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# New Polymeric Composites Derived from Waste PET: Synthesis, Characterization, and Antimicrobial Activity

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## Abstract

Many chemical, physical, and biological methods have been applied for plastic pollution as a global environmental problem. Polyethylene terephthalate (PET) is a commonly used polymer for many human life-related applications (food and water containers). In this paper, PET was hydrolyzed in a alcoholic NaOH medium at 110°C to obtain sodium terephthalate (Na-Terephthalate) that refluxed with glycerine and polyvinyl alcohol (PVA). PVA-terephthalate polymeric composite formed a flexible thin film. Other composites were prepared using PVA, Na-Terephthalate, and glycerine and $NiCl_2.6H_2O$, $ZnSO_4.7H_2O$, $Sr(NO_3)_2$, or $FeCl_3$. All PVA-terephthalate composites were characterized using FTIR, XRD, SEM, and DTA. Additionally, all prepared composites were screened for their microbial activity against *E. coli, S. aureus, P. aeruginosa*, and *Candida albicans*. The PVA-terephthalate composite showed a good to moderate inhibition zone towards *S. aureus, P. aeruginosa*, and *Candida albicans*. All metallic composites showed a negative response against *P. aeruginosa* and *Candida albicans*. The Zinc composite exhibited a noticeable inhibition zone against *E. coli* and *S. aureus*. These newly prepared composites (PVA-terephthalate, Nickel, and Iron composites) were evaluated for endo- and/or exothermic DTA peaks. The exothermic temperature of the Nickel composite was higher than the Iron composite, while PVA-terephthalate did not show any exothermic reaction. The obtained characteristics suggest promising materials for industrial applications such as seedling planting bags, food coating, and packaging materials, starting with minimizing PET quantity in the environment. The addition of metal salt to the blend composition may be a good suggestion in other research fields such as optical or electric applications.

## 1. Introduction

Plastics are commonly used in agriculture, packaging, building, medical, absorption … etc. according to their characteristics: rigidity, crystallization, mechanical, thermal, biological properties. Plastic pollution has been considered as global environmental problems [1 -4]. Many chemical, physical, biological methods were applied to solve this problem. Besides the environmental impacts of polymer pollution, recycling or degradation processes are considered crucial factors, especially costs, types of decomposition products from sunlight, oxygen, and heat, and their influences on the environment. [5-12]

Polyethylene terephthalate (PET) is a common polymer in modern-day community used for textile, water and food packing. A global crisis of daily PET waste found in natural water sources and landfills. Degradation of any polymers ensures obtaining the raw materials that formed PET followed by synthesis of new materials as next steps [12-15].

Polyvinyl alcohol (PVA) is another synthetic polymer that has various applications in food, medical, industrial, construction, and other fields because of its binding and film-forming capabilities. It is a non-toxic, water-soluble, biodegradable polymer with excellent crystallinity, thermal properties, emulsification, hydrolysis, and adhesion properties. These properties and applications reflected its molecular weight (or degree of polymerization), viscosity, and chemical composition of aliphatic chain with pending hydroxyl group that influence polymer water solubility, thickness of the formed film or coating, resistance to chemicals especially organic solvent, oxygen or other gases [16-19]. The presence of the intra- and inter-hydrogen bonding between hydroxyl groups plays a significant role in eco-friendly composite science and technology by considering both its advantages and disadvantages such as surface activity, porosity, water (or acid, alkali, or organic solvent) solubility or swelling behavior, magnetic, structural stability under forced conditions. PVA matrix interactions to other constituents such as organic aldehydes, acids, polyols, micro- or nano- cations exhibited numerous characteristics such as dielectric, adhesion, dispersion, magnetic, tensile strength and others [16-23]. In many research articles, various materials were added as cross-linkers that enhanced flexibility for more flow character depending on the effective distribution, crystallinity, size, chemical structure …etc. Boric acid, citric acid, glycerine and other biodegradables were operative in enhancement of PVA matrix towards films or coating blends [16-27].

Glycerine (1,2,3-Propanetriol, trihydroxypropane, or glycerol) is a triols produced from natural sources or by chemical synthesis used to bind and hold water molecules through hydrogen bonding. Its role is varied in cosmetics, pharmaceutical preparations, solvent, plasticizer, sweetener, in the manufacturing of dynamite, soaps, printing materials, lubricants, glues, fabrics industry, anti-freezing, and other industrial applications [28-32]. Glycerine and PVA are polyols materials that present a three dimensional network. This 3D-network in the polymeric matrix or formed blend or bed gel is reflection of hydrogen bonding between hydroxyl groups or other groups that form this crosslinking matrix. The formation of such a 3D configuration enhanced polymeric characteristics such as transparency, plasticity, thermal properties and others. In this subject, various natural or industrial polymeric combinations were prepared and studied such as cellulose, polyacrylamide, and PVA [24, 28-36].

In this study, we aimed to prepare new blends starts from alkaline hydrolysis of waste PET then mixing the produced terephthalate salt with PVA and glycerine under heating condition (Figure 1). Another preparation step involved mixing water-soluble metal salt of Nickel, Iron, Zinc, and Strontium with terephthalate, PVA, and glycerine mixture as one-pot step. In a primary step, the prepared blends were subjected to various characterization techniques to specify their spectral, thermal, and biological properties.

## 2. Experimental Procedure

### 2.1. Materials

Waste polyethylene terephthalate (PET) was obtained from water cups after use from local market. Nickel chloride ($NiCl_2.6H_2O$, BDH, England), zinc Sulfate ($ZnSO_4.7H_2O$, Riedel-de Haën, Germany), strontium nitrate ($Sr(NO_3)_2$, Perak, Germany), ferric chloride ($FeCl_3$, anhydrous, BDH, England) were used without any further purification. Glycerine was made in UAE (AMEYA FIC) while absolute ethanol was purchased from Hyman, GCC, India.

