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Green Preparation of Aluminum-based Metal-Organic Framework (Al-MOF) from Waste Plastic Bottles and Waste Aluminum Scraps

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ABSTRACT: The vast use of polyethylene terephthalate (PET) drinking water bottles has increased dramatically worldwide in recent decades, inflicting severe consequences on the environment. According to the latest survey, a million plastic bottles are bought around the globe every minute. The non-biodegradable nature of PET materials has led to a huge accumulation of plastic in waste landfills. Among the current recycling methods used to solve this environmental problem is chemical recycling. In this method, PET bottles are converted back cleanly into their starting materials: terephthalic acid and ethylene glycol. This paper unveils the exploitation of recycling products from PET bottles and aluminium scraps in order to prepare a metal-organic framework (MOF) material. The characterization of the prepared MOF substance was carried out using different techniques such as IR, XRD, SEM and elemental analysis.

Keywords: Polyethylene terephthalate (PET); Aluminium scarp; Metal-organic framework (MOF).

تصنيع مركب كيميائي من معدن الألومنيوم مرتبط بمادة عضوية (Al-MOF) من مخلفات الألومنيوم الصناعية ومخلفات العلب البلاستيكية

صالح البوصافي و يحيى الشافوري

الملخص: إن الزياده المتطرده في استخدام علب المشروبات البلاستيكية المصنعة من مادة البولي إيثيلين تيريفثالات أدت الى تراكم هذه العلب في البيئه بشكل ملحوظ. أن خطورة هذه المواد ناتجه في المقام الأول من مقاومتها للتحلل البيولوجي في البيئه وبالتالي بقائها لفتره زمنيه طويله نسبيا. أفضل الحلول المطروحه للتخلص من النفايات البلاستيكية هي إعادة تدوير هذه المواد كيميائيا لإعادتها الى موادها الأوليه وهي:حمض التيريفثاليك وإيثيلين جلايكول. في هذه الورقه العلمية قمنا بتدويرعلب الماء البلاستيكيه كيميائيا وإنتاج مادة حمض التيريفثاليك وأيضا قمنا بتدوير خردة الألمنيوم وتحويلها الى مادة كلوريد الألمنيوم. وفي النهاية قمنا بتفاعل هاتين المادتين (حمض التيريفثاليك و كلوريد الألمنيوم) لانتاج مادة مبلمرة تحتوي على فلز الألمنيوم مرتبط بوحده عضوية (Al-MOF). تم اسخدام وسائل الكم الطيفي لتحديد صفات المركب الجديد.

الكلمات المفتاحية: بولي اثيلين تيري فثالات، خردة الألمنيوم، مركبات فلزيه-عضويه.

1. Introduction

Used plastic drinking bottles, among other plastic waste, present a serious environmental problem in our society. Although plastic materials have been playing a significant role in our modern life for their use in such as food packaging, water and beverage packaging, and mobile phone manufacturing, among other useful daily life applications [1], their non-degradable nature leads to rapid accumulation in the surroundings causing appalling environmental effects [2]. In particular, polyethylene terephthalate (PET) plastic, which is produced by the condensation reaction of terephthalic acid (Benzene dicarboxylic acid, BDC) and ethylene glycol (EG) has received the greatest attention because of its widespread use in manufacturing most water and soft-drinks bottles [3]. The vast growth of the use of PET plastics globally is reflected in the increased worldwide production of these materials, which reached over 26 million tonnes (around 68 billion bottles) in 2019, of which bottled beverages was the largest end-use market accounting for 30% of global PET packaging consumption [4]. In Oman, almost 50% of the plastic wastes, or 190,000 metric tonnes, dumped in landfills in 2019 were of PET [5]. Currently, the common practice for disposing of used PET bottles is by landfilling, which has led to serious environmental problems due to the poor biodegradability of PET bottles [6]. Another reported method is for recycling PET bottles by converting them back to their chemical constituents, BDC and EG [7]. This chemical method of recycling PET drinking bottles is considered a green and economical source of terephthalic acid and can contribute an acceptable way to eliminate PET waste landfilling, and thus lead to a cleaner environment.

