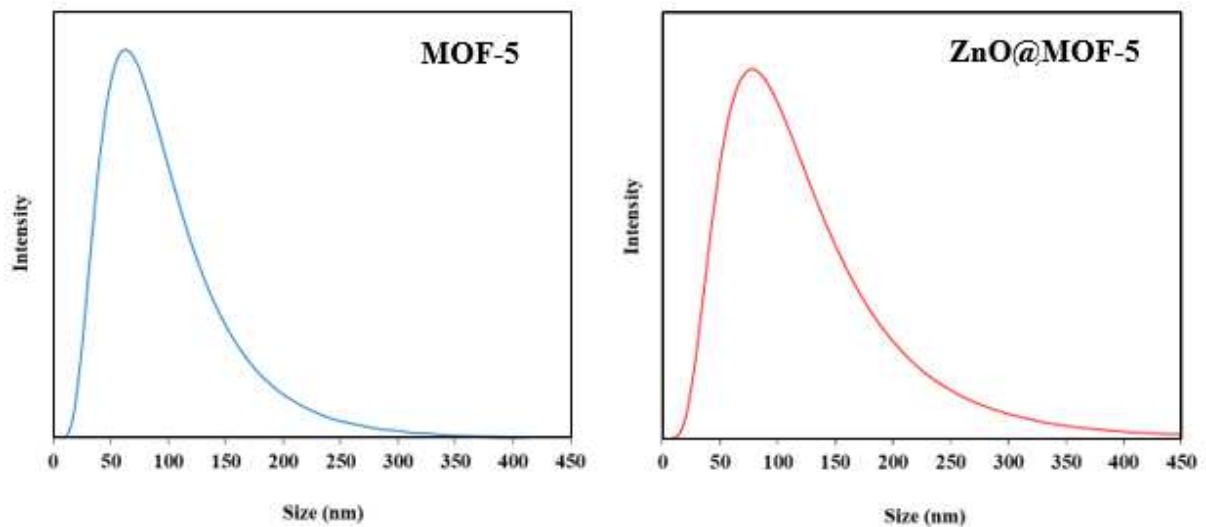


Fig. 6 DLS measurement of MOF-5 and ZnO@MOF-5 nanocrystals.

Based on analytical studies carried out in this study, it was found that ZnO@MOF-5 nanoparticles contain a higher percentage of ZnO molecules in their structure compared to MOF-5 nanoparticles. To investigate the effect of the presence of additional ZnO molecules on the magnetic properties of the ZnO@MOF-5 structure, the magnetic characterization was performed on both MOF-5 and ZnO@MOF-5 (Fig. 7). The results showed that the MOF-5 nanocrystals have diamagnetic properties, as expected. However, for ZnO@MOF-5 nanocrystals, a ferromagnetic-like character was observed in the scanned field range, as shown in Fig. 7. A saturation value of about  $2.59 \times 10^{-3}$  emu/g was measured for the ZnO@MOF-5. Therefore, it can be concluded that the presence of additional ZnO molecules in the ZnO@MOF-5 structure will cause fundamental changes in the magnetic properties of MOF-5 nanocrystals. However, at room temperature, the single ZnO species exhibit diamagnetism behavior [25]. The DFT results presented in the literature showed that the additional ZnO molecules in the ZnO@MOF-5 nanocrystals interacted with organic ligand [23]. It was also proven that the electronic property of ZnO molecules can be strongly changed after interaction between ZnO molecules and organic molecules, and strong ferromagnetic-like behavior can be achieved [25]. Therefore, the combination of these two phenomena could be the main cause of these changes in magnetic properties.