### 2.2. Instrumental Characterization

FTIR spectra were utilized with Infrared spectrophotometer, Shimadzu, Japan while X-Rays Diffraction (XRD) recorded by Lab X (XRD-6000), Shimadzu, Japan where its operational details were Voltage: 40.0 kV, Current:

30.0 mA, Speed: 5 deg. / min. with Cu-Kα radiation target at Wave: 1.54060 Å. Scanning Electron Microscope (SEM) type VEGAS III Tescan Algiua origin was used to show the configurations linked to each preparation step.

## 2.3. Preparation Steps

### 2.3.1 Degradation Step of Waste PET [37]

Waste PET was degradation by alkaline medium according to Japanese article. Alkaline hydrolysis (10% NaOH) of waste (cutted pieces PET, total weight 2 grams) resin in water-ethanol (20:80) wt% at 110oC was done for 0.5 hour. Residual PET was filtered then the liquid filtrate was dried and washed with water then dried to obtain sodium terephthalate (Figure 1).

### 2.3.2. One-Pot Synthesis of PVA – Terephthalate Polymeric Composite

2 grams (PVA), 2 grams (sodium terephthalate), and glycerine (4 grams) in (30 mL) water: (30 mL) ethanol medium were mixed and refluxed for five hours then the mixture was poured in three Petri dishes.

### 2.3.3. One-Pot Synthesis of Iron Polymeric Composite

2 grams (PVA), 2 grams (sodium terephthalate), glycerine (4 grams), ferric chloride ($FeCl_3.6H_2O$, 1gram) were mixed and refluxed for five hours then the mixture was poured in Petri dish. The resulted film was red dark flexible thin film.

### 2.3.4. One-Pot Synthesis of Metal Polymeric Composite

The same methodology of one-pot synthesis of iron polymeric composite was applied with the presence of metal salt (one gram).

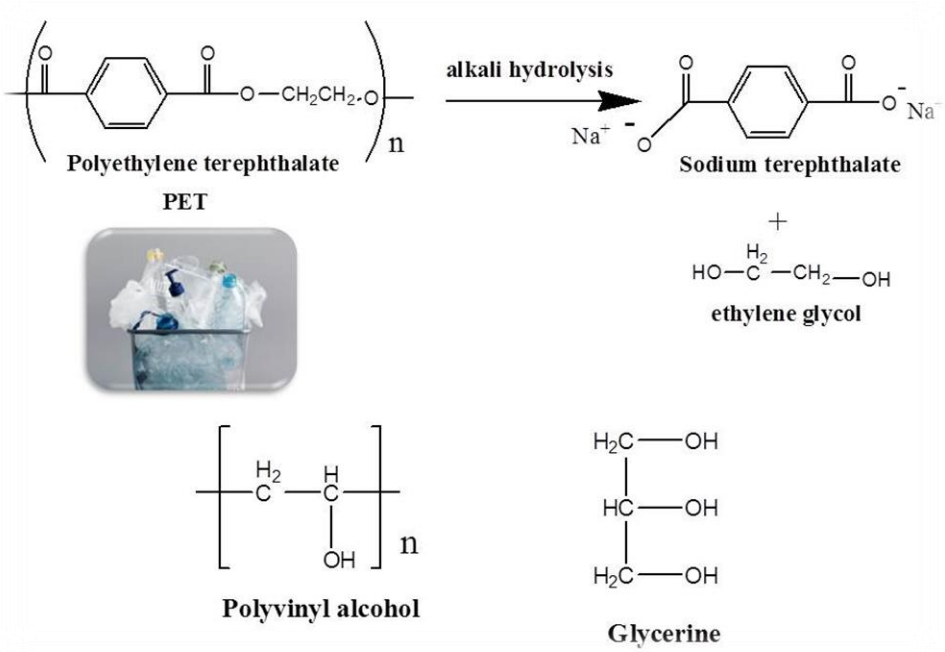

**Figure (1):** Alkali hydrolysis scheme of PET and structures of PVA and glycerine.

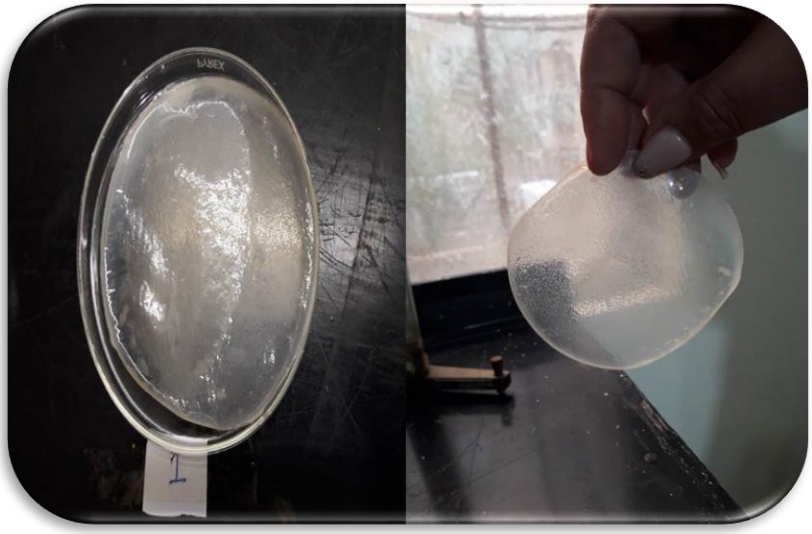

**Figure (2):** PVA – terephthalate polymeric composite (flexible hydrogel).

### 2.3.5. Anti-Biological Assessment of the Prepared Blends

All prepared blends were subjected to biological assessment against E. coli ATCC 10536, S. aureus ATCC 6538, P. aeruginosa ATCC 14442, and Candida albicans ATTC 10231. The semi-flexible films were cut into disks using a paper puncher with a diameter of 5.5 mm. Then, these discs were placed onto the Mueller-Hinton agar medium (Sigma-Aldrich, Germany) of the tested biological species and incubated at 37°C for 24 hours. The diameter of the inhibition zone was measured as an indicator of antimicrobial activity.

## 3. Results and Discussion

A long -life methodology of non-toxic and biodegradable materials made from waste or recycled materials is considered a green approach. Eco-friendly research based on plastics has become a part of the modern material industry and presents new materials with very unique properties. Depending on waste PET in any synthesis method presents a free charge step that reduces total costs. In this work, PVA–terephthalate – glycerol transformed from flexible aqueous Ethanolic solution into a semi - hard film with time at room temperature, due to the formation of PVA crystallites having a network of hydrogen bonding between hydrogen donor and acceptor centers presented in PVA and glycerol structures. The function of glycerol as a plasticizer improves the elongation at the break of the film [17] and gives a semi-flexible form beside on the stronger intra- and inters-hydrogen bonding between polyvinyl alcohol and glycerol molecules. These shorter interactions may be broken due to transformation from the semi-liquidized form to the dried form and the disordered PVA and glycerol. The intra Hydrogen bonding can increase the folding of PVA chains while the inter hydrogen bonding led to reposition of hydroxyl groups. These transformations had directly influence on the tested properties especially crystallization and thermal characters [38, 39]. These remarks can be observed on FTIR, XRD, and DTA. In this work, various components were mixed leading to physical and chemical changes presented in their characteristic peaks. FTIR spectrum of PVA – terephthalate sample (Figure 3) showed a broad band on ((3500-3300) and 2939) cm$^{-1}$ related to stretching vibrations of (OH, hydrogen bonding), aliphatic C-H of the alkyl groups respectively.

The peaks at (1107 and 1047) cm$^{-1}$ indicate C-O stretch while other peaks at (1651and 1560) cm$^{-1}$ can be attributed to symmetrical and non-symmetrical C=C stretching vibration in the phenyl ring (aromatic system). In addition, peak at 837 cm$^{-1}$ is related to C–C stretching mode. The bending vibration due to hydrogen bonding between hydrogen bonding sources was noticed at 1728 cm$^{-1}$ while 923 cm$^{-1}$ referred to C-H rocking vibrational mode of PVA – glycerine- terephthalate sample. This rocking mode indicates a successful blending of the three components. FTIR spectra of the prepared Zinc, Nickel, and strontium composites (Figures 3-6) showed the same important vibrational modes with little (blue or red) shifts confirming that the metal salt did not make chemical changes in the PVA –glycerine – terephthalate blend [40].