Another issue is that global aluminium production reached 63.7 million metric tonnes in 2019 with 5.8 million metric tonnes being from Gulf Cooperation Countries (GCC) countries [8]. Due to its marvellous industrial properties such as light weight, conductivity, and corrosion resistance, aluminium finds its way into the manufacturing of a wide range of metallic applications such as automobiles, aeroplanes, kitchen tools, cabinets, foils, beverage cans, cables, and window and door frames [9-10]. To meet the increasing global consumption of aluminium, huge amounts of bauxite must be excavated, which in turn produces a significant amount of wastes. One way to minimize the production of bauxite, and hence reduce the burden on the environment, is by recycling used aluminium. Statistics show that for every 1 Kg of recycled aluminium, 8 Kg of bauxite can be saved [11]. In this context, waste aluminium foils and cans have been recycled to prepare alumina (Al_2O_3) and aluminium nitrate $Al(NO_3)_3$ [12-14]. In addition, aluminium scraps resulting from machining and trimming operations can be recycled to make beneficial chemicals.

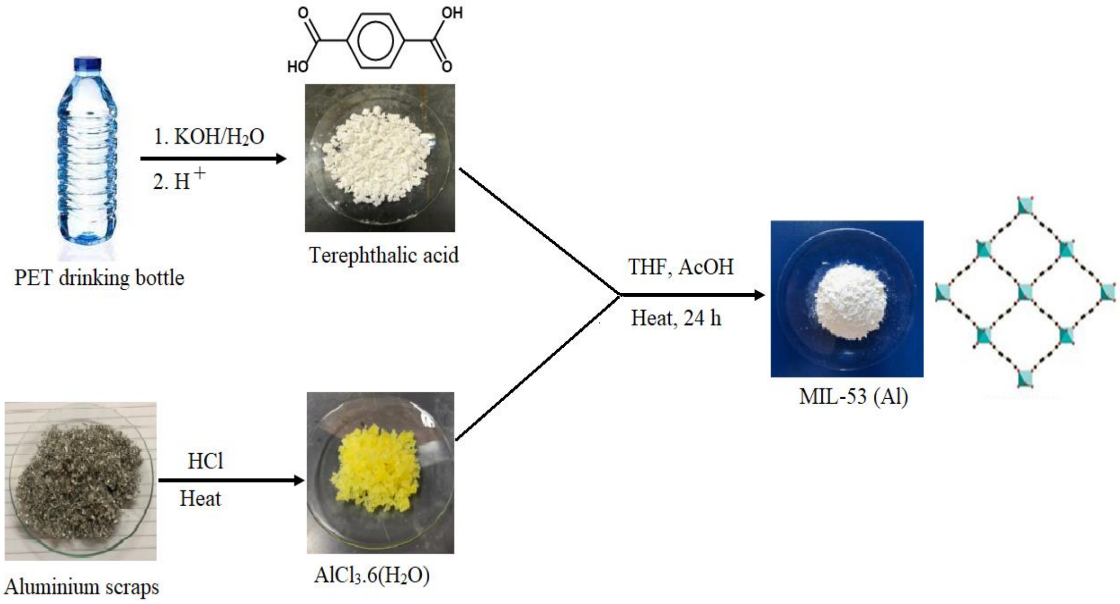
During the past two decades, Metal-Organic Frameworks (MOFs) have become the focus of much investigation due to their versatile properties such as high surface area, high porosity, and tuneable pores [15-17]. MOFs are coordination polymers prepared from metal ions (clusters) and organic linkers, giving rise to extended 3-D open framework structures, with incorporated solvent molecules [18]. MOFs have rapidly found their way in many applications such as separations, gas storage, heterogenous catalyses, molecular recognition, and drug delivery [19-23]. Different metals such as Al, Ni, Co, Cu, Zn, Cr, Ga, Fe, V, and Zr have been used to construct MOFs [24-26]. Among the known MOFs, MIL-53 (Al) has attracted attention due to its double structural transitions (breathing) upon some gas adsorption [27, 28]. In addition, MIL-53 (Al) has been used as a catalyst for Friedel-Crafts alkylation of benzene with ethanol [29]. The source of aluminium in all reported syntheses of MIL-53 (Al) is either $Al(NO_3)_3.9(H_2O)$ or $AlCl_3.6(H_2O)$ [30, 31].

Terephthalate (benzene dicarboxylate, BDC) is used extensively as an economical and viable source of a dicarboxylate linker to construct many nanoporous MOFs such as MIL-101(Cr), MIL-47(V), and MIL-53(Al, Ga, Cr) [32]. In this paper, we report the synthesis of MIL-53 (Al) from terephthalic acid produced from recycled drinking water bottles and aluminium chloride hexahydrate produced from recycled aluminium scrap as shown in Scheme 1.

2. Materials and Methods

2.1 General

All reagents are commercially available and were used as received without further purification. Waste drinking PET bottles were washed well and cut into small pieces and used to prepare terephthalic acid. Aluminium scraps were collected from Maabella industrial state and used to prepare $AlCl_3.6(H_2O)$. Melting points were determined using Stuart SMP20 Digital Melting Point Apparatus (SLS Scientific Laboratory supplies, UK). Infrared spectra (IR) were measured on Agilent technologies Cary 630 FT-IR (Agilent Technologies, USA). ^1H NMR and ^{13}C NMR spectra were carried out in a JEOL 400 MHz spectrometer (Japan) with $CDCl_3$ as the NMR solvent. Elemental analysis measurements were performed using the Euro vector CHN elemental analyser EA3000 model. Aluminium content in MIF-53 was performed on an ICP-OES instrument using the nitric acid extraction method. ESI-MS spectra were recorded with Agilent, 6460 Triple quad LC/MS, 1200 Infinity series (Germany) equipped with an electrospray ionization (ESI) interface and operated by Mass Hunter software. Powder XRD diffraction (PXRD) measurements were performed on a Malvern Panalytical X'Pert Pr instrument using Cu Kα1.5405 Å. The microstructure and sample surface morphology of $AlCl_3.6(H_2O)$ and MIL-54(Al) were examined with scanning electron micrographs (SEM) model JSM-7600F- JEOL.



Scheme 1. Graphic illustration of the synthesis of MIL-53 (Al) from recycled PET bottles and aluminium scraps.

2.2 Conversion of PET bottles into terephthalic acid

Clean PET chips (10 g) and KOH (10 g) were mixed with water (10 mL) and ethanol (200 mL) in a round-bottomed flask (500 mL) and the mixture was heated under reflux for one day. After cooling to room temperature, the white solid was filtered by suction and allowed to dry. The resulting white solid was dissolved in water and the solution was acidified by adding aqueous H_2SO_4 (1M) until the white solid reappeared. The solid was filtered by suction filtration, and dried first under vacuum and then in the oven to yield 8.42g (84.2% w/w) of a white solid. Mp 298.2 ºC, Elemental analysis: %C = 58.091, %H = 3.234, %O = 38.676 (Empirical formula = $C_8H_6O_4$); IR (Neat): 2515-3311(broad band), 1672, 1574, 1509, 1422, 1279, 927, 878, 782, 726 cm^{-1}. ^{1}H-NMR (400 MHz, $CDCl_3$): δ_{ppm} = 8.04 (4H, s); ^{13}C-NMR (100.6 MHz, $CDCl_3$): δppm = 129.64 (4C), 134.60 (2C), 166.89 (2C). ESI m/z: calcd for $C_8H_6O_4$ 166; found 166.

2.3 Conversion of aluminium scraps into $AlCl_3.6H_2O$

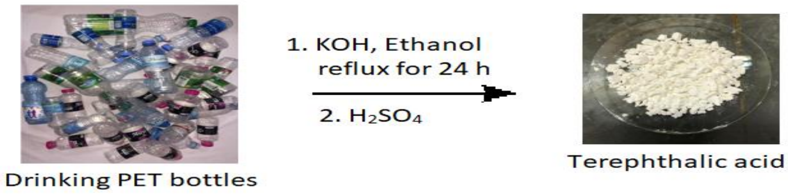
Aluminium scraps (10 g) were transferred to a conical flask containing 375 mL HCl (6 M) and the mixture was stirred at room temperature. The exothermic reaction started after 1 minute and lasted for 30 minutes. Then the cold mixture was filtered to remove the unreacted Al pieces by suction and the clear filtrate was left in the fume hood for one week until yellow crystals of aluminum chloride hexahydrate formed. The mixture was filtered by suction and the yellow crystals were dried under vacuum to yield 27.60 g (30.8%). The yellow crystals decomposed after 300 ^{0}C, IR (Neat): 2600-3200 (broad band), 2405, 1950, 1622, 1139, 817, 758, 608 cm^{-1}.

2.4 Synthesis of MIL-53 (Al) from terephthalic acid and $AlCl_3.6H_2O$

A mixture of terephthalic acid (1.0 g, 6.1 mmol), $AlCl_3.6H_2O$ (1.5 g, 6.1 mmol), DMF (150 ml) and acetic acid (1.5 ml) was heated under reflex for 3 days. The white solid was filtered by suction, then washed with DMF to remove unreacted terephthalic acid, and next washed with water several times to remove DMF. The product was dried first under vacuum and then in the oven to yield MIL-53 as a white solid 1.75g. The white solid decomposed after 300 ºC. Elemental analysis: %C = 39.078, %H = 2.636, %O = 44.450, %Al = 13.820%, (Empirical formula = $C_6H_5AlO_5$); IR (KBr): 1695, 1594, 1509, 1413, 1274, 988, 833, 751 cm^{-1}.

3. Results and discussion

3.1 Preparation of terephthalic acid from PET drinking bottles



Scheme 2. Conversion of PET drinking bottles into terephthalic acid.

Hydrolysis of PET can be achieved either under acidic or basic conditions. The process under an acidic condition was reported to be slow compared to that under a basic one [33]. Moreover, hydrolysing PET under a basic condition enabled us to separate the unreacted PET flakes before the acidifying step. After mixing PET pieces with potassium hydroxide in a mixture of ethanol and water (20:1), a white potassium terephthalate salt rapidly started to appear. After 24 hours of heating under reflux, the white solid, along with unreacted PET flakes, was separated and dissolved in water. Another filtration was done to separate the unreacted PET, followed by acidification with aqueous H_2SO_4 to recover terephthalic acid as a white solid.

3.1.1 IR analysis

The IR spectrum of the prepared terephthalic acid revealed the characteristic broad stretching O-H band of carboxylic acid in the range 2515-3311 cm^{-1} and a strong C=O stretching band at 1672 cm^{-1} (**Figure 1**). The lower wavenumber of C=O is due to conjugation with a benzene ring. Other distinctive bands appeared at 1574 (C=C stretching), 1279 (C-O stretching), and 782 (C-H bending). The IR spectrum showed good matching when compared with a previously reported spectrum [34].

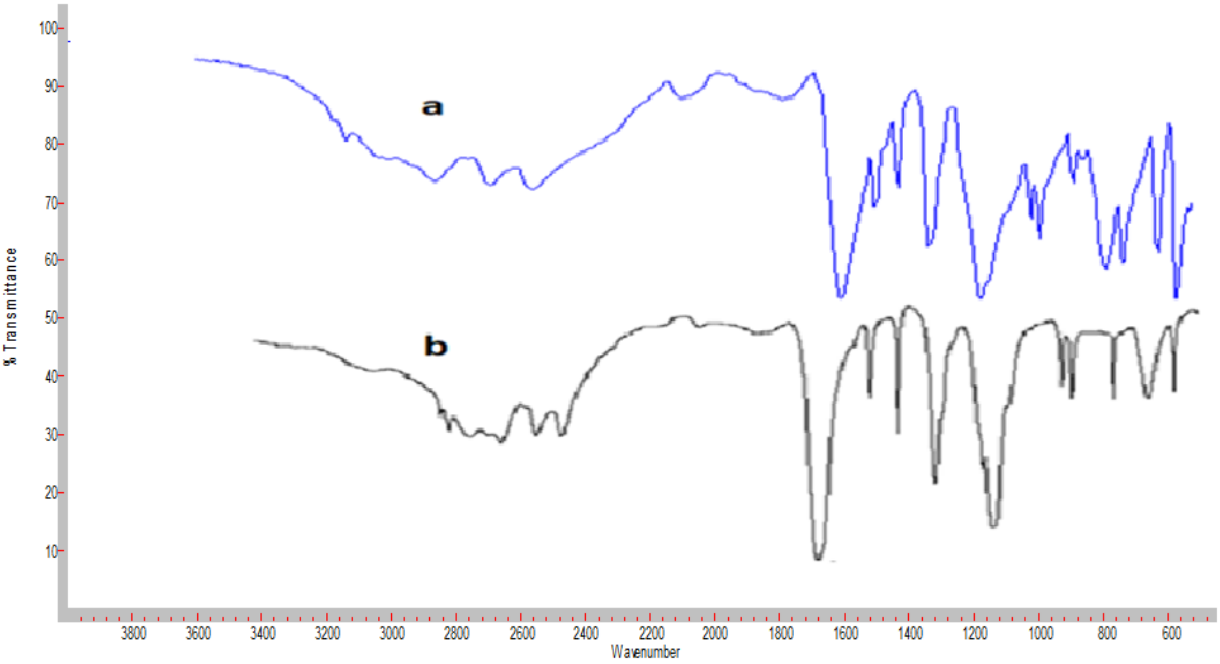
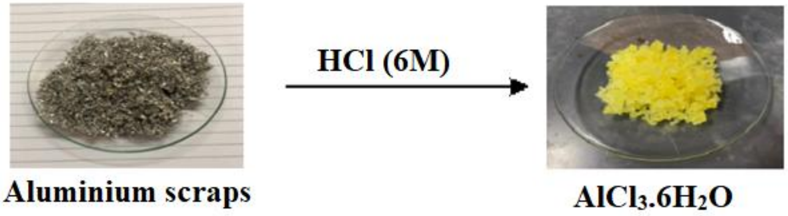


Figure 1. (a) IR spectrum of the prepared terephthalic acid and (b) the reported spectrum

3.1.2 NMR analysis

The ^1H-NMR spectrum of the prepared terephthalic acid exhibited a characteristic signal at 8 ppm for the four hydrogens in the benzene ring (**Figure 2**). The other signal at 2.5 belongs to the DMSO-d_6 solvent used in the NMR analysis. In addition, the ^{13}C- NMR spectrum showed two signals for the benzene ring carbons, one signal at 129.6 ppm for the four CH carbons and one at 134.6 ppm for the two quaternary carbons. The signal at 166.9 ppm is for the carboxyl group. The NMR spectrum of the prepared terephthalic acid exactly matched the reported one [35].

3.2 Preparation of $AlCl_3.6H_2O$ from aluminium scraps



Scheme 3. Conversion of Al scraps into hydrated aluminium chloride.

One minute after mixing aluminium scraps with HCl (6 M) an exothermic reaction started, indicating a reaction between Al and HCl. The heat subsided after 30 minutes to give a milky mixture which was filtered to remove unreacted Al scraps. The mixture was then left out in the fume hood to evaporate slowly, and following this, left in the sun until yellow crystals formed.

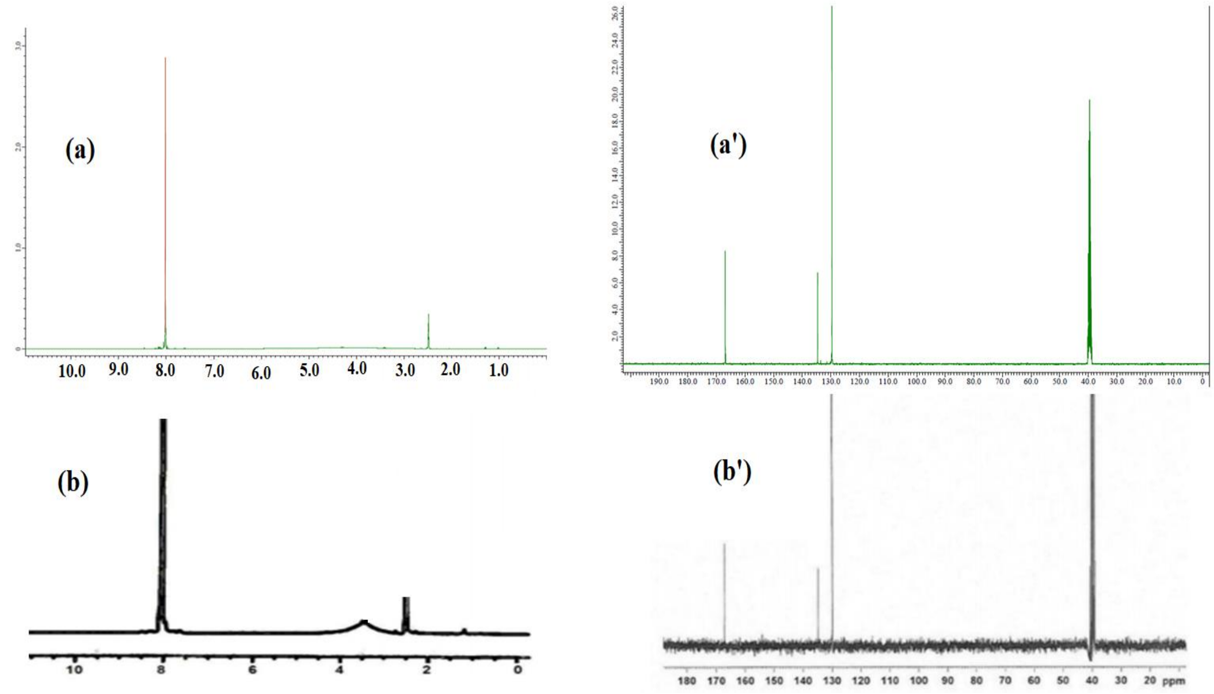


Figure 2. (a and a′) ^{1}H- and ^{13}C-NMR spectra of the prepared terephthalic acid and (b and b′) of the reported sample

3.2.1 IR analysis

The main characteristic IR band of aluminium chloride is the Al-Cl bond stretching located at 610 cm^{-1} as depicted in **figure 3** [36]. The other bands belong to water molecules, which are the O-H stretching broad band at 3500 cm^{-1} and the O-H bending (Scissoring) which appears at 1622 cm^{-1}. Furthermore, a smaller band is located at 2404 cm^{-1}, which is the result of coupling of the scissors-bending and another broad liberation band at 812 cm^{-1}[37].

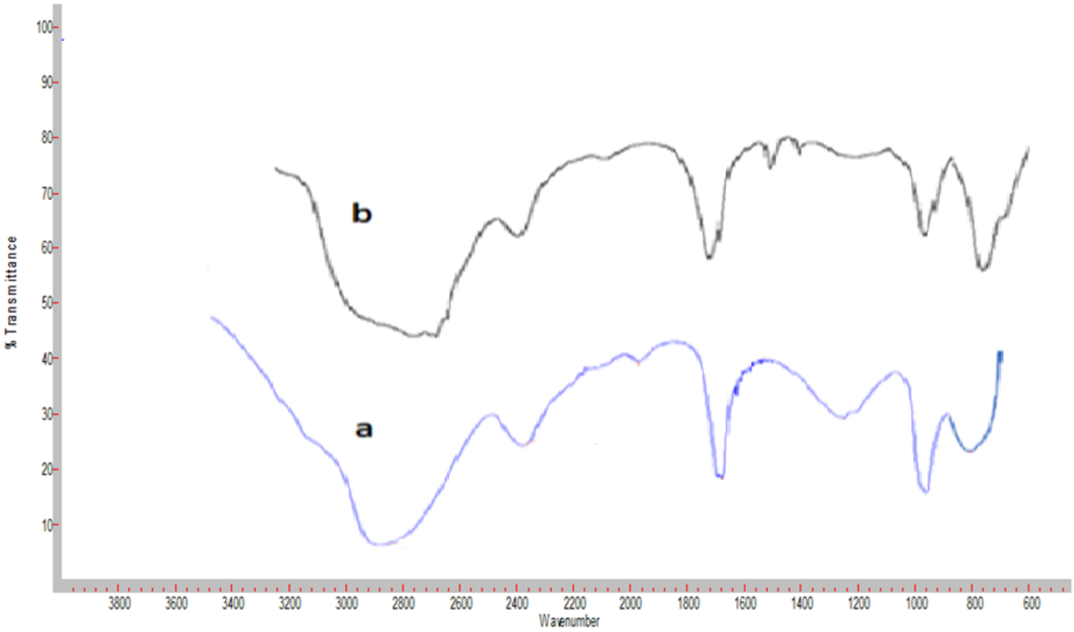


Figure 3. (a) IR spectra of the prepared $AlCl_3.6H_2O$ and (b) of the reported one.

3.2.2 XRD and SEM analysis

The XRD pattern of the prepared aluminium chloride hexahydrate is in accordance with the reported pattern [38]. The pattern showed coinciding 2Θ values at 8.4°, 9.2°, 10.1° and 28.1° (**Figure 4**). The SEM image of the prepared $AlCl_3.6H_2O$ shows granular particles with a size of about 233 µm.

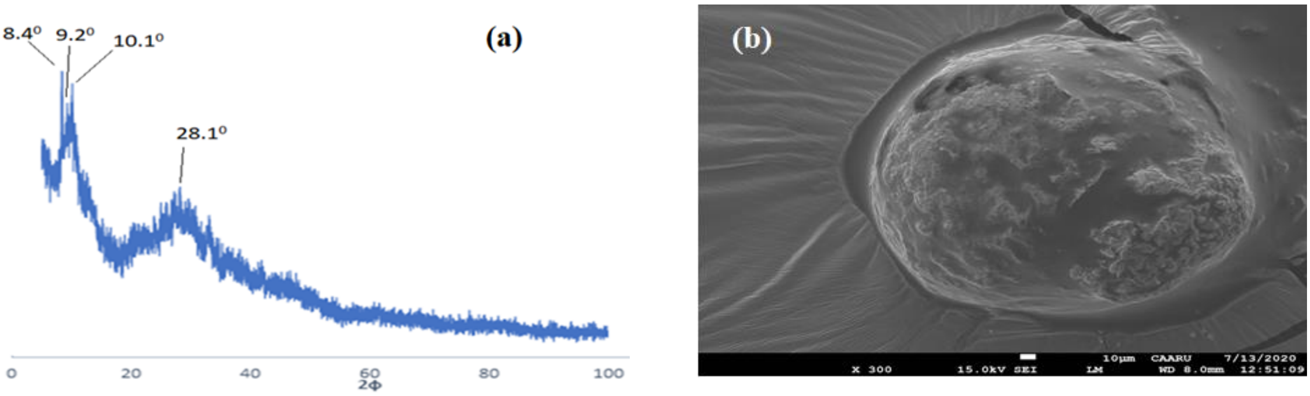
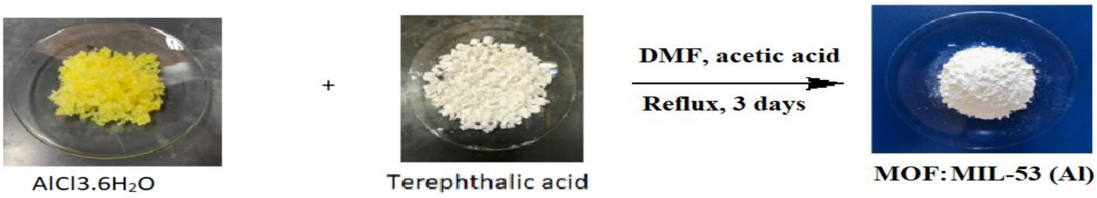


Figure 4. (a) XRD pattern and (b) SEM image of the prepared $AlCl_3.6H_2O$.

3.3 Preparation of aluminium-terephthalate framework, MIL-53 (Al)



Scheme 4. Synthesis of MIL-53 (Al) from hydrated aluminium chloride and terephthalic acid.

The reaction of terephthalic acid with $AlCl_3.6H_2O$ was carried out in a refluxed DMF for 3 days. A white MIL-53 (Al) solid formed in the reaction flask after 5 hours. TLC study showed total consumption of terephthalic acid. The prepared white solid polymer showed good thermal stability and resisted melting even after heating to above 300 °C. Elemental analysis of aluminium in the resulting solid was performed in ICP-OES and it was found to contain 13.82% of Al. Other elemental analyses for carbon, hydrogen and oxygen were carried out in a EuroEA elemental analyser and they were found to constitute 39.08%, 2.64%, and 44.45%, respectively.

3.3.1 IR analysis

The IR spectrum of the prepared MIL-53 (Al) revealed the expected characteristic absorption bands, such as C=O stretching bands at 1695 cm^{-1}, a C = C stretching band at 1594 cm^{-1}, a C-O stretching band at 1400 cm^{-1}, and an aromatic C-H bending band at 751 cm^{-1} (**Figure 5**). In addition, the stretching band of the Al-O single bond appears at 600 cm^{-1}, which is in agreement with the reported absorption [39].

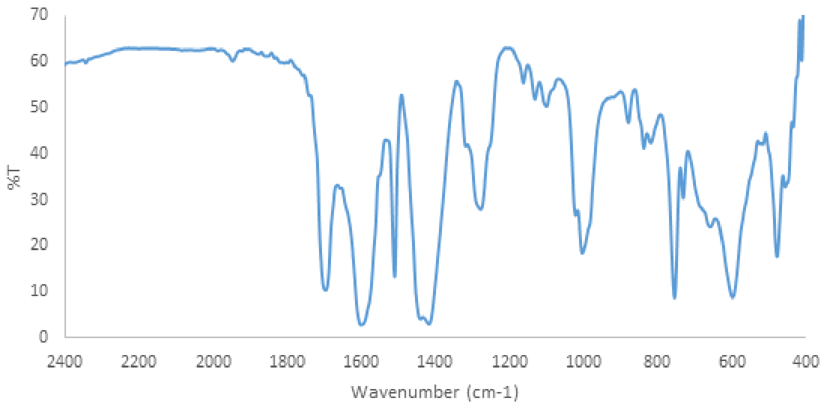


Figure 5. IR spectrum of MIL-53 (Al).

3.3.2 XRD and SEM analysis

Figure 6 illustrates the XRD pattern and SEM image of MIL-53 (Al). The main 2Θ values found in the XRD graph are: 9.5°, 15.3°, 17.9 ° and 20.6° which are comparable to the values in the literature [40]. SEM images of MIL-53 (Al) reveal an irregular brick-like morphology of the crystallites, with a size distribution of 100 to 500 nm (Figure 6(b)).

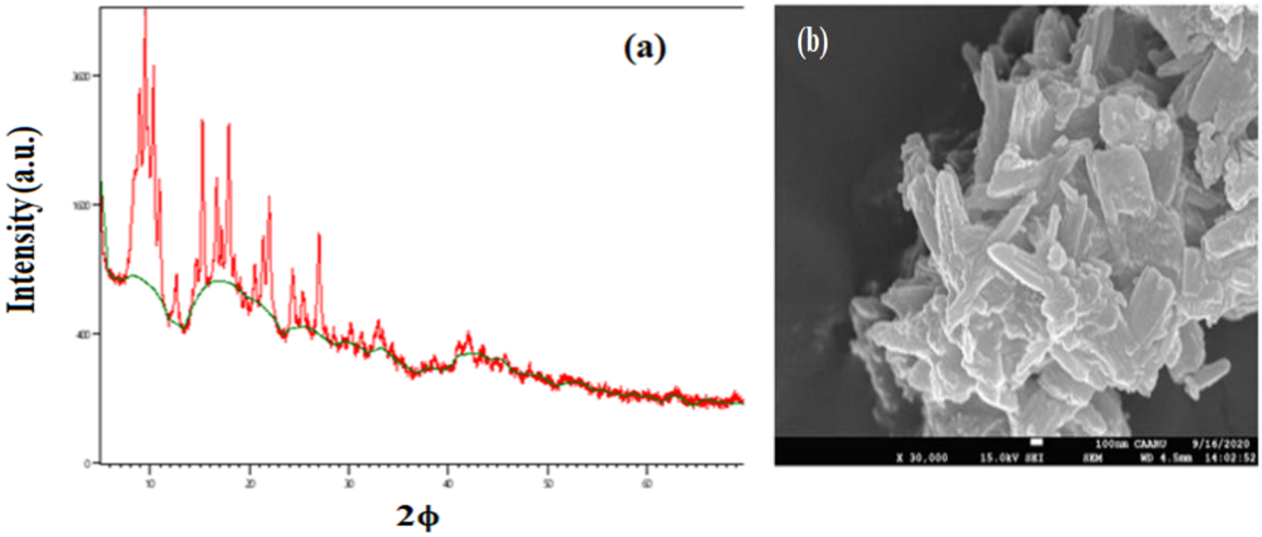


Figure 6. (a) XRD pattern and (b) SEM image of the prepared MIL-53 (Al).

4. Conclusion

In this study, an aluminium MOF (MIL-53 (Al)) material was prepared from recycled PET drinking water bottles and aluminium scraps. The characterization of the prepared substance was carried out using elemental analysis, IR, NMR, XRD, and SEM techniques. Investigation of the use of this material as a gas and pollutant absorbent is in progress.

Conflict of interest

The authors declare no conflict of interest.

Acknowledgment

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