

**UNIVERSITI TEKNOLOGI MARA**

**LOW COST AND GREEN  
SYNTHESIS OF REDUCED  
GRAPHENE OXIDE-ZEOLITIC  
IMIDAZOLE FRAMEWORK 8 (RGO-  
ZIF-8) USING MOTHER LIQUOR  
RECYCLING**

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## ABSTRACT

Zeolitic imidazole frameworks-8 are have a high interest in industries application due to its properties which are high surface area, modifiable organic and high chemical and thermal stability. However, to synthesized ZIF-8 in large scale, there might be a problem on the cost because of the expensive imidazole species solvent. To this work, Green hybrid nanocomposite of reduced graphene oxide/zeolitic imidazolate framework-8 (rGO-ZIF-8) has been successfully fabricated via multi-recycling approach by re-using non-reactant reagents in supernatant known as mother liquor. This approach allowed minimal addition of fresh reactant and rGO was used as a template for in situ growth of ZIF-8. The solid product of each batch was collected and characterized by using XRD, TGA, FTIR, BET, FESEM and TEM. The mother liquor then recovered and re-use for the subsequent cycle of synthesis up to 4 times. The XRD analysis shows that the ZIF-8 samples have diffraction peaks  $2\theta$  at 7.28, 10.35, 12.66, 14.64, 16.39, 17.97 peaks indicate that the formation of ZIF-8 was successful. The diffraction peaks for rGO-ZIF-8 shows the same pattern indicates that the addition of rGO does not changing the crystal structure of ZIF-8. The TGA results revealed that there was no significant difference in thermal stability of ZIF-8(Z1) and all the recycled ZIF-8 samples. For rGO-ZIF-8 samples, higher amount of rGO added was found to increase the thermal stability of the samples. The surface area and pore volume of the recycled ZIF-8 samples were lowered compared to ZIF-8(Z1). The surface area of 70r-Z and 140r-Z decrease proportionally with the addition high amount rGO suggests that there might be strong association between ZIF-8 and rGO that the cavities inside ZIF-8 might be partially blocked by rGO sheets. It was demonstrated that ZIF-8 nanoparticles anchored well on the surfaces of rGO and high product yield can be maintained. This approach allows low cost and green synthesis of rGO-ZIF-8 synthesis as the waste discarded is minimal.

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# CHAPTER ONE

## INTRODUCTION

### 1.1 Research Background

Metal Organic Frameworks (MOF) are multidimensional networks that consist of metal centre and organic linker. Nowadays, there are many applications of MOFs due to their attractive properties such as tunable pore size and functionality. Certain classes of MOFs have a very high chemical stability and thermal stability. These properties have encouraged researchers over the past 15 years to unlocked more potential application of MOFs. Gas storage and separation, gas adsorption, drug delivery and catalyst are examples of MOFs application (Marshall, 2017). Most MOFs do not require high temperature and pressure condition make it easier in synthesized process. There are several methods to synthesized the MOFs such as solvothermal, hydrothermal centrifugal and microwave induced thermal methods.

There is a wide variety of MOFs known and it can be increased by the post-synthetic modification (PSM) which can modified desired properties and structure of the MOFs towards their applications. In PSM, the modification can be done by adding functional groups to the linker or changing the metal ions in the MOFs. Direct synthesized of MOFs does not have their desired properties thus variety of PSM process can be performed. Covalent post synthetic modification, post synthetic deprotection (PSD) and post synthetic exchange (PSD) are the most common in MOFs modification.

Zeolitic imidazole frameworks (ZIF) is a subclass of MOFs which consist of crystalline porous materials. Their topology is similar to zeolites and have many valuable properties such as high surface area, modifiable organic and high chemical and thermal stability. These properties make them have a high potential in the gas sensors, catalytic membrane reactors and gas separation membranes application. ZIF-8 has the most interest due to their robust synthesis and their potential to separate small gases. It has large pores of 11.6 Å and small pores apertures 3.4 Å (Kwon & Jeong, 2013). Their thermal stability also up to nearly 400 °C. The expensive and environmentally undesired