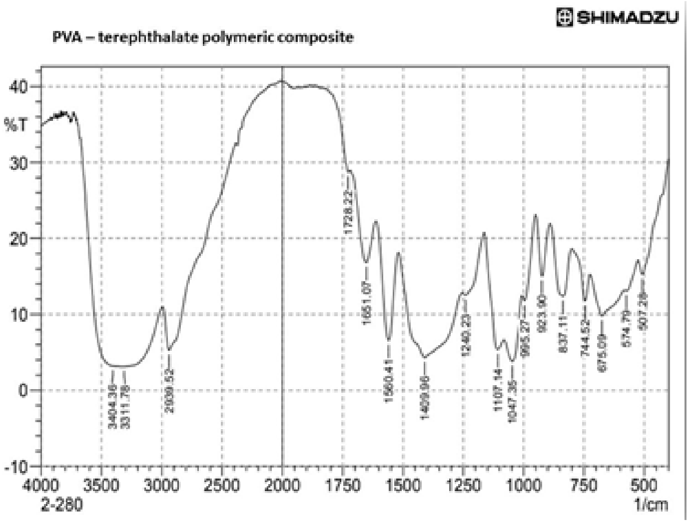

**Figure (3):** FTIR spectrum of PVA – terephthalate polymeric composite.

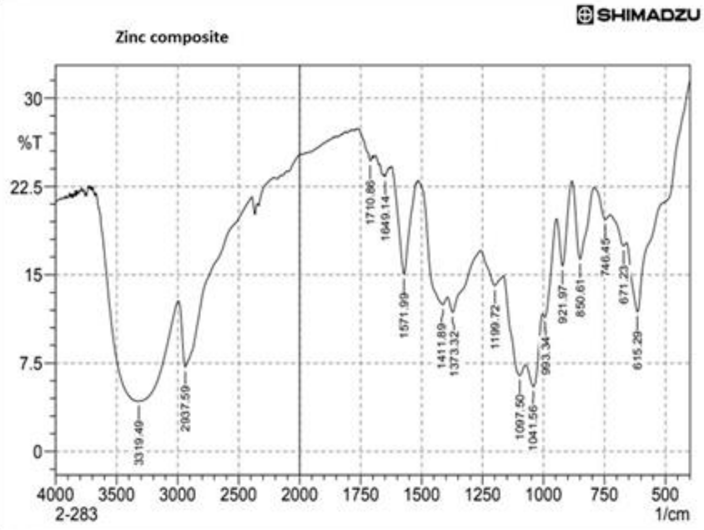

**Figure (4):** FTIR spectrum of Zinc composite.

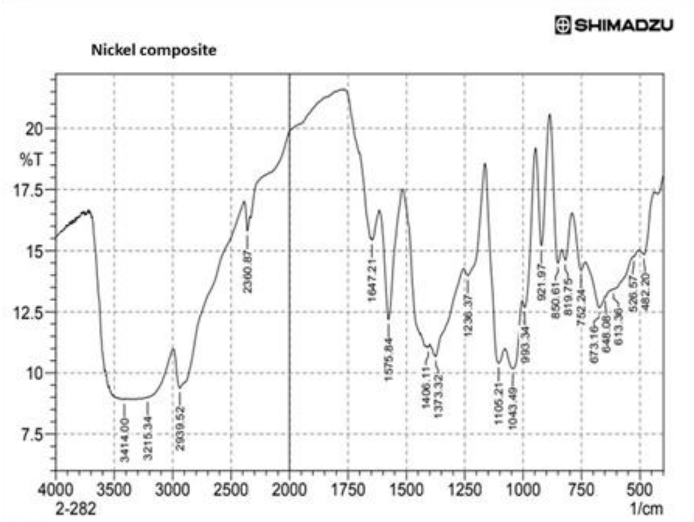

**Figure (5):** FTIR spectrum of Nickel composite.

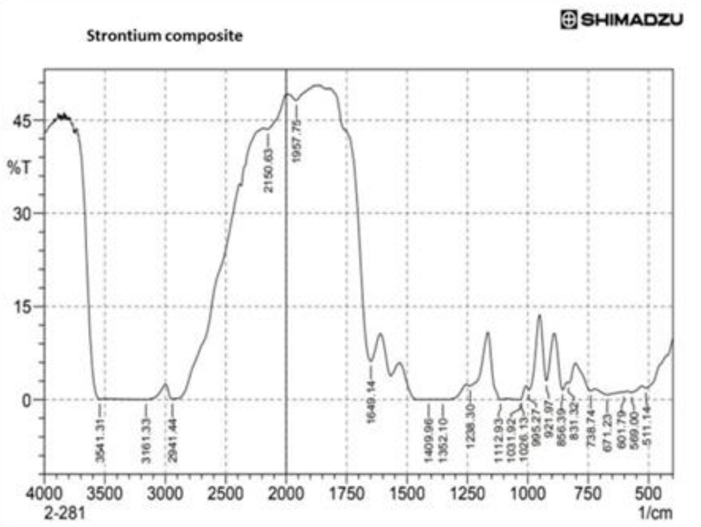

**Figure (6)**: FTIR spectrum of Strontium composite.

From the obtained XRD patterns, the average crystallite size is estimated from the diffraction line broadening of the three strongest peaks. The Scherrer formula ($D = k\lambda/\beta \cos(\theta)$) determines this important characteristic (Table 1), where d is the crystallite size, $\lambda$ is the wavelength of the X-ray (Cu-K$\alpha$, 1.54056 Å or 0.154056 nm), k is the shape factor ($k = 0.91$), $\theta$ is the diffraction angle of the peak, and $\beta$ is the full width at half - height of the peaks (FWHM, in radians ) of the tested sample.

Gupta et al. prepared PVA hydrogels with a relatively broad peak at 22.5° (no sharp X- ray diffraction peaks) while the monoclinic unit cell of the PVA powder exhibits strong crystalline reflections at two angles ($2\theta$ (hkl) = 19.88° (101) and 20.18° (101)) in an atactic trans-planar conformation. These researchers claimed that the low degree of crystallinity and high scattering indicate the presence of a nanocrystalline phase dispersed within the bulk amorphous phase [41].

Kovtun et al. studied various weights of glycerol in PVA films, and their XRD diffractograms were compared with pure PVA, which has characteristic peaks at 11.2°, 16.1°, 19.4°, 20.1°, 22.5°, and (40-42)° associated with the 100, 001, 101/101, 200, and $(1\bar{1}1)$/(210) planes, respectively. The authors attributed their XRD patterns to being semi-crystalline. Additionally, the broad amorphous regions of disordered non-crystalline areas were reduced due to a polymeric rearrangement influenced by the addition of glycerol. The addition of glycerol led to a decrease in the peak at 22.5° compared to pure PVA, with no change at (40.7°) [42]. In another research article, Thavornsetawat et al. hydrolyzed post-consumer PET bottles using alkaline treatment to produce terephthalic acid salt, which exhibited characteristic peaks at 17.3°, 25.2°, 27.9°, 31.8°, and 35.1° [43].

As previously mentioned, PVA is classified as a semi-crystalline polymer (Figure 7a) showing the unique XRD diffraction peaks associated with its crystal domain. The most prominent peak of the XRD pattern generally corresponds to a $2\theta$ value ≈ 19.5° for the (101) plane confirming semi-crystalline characteristics. We can also notice small peaks around 11.4° and 22.8°, which are related to the (100) and (200) crystallographic planes, respectively. The exact position and intensity of peaks will depend on the composition ratio between components, processing conditions, and their polymers degree of crystallinity. To that end, the terephthalate is combined with PVA; an additional peak may appear depending on the degree of crystallinity of the terephthalate phase $2\theta \approx 26°$ (010) plane.

The peaks of the PVA-Fe composite were located at $2\theta$ = 30.1°, 35.5°, 43.1°, 53.4°, 57.0°, and 62.6° that can be indexed to (220), (311), (400), (422), (511), and (440) planes, besides pure PVA peak as shown in Figures (7b) and Figure (8).

Among the PVA-Zn composite (Figure 7c and Figure 9), one can see a similar combination of the broad peaks from the PVA and the sharper peaks corresponding to the crystallinity of the Zn compound at 2θ ≈ 31.8° from the (100) plane and 34.4° (002), 36.3° (101), 47.6° (102), 56.6° (110) and 62.9° (103).

The PVA-Ni composite (Figure 7d) is semicrystalline, and its contribution to the XRD pattern is generally weak and broad at around 2θ = 19.5°. This peak is consistent with the accumulation inter-planar spacing of the polymer chains and referencing common diffraction peaks on the JCPDS card No. 47-1049 m for the Miller index diffract peaks as it (111) 37.2° (200) 43.3°, (220) 62.9°, (311) 75.4° respectively.

Lastly, a PVA-Sr composite displays a wide and amorphous peak at approximately 2θ = 19.5° as a consequence of the semi-crystalline nature of PVA and the peaks at 2θ ≈ 32.9°, 38.3°, 55.2°, 64.8° as a cubic phase (card No.01-074-1258), as shown in Figure (7e).

The XRD of PVA-terephthalate (Figure 7a) presented peaks at 22. 4725°, 25. 2531°, 27. 892°, 31. 7412°, and 35. 0379° with low intensities (6, 6, 14, 6, and 3), respectively. Additionally, Table (1) and Figures (7-9) show that new characteristic peaks were not found in both references [42 and 43], indicating the formation of a new blend with distinct features. Strontium and zinc composites exhibited the lowest crystallite size at 2θ = 19° alongside various 2θ values related to each prepared sample, characterized by the X- ray diffraction technique (Figures 7-9, Table 1).

**Table (1)**: XRD data and Scherrer equation.

| Sample ID | 2θ, deg. | FWHM, deg. | D, nm | d-spacing, Å |
|---|---|---|---|---|
| PVA – terephthalate | 33.5159 | 0.2783 | 30.1439 | 2.6716 |
| | 28.7082 | 0.3477 | 23.84708 | 3.107127 |
| | 27.6119 | 0.3131 | 26.4189 | 3.227952 |
| **Iron composite** | 31.6556 | 0.1638 | 50.97275 | 2.824228 |
| | 31.4416 | 0.0978 | 85.32651 | 2.842961 |
| | 45.3457 | 0.1584 | 54.95903 | 1.998346 |
| **Strontium composite** | 19.6638 | 1.23 | 6.628069 | 4.51106 |
| | 20.3618 | 0 | - | 4.357969 |
| | 21.3092 | 0 | - | 4.166307 |
| **Nickel composite** | 31.6948 | 0.2768 | 30.16671 | 2.820825 |
| | 45.392 | 0.2404 | 36.21872 | 1.996415 |
| | 56.3901 | 0.2175 | 41.90429 | 1.630352 |
| **Zinc composite** | 19.9131 | 1.25 | 6.524495 | 4.455143 |
| | 20.8604 | 0 | - | 4.254918 |
| | 49.368 | 0.3055 | 28.93799 | 1.844529 |

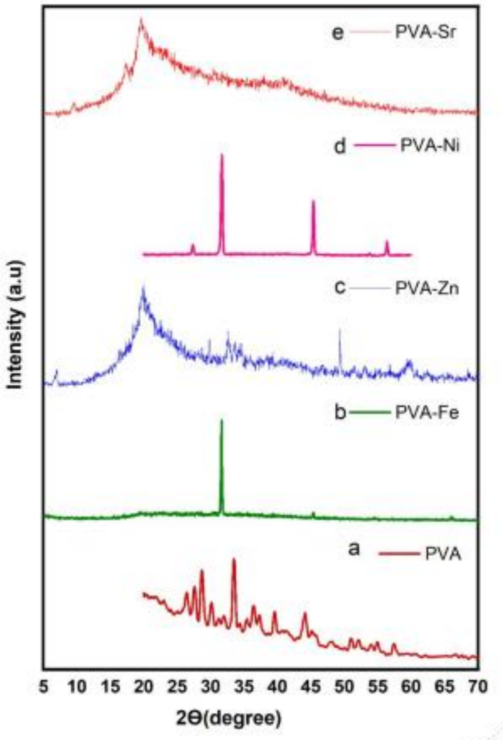

**Figure (7):** X-Rays diffractograms of all samples.

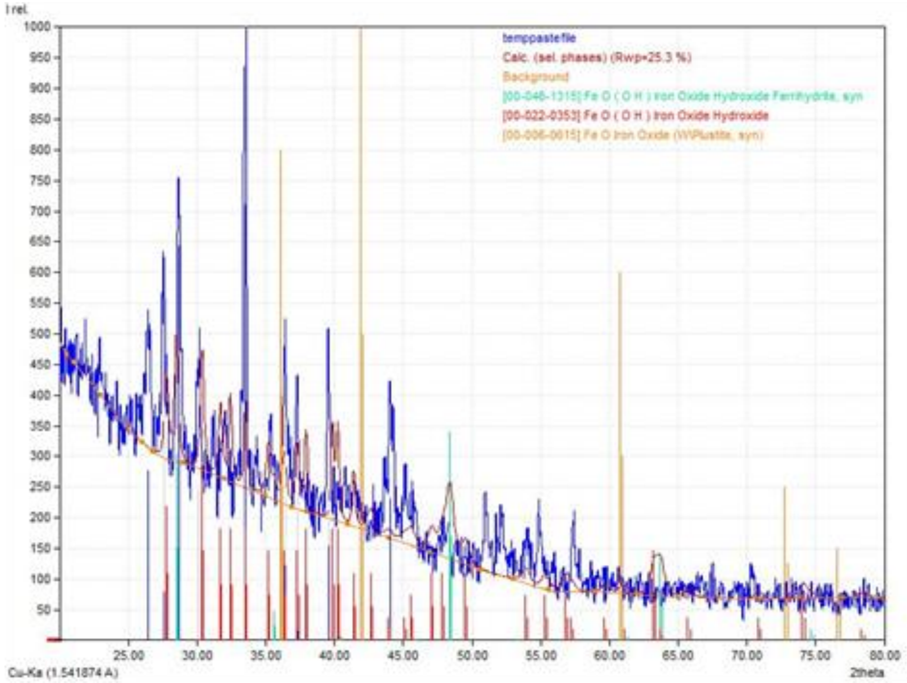

**Figure (8):** X-Rays diffractogram of iron composite compared with JCPDS cards.

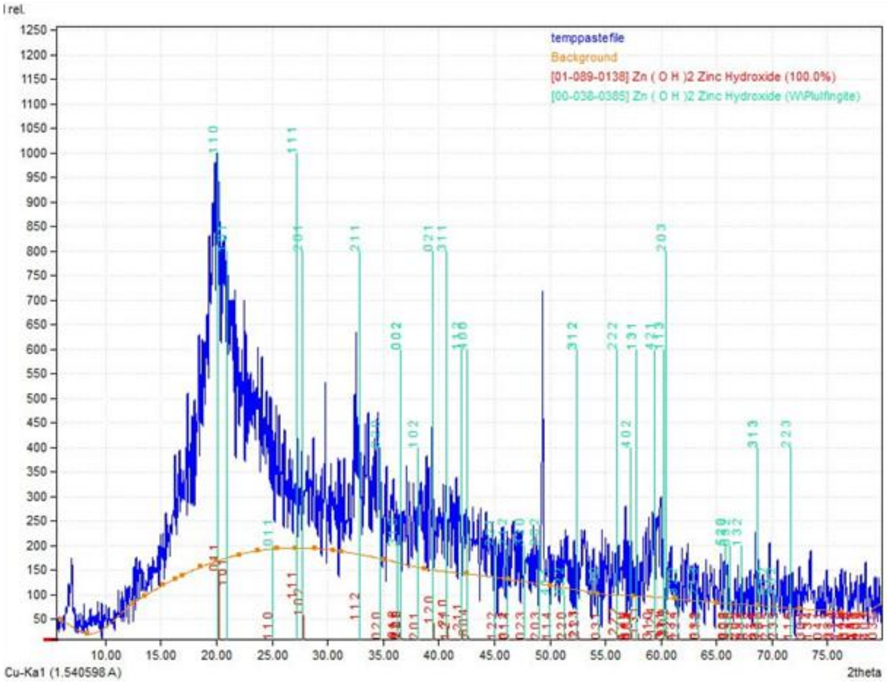

**Figure (9):** X-Rays diffractogram of Zinc composite compared with JCPDS cards.

Scanning Electron Microscope (SEM) images of the prepared samples was another spectral characterization used to identify the newly blends. SEM images displayed semi- cubic structure with high meters (out range of nano-scale) (Figures 12-14). These semi–cubic structures were more obvious with metal composites compared to PVA – terephthalate reflecting the influence of the metal in the final formed structure. The observed dimensions in all SEM images (Figures 12-14) were non-compatible with XRD-Scherrer equation results (Table 1). So, nano-structural features were absent in all tested samples according to SEM images (Figures 10-14).

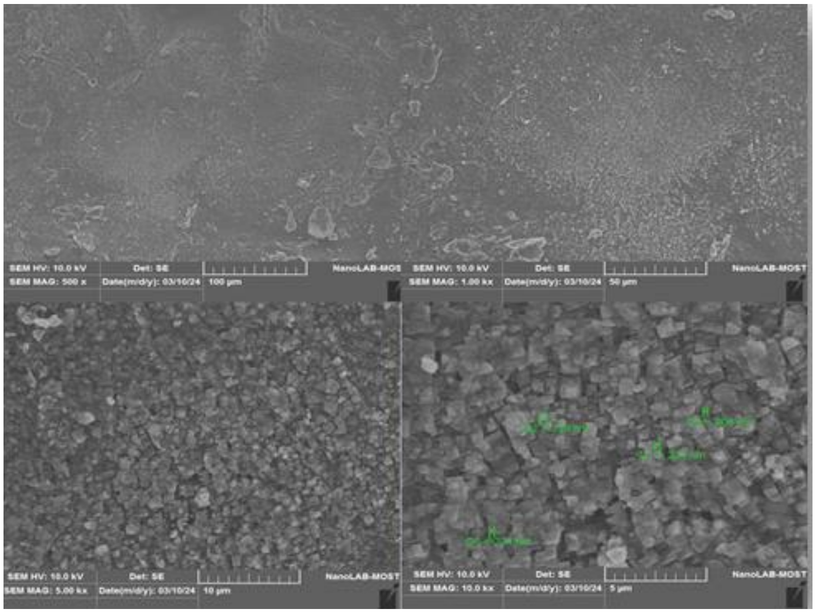

**Figure (10):** SEM images of PVA – terephthalate sample.

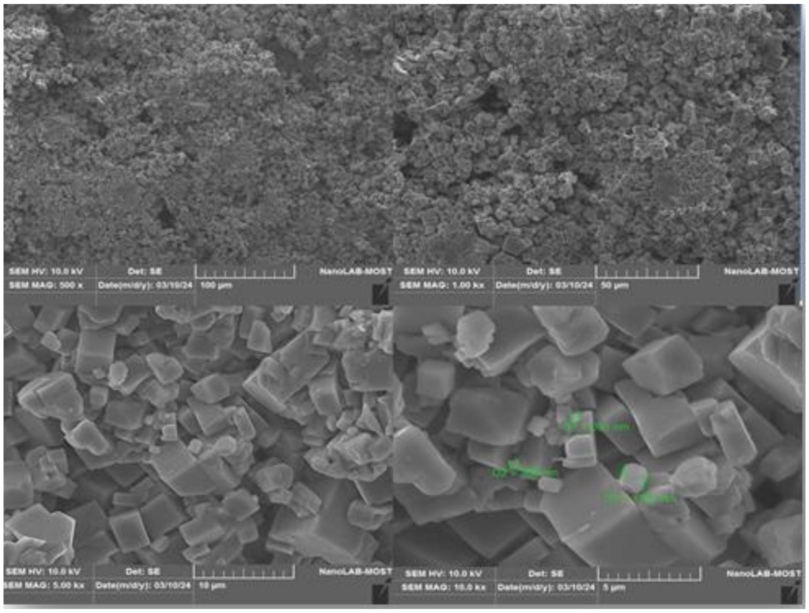

**Figure (11):** SEM images of Nickel composite.

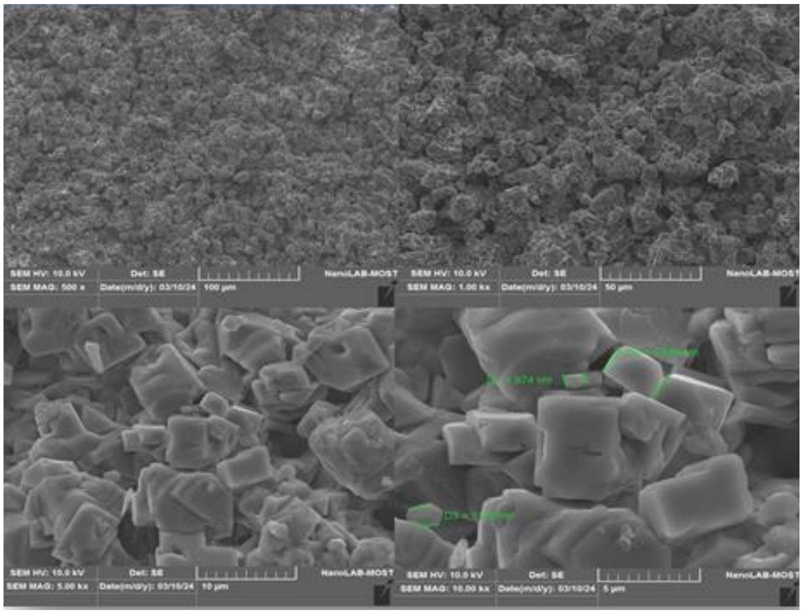

**Figure (12):** SEM images of iron composite.

According to DTA, these newly prepared composites (PVA-terephthalate, Nickel, and Iron composites) were evaluated for endo- and/or exothermic peaks. The exothermic temperature of the Nickel composite was higher than the Iron composite, while PVA-terephthalate did not show any exothermic reaction (Table 2, Figures 15-17).

Firstly, PVA – terephthalate sample (Figure 15) showed two endothermic peaks that absorbed (5.1 and 3.19) kJ/g. The presence of nickel and iron exhibited an exothermic point besides the endothermic point(s) (Figures 16 & 17) where the released heat varied in their values with maximum released heat at 3.17 kJ/g in DTA of Iron composite. In temperature scale, Nickel composites revealed the highest temperature in both endothermic and exothermic points compared to PVA –terephthalate and iron samples. Also, the addition of metal salt to PVA – terephthalate dropped the first endothermic temperature compared to non- metallic sample (Table 2, Figures 13-15).

**Table (2):** Thermal analysis (DTA) results of the prepared samples.

| Sample ID | Peak information | | | | |
|---|---|---|---|---|---|
| | Type | Time, min. | Temp., ºC | Heat, J (kJ/g) | Height, µV |
| PVA – terephthalate | Endothermic | 12.08 | 125.63 | -31.90 (-5.10) | -137.24 |
| | Endothermic | 23.27 | 245.26 | -19.94 (-3.19) | -108.09 |
| **Iron composite** | Endothermic | 9.13 | 116.05 | -10.44 (-2.02) | -52.47 |
| | Exothermic | 27.40 | 316.51 | 16.41 (3.17) | 126.44 |
| **Nickel composite** | Endothermic | 8.25 | 104.79 | -71.19 (-15.15) | -171.18 |
| | Endothermic | 21.88 | 248.05 | -11.20 (-2.38) | -51.34 |
| | Exothermic | 31.88 | 356.33 | 6.94 (1.48) | 34.46 |

The thermal behavior of the prepared blends is related on the crystalline structure, glycerol plasticization, and the absence of cross-linking chains in PVA. The role of temperature in changing characteristics mainly depends on the aggregation of PVA- glycerol -terephthalate, influenced by the formation of hydrogen bonds due to the degree of crystallinity. This degree is minimized by the presence of glycerol plasticizer, which disrupts the hydrogen bonding framework and increases mobility.

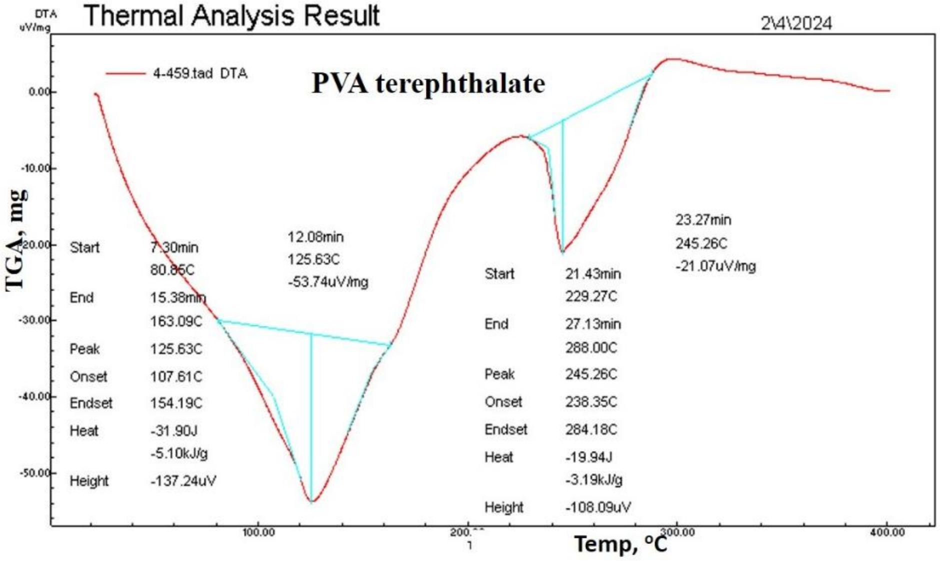

**Figure (13):** Thermal behaviour of PVA-terephthalate.

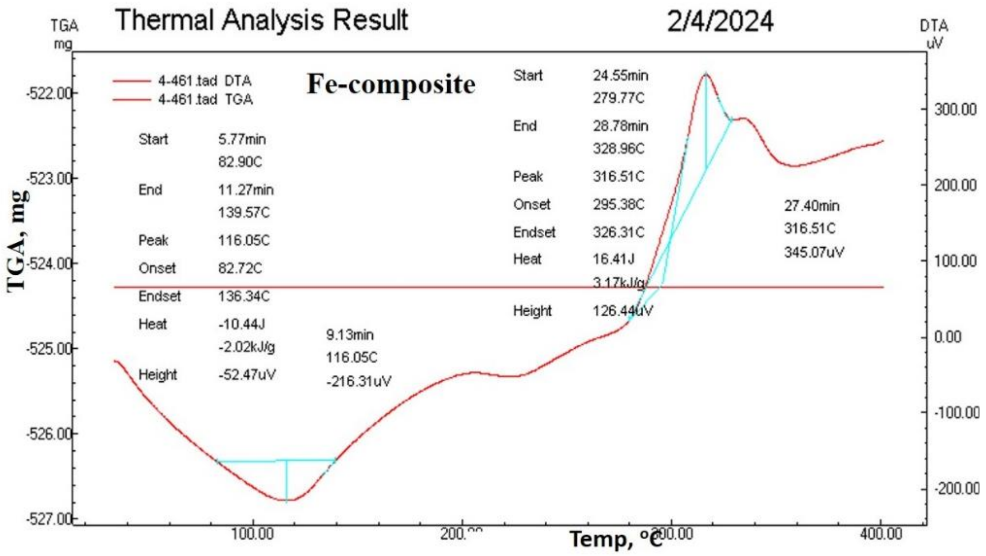

**Figure (14):** Thermal behaviour of Fe composite.

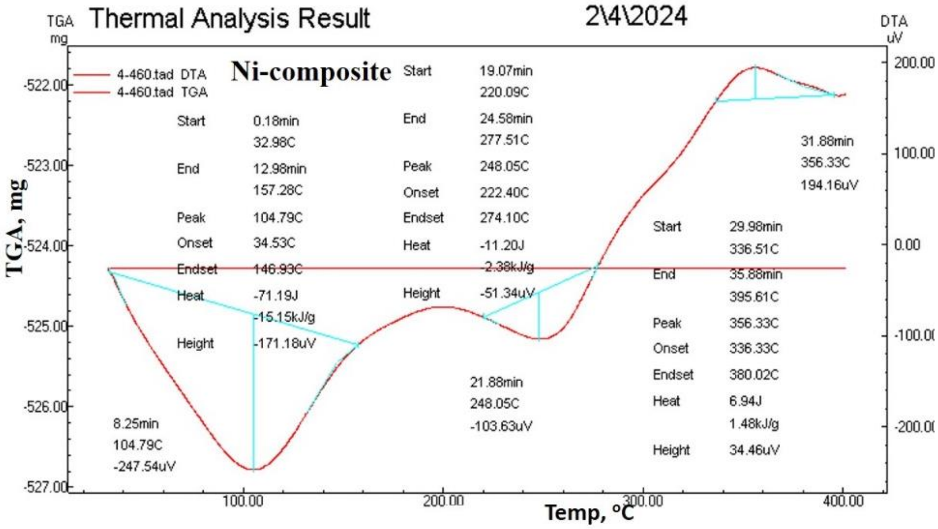

**Figure (15):** Thermal behaviour of Ni composite.

The prepared samples were subjected to microbial testing and were screened against *E. coli ATCC 10536*, *S. aureus ATCC 6538*, *P. aeruginosa ATCC 14442*, and *Candida albicans ATTC 10231* (Table 3, Figure 16). The PVA-terephthalate polymeric composite showed a good to moderate inhibition zone towards *S. aureus*, *P. aeruginosa*, and *Candida albicans*. All metallic composites showed a negative response against *P. aeruginosa* and *Candida albicans*. The Zinc composite exhibited a noticeable inhibition zone against *E. coli* and *S. aureus*. Our prepared blends presented a promising antimicrobial active packaging that differs from the traditional packaging materials in its ability against the growth of pathogenic microorganism on food surface [44].

**Figure (16):** Biological assessment of the prepared materials.

PVA was investigated as seedling bags that decomposed within a short time. The addition of glycerol as a non-toxic material to PVA increases the polymer flow and improves solubility, absorption, and evaporation.

Zinc oxide was selected by many researchers because of its antimicrobial effect, optical, electrical, dielectric, and thermal properties that adapt PVA characters [45]. Therefore, our blends had good antimicrobial properties and could be applied as a new food packaging material with antimicrobial activity. Also, the addition of metal salt to the blend composition may be a good suggestion in other research fields such as optical or electric applications such as UV sensing.

**Table (3):** Biological activity of PVA –Terephthalate samples.

| **Sample ID** | ***E. coli*** | ***S. aureuse*** | ***P. Arginosa*** | ***Candida albicans*** |
|---|---|---|---|---|
| PVA – terephthalate | Negative | 20 | 13 | 7 |
| **Iron composite** | Negative | Negative | Negative | Negative |
| **Strontium composite** | Negative | 11 | Negative | Negative |
| **Nickel composite** | 13 | Negative | Negative | Negative |
| **Zinc composite** | 27 | 10 | Negative | Negative |

## 4. Conclusions

Pollution reflects a global problem to all environmental species and when it related to plastic, it becomes more complicated. By chemical, physical, and biological methods, many types of plastic had been subjected to these methods to convert them to other beneficial materials. Polyethylene terephthalate (PET) is a commonly used polymer for many human life-related applications such as food and water containers. In this paper, PET was hydrolyzed in an alkali medium (chemical method). The obtained sodium terephthalate (Na-Terephthalate) was mixed with glycerine and polyvinyl alcohol (PVA) to form a new composite as a flexible thin film. Other composites were prepared using the same starting materials (PVA, Na-Terephthalate, and glycerine) and metal salts (Nickel chloride, Zinc Sulfate, Strontium nitrate, or Ferric chloride). PVA-terephthalate composite and its four metallic composites were characterized by (FTIR, XRD, SEM, and DTA) techniques. Additionally, all prepared composites were screened for their microbial activity against *E. coli, S. aureus, P. aeruginosa, and Candida albicans*.

The PVA-terephthalate polymeric composites showed a varied inhibition activity towards the tested microorganisms where all metallic composites showed a negative response against *P. aeruginosa* and *Candida albican* beside a noticeable inhibition zone against *E. coli* and *S. aureus* by Zinc composite. Thermal testing by DTA showed various endo- and/or exothermic peaks where the highest exothermic temperature found in the Nickel composite than the Iron composite, while PVA-terephthalate did not show any exothermic reaction. Here, waste PET hydrolysis was a start step to obtain encouraging materials for industrial applications such as seedling planting bags, food coating, and food packaging materials beside other applications that mainly depend on optical or electric applications such as UV sensing.

**Conflict of Interest:** The authors declare that there are no conflicts of interest associated with this research project. We have no financial or personal relationships that could potentially bias our work or influence the interpretation of the results.

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