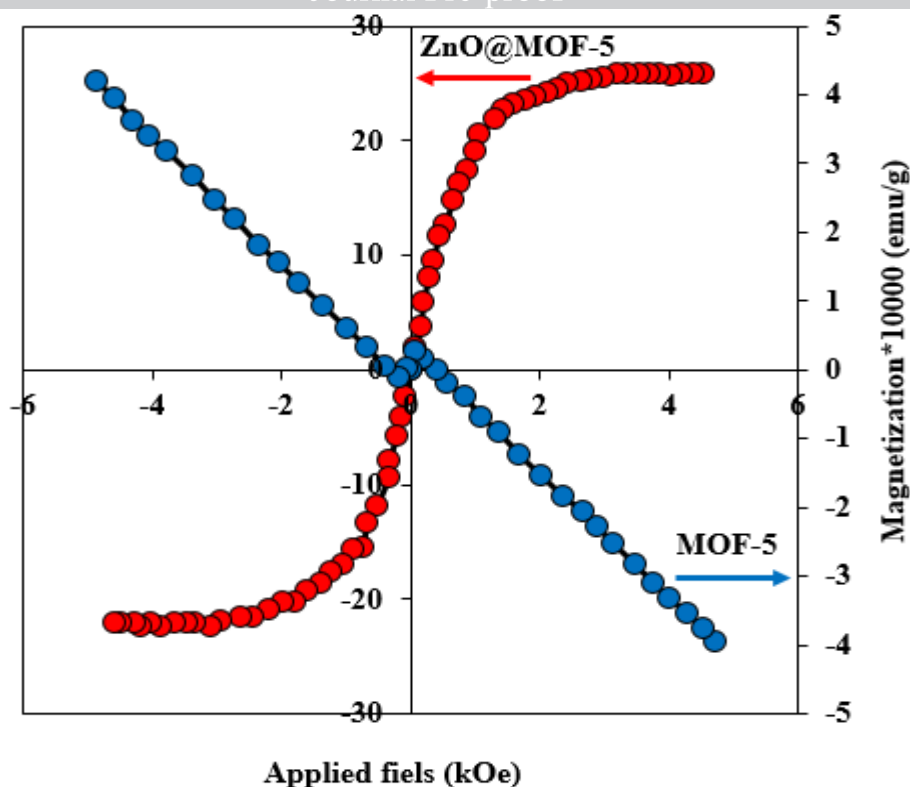


Fig. 7 Magnetization curves of MOF-5 and ZnO@MOF-5 nanocrystals.

## 5 Conclusions

We reported for the first time, a facile and efficient approach to increase the magnetic property of MOF-5 based on the encapsulation of cluster-oxygen composition during the synthesis process. The following key points were concluded from this study:

- i) The FTIR spectra confirmed the presence of additional ZnO molecules in the ZnO@MOF-5 structure
- ii) The XRD patterns showed that the structure of ZnO@MOF-5 differs from the MOF-5 structure
- iii) The TGA results showed that the ZnO@MOF-5 contain 15.23 wt% ZnO more than that of MOF-5

iv) The FESEM and DLS results showed that the average size of MOF-5 and ZnO@MOF-5 nanocrystals are below 100 nm, with no defined morphology

v) The magnetization measurements showed that the MOF-5 and ZnO@MOF-5 nanocrystals have diamagnetic and ferromagnetic-like properties, respectively, in the scanned field range.

vi) The saturation value of the magnetization curve for the ZnO@MOF-5 is about  $2.59 \times 10^{-3}$  emu/g.

Finally, it can be concluded that the presence of additional ZnO molecules in the ZnO@MOF-5 structure will cause fundamental changes in the magnetic properties of MOF-5 nanocrystals. The success of this facile and hassle-free approach can be an important step towards enhancing the magnetic properties of MOFs.

### **Acknowledgment**

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## Research highlights

- A facile and efficient approach to increase the magnetic property of MOF-5
- ZnO@MOF-5 contain 15.23 wt% ZnO species more than that of MOF-5
- Evaluation of the impact of additional ZnO species on the magnetic property of MOF-5
- A ferromagnetic-like character was observed for ZnO@MOF-5 nanocrystals

<b>Term</b>	<b>M. Arjmandi</b>	<b>A. Altaee</b>	<b>M.P. Chenar</b>	<b>A. Arjmandi</b>	<b>M. Peyravi</b>	<b>M. Jahanshahi</b>
Conceptualization						
Methodology						
Software						
Validation						
Formal analysis						
Investigation						
Resources						
Data Curation						
Writing - Original Draft						
Writing - Review & Editing						
Visualization						
Supervision						
Project administration						
Funding acquisition						

**Declaration of interests**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: