

Synthesis of Amorphous Titanium Phosphate using Dihydrate Wet-Process Phosphoric Acid

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Abstract: Cost-effective synthesis of titanium phosphate using phosphoric acid produced from Abu-Tartur phosphate concentrate was achieved with titanyl sulfate. Five controlling factors that affect the synthesis conditions of titanium phosphate were thoroughly studied. The obtained results showed that the optimum precipitation conditions are 65 °C reaction temperature, 35.8 % H₃PO₄ phosphoric acid concentration, 30 minutes reaction time, 5 minutes phosphoric acid addition time, and 400 rpm stirring speed. Based on these conditions, amorphous titanium (IV) phosphate particles were obtained, which belongs to the class of tetravalent metal acid salt (TMA). The synthesized titanium phosphate was characterized using FTIR, XRD, TGA/DTA, SEM, EDX and Ion exchange capacity for Na⁺.

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Keywords: Abu-Tartur phosphate concentrate; Phosphoric acid; Titanium phosphate; Tetravalent Metal Acid Salt (TMA).

1. Introduction

The wet-process presents 90% of the world current phosphoric acid production (cheap in cost) (**Gonzalez et al., 2002; Krea and Khalaf, 2004**).

Wet-process phosphoric acid is an important intermediate product for production of fertilizers. It is mainly produced reaction of phosphate concentrate with sulfuric and weak phosphoric acids to produce phosphoric acid and calcium sulfate crystallization occurs as leaching is taking place. According to the process adopted, calcium sulfate dihydrate (gypsum) (CaSO₄.2H₂O) or calcium sulfate hemihydrate (CaSO₄.0.5H₂O) or calcium sulfate anhydrite (CaSO₄) is crystallized. Wet process phosphoric acid includes acids rather than sulfuric acid such as nitric acid and hydrochloric acid but to much less extent (**Abdel-Aal, 1984, 1989; El-Shall et al., 1999, 2000; Abdel-Aal, 2004**).

Synthesis and characterization constitute the most essential aspects of materials research. A combination of the two, leads to the preparation of tailor made materials, to perform a specific function with the desired properties as well as discovery of new materials and advanced synthesis methodologies.

Amorphous titanium phosphate as tetravalent metal acid salt (TMA) has been widely explored as an ion exchangers (**Pandit and Chudasama, 1998; Parikh and Chudasama, 2003; Maheria and Chudasama, 2006a**), solid acid catalysts (**Chudasama, 1999; Parikh et al., 1999; Patel et al., 2001; Patel et al., 2002**) and solid state proton

conductors (**Patel et al., 2005; Patel and Chudasama, 2006; Patel and Chudasama, 2007**).

The main objective of the present work is to determine the optimum conditions required for synthesis of titanium phosphate by reaction of titanyl sulfate with wet-process phosphoric acid produced from Abu-Tartur phosphate concentrate (New Valley, Egypt). Characterization of the produced titanium phosphate using FTIR, XRD, TGA/DTA, SEM and EDX is another goal of this study.

2. Experimental

Materials:

All chemicals used are analytical grad. Sulfuric acid ≥ 97.9 % was obtained from ADWIC, Egypt. Titanium dioxide and ammonium sulfate were supplied by (MERCK, Germany). Phosphoric acid produced laboratory from Abu-Tartur phosphate concentrate, by dihydrate process. The chemical composition of the produced acid is given in Table (1).

Apparatus:

The reaction was carried out in a covered 1 L beaker. It was fitted with Teflon-coated stirrer with 4 cm diameter and placed in thermostatically controlled water bath. The impeller tip speed was adjusted at 450 rpm. Filtration was performed using Buchner type filter of 8.5 cm diameter filter funnel with polypropylene filter cloth. A vacuum pump was used for filtration.

Table (1): Chemical analysis of phosphoric acid produced from Abu-Tartur phosphate concentrate by dihydrate process.

Component	Concentration
P ₂ O ₅	29.2 %
CaO	0.29 %
Fe ₂ O ₃	2.41 %
MgO	0.53 %
Al ₂ O ₃	0.35 %
SO ₄ ²⁻	2.19 %

Procedure:**Preparations of titanyl sulfate:**

Titanyl sulfate has been prepared by mixing 50 g of titanium oxide with 500 g of ammonium sulfate and 1.0 liter of sulfuric acid having a specific gravity of 1.825 (97.97%). The mixture is gradually heated to 200°C with continuous stirring (Marie, 1968). A clear solution of titanyl sulfate in highly concentrated sulfuric acid is obtained.

Synthesis of titanium phosphate:

For each run of titanium phosphate synthesis, a calculated amount of phosphoric acid was mixed with the proper amount of titanyl sulfate solution in the reactor. After the desired reaction time, the produced titanium phosphate gel was kept in contact with mother liquor overnight for aging, and then washed with hot distilled water (60 °C) until pH 3.8-4.2 followed by filtration and drying on dryer for 5 hours at 60 °C. After drying, the titanium phosphate has been weighed as white powder. During the reaction time or after certain time, the turbidity measurements (NTU) of the stirred solution were taken using Orbeco-Hellige Digital Direct-Reading Turbid meter.

Characterization of the synthesized titanium phosphate:

Titanium and phosphorus content in the obtained precipitate were analyzed by X-ray powder diffraction (XRD) analysis for the prepared precipitate was carried out using a Bruker AXS diffractometer (D8-ADVANCE) with Cu K α ($\lambda=1.54056 \text{ \AA}$) radiation, operating at 40 kV and 10 mA. The diffraction data were recorded for 2θ values between 5° and 70° and the scanning rate was 0.5° min⁻¹. SEM-EDX, a Scanning Electron Microscope and Energy dispersive X-ray spectroscopy was employed to evaluate the atomic composition of produced titanium phosphate. Fourier Transform Infrared Spectrum (FT-IR) analysis was performed using JASCO 3600 spectrophotometer and the resolution was set to 4 cm⁻¹ for all the samples. Thermal analysis (TGA/DTA) was carried out on a Shimadzu thermal analyzer at a heating rate of 10°C/min. Chemical resistivity in various media (acids, bases and organic solvents) was studied by taking 500 mg of the prepared titanium phosphate in 50 mL of the particular medium and allowing to stand

for 24 h. The change in its colour, nature and weight was observed.

3. Results and Discussion

Reaction of Abu-Tartur phosphoric acid with titanyl sulfate was carried out. The parameters affecting the reaction (precipitation) conditions such as reaction temperature, phosphoric acid concentration, reaction time, time of phosphoric acid addition, and stirring speed are thoroughly studied. Their effects on the titanium phosphate precipitate turbidity have been investigated.

a. Effect of Precipitation Temperature:

The effect of reaction temperature on titanium phosphate precipitation process was investigated for the temperatures ranging from 25 to 150 °C, at reaction time of 30 min, phosphoric acid concentration of 43.3% H₃PO₄, with phosphoric acid/ titanium oxide mass ratio, ml/ g, 5.0, phosphoric acid rate of addition of 10 min, stirring speed of 400 rpm and nucleation time of 30 min. The obtained results were plotted in Figure (1) as a relation between turbidity (NTU) and temperature. From the results, it is clear that, by increase reaction temperature from 25 to 65 °C the solution turbidity increased from 150 to 790 NTU. However further increase in the reaction temperature up to 150, the solution turbidity decrease to 300 NTU. This behavior may be attributed to the dissolution of titanium phosphate precipitate. The obtained results clear that the reaction temperature within the range from 25 to 65 °C has positive effect on the precipitation of titanium phosphate. Therefore, temperature of 65 °C is the preferred reaction temperature for the precipitation process.

b. Effect of Phosphoric Acid concentration:

The effect of phosphoric acid concentration on titanium phosphate precipitation process, have been investigating by conducting several experiments at different phosphoric acid concentration ranging from 25.5- 75.7 % H₃PO₄ while the other parameters were fixed at; 30 min reaction time, temperature of 65 °C, phosphoric acid rate of addition of 10 min, stirring speed of 400 rpm, and nucleation time of 30 min. The results obtained were presented in Figure (2) as a relation between turbidity (NTU) and phosphoric acid concentration. The results in Figure (2) indicated that, as the phosphoric acid concentration increases from 25.5 to 35.8 % the solution turbidity increased from 500 to 805 NTU. Further increase in the phosphoric acid concentration up to 75.7 % H₃PO₄ has a slightly decrease in the measured turbidity (NTU). This means that, the increase in phosphoric acid concentration up to 35.8 % H₃PO₄ is enhancing titanium precipitation process. Accordingly the phosphoric acid concentration of 35.8% H₃PO₄ is the choice concentration for the titanium phosphate precipitation

process from titanium oxide by Abu-Tartur phosphoric acid.

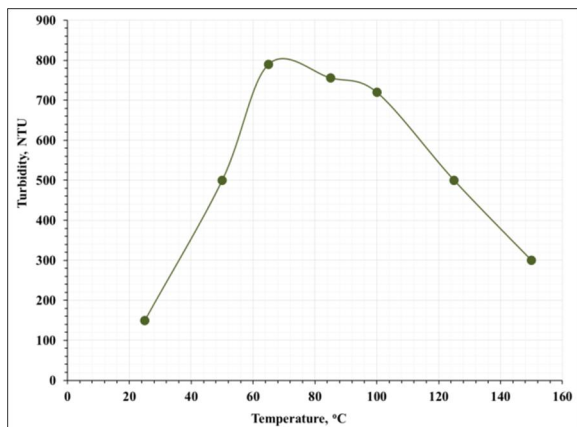


Figure (1): Effect of temperature on titanium phosphate precipitation (reaction time of 30 min, phosphoric acid concentration of 43.3% H_3PO_4 , with phosphoric acid/ titanium oxide mass ratio, ml/ g, 5.0, phosphoric acid rate of addition of 10 min, stirring speed of 400 rpm and nucleation time of 30 min).

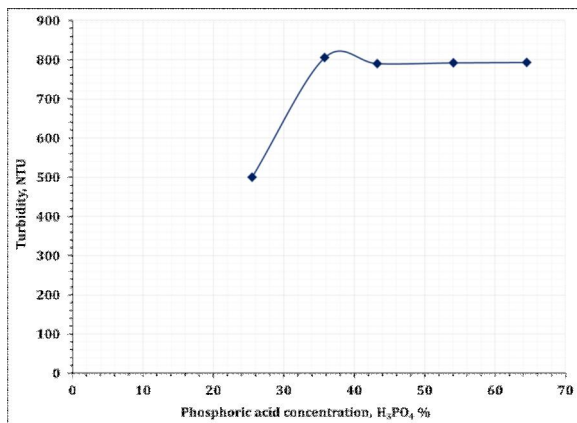


Figure (2): Effect of phosphoric acid concentration, H_3PO_4 %, on titanium phosphate precipitation (reaction time of 30 min, temperature 65 °C, phosphoric acid rate of addition of 10 min, stirring speed of 400 rpm and nucleation time of 30 min).

c. Effect of Precipitation Time:

To study the effect of reaction time on titanium phosphate precipitation process from titanium oxide by using 35.8 % H_3PO_4 phosphoric acid several experiments were carried at different times ranges from 10 to 45 min at a reaction temperature of 65 °C, phosphoric acid/ titanium oxide mass ratio, ml/ g is 5.0; phosphoric acid rate of 10 min, stirring speed of 400 rpm and nucleation time is 30 min. The experimental results are graphically presented in Figure (3) as a relation between turbidity (NTU) and time. From the Figure it is clear that, as the reaction

time increases from 10 to 30 min, the solution turbidity increased from 130 to 800 NTU, Further increase of reaction time from 30 up to 45 min leads to slightly decrease in the solution turbidity from 800 to 780 NTU. These results clear that the stirring time within the range from 10 to 30 min is improving the titanium phosphate precipitation process. Therefore, 30 min represents the preferred time to maximize the titanium phosphate formation from titanium oxide by using Abu-Tartur phosphoric acid.

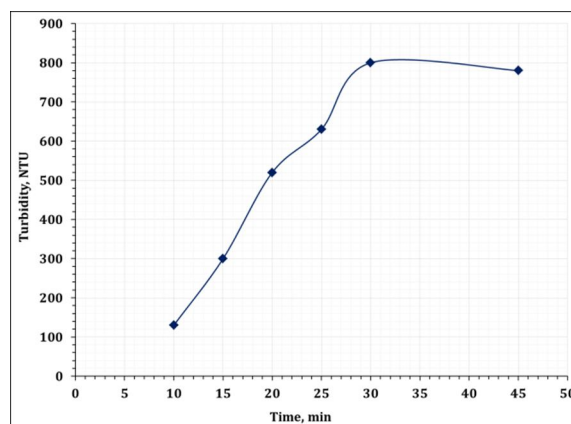


Figure (3): Effect of reaction time on titanium phosphate precipitation (phosphoric acid concentration, 35.8 % H_3PO_4 %, temperature 65 °C, with phosphoric acid/ titanium oxide mass ratio, ml/ g, 5.0, phosphoric acid rate of addition of 10 min, stirring speed of 400 rpm and nucleation time of 30 min).

d. Effect of Phosphoric Acid Rate of Addition:

Several precipitation experiments were performed by adding 35.8 % H_3PO_4 phosphoric acid to titanium oxide with different rates ranging from 1.0 to 15.0 min and reaction time of 30 min, with phosphoric acid/ titanium oxide mass ratio, ml/ g, 5.0, stirring speed of 400 rpm and nucleation time of 30 min at a temperature of 65 °C to study the effect of phosphoric acid rate of addition on the titanium phosphate precipitation process. The experimental results are given graphically in Figure (4) as a relation between turbidity (NTU) and phosphoric acid rate of addition. As can be seen from the Figure, by increase phosphoric acid rate of addition from 1 to 5 min the solution turbidity increased from 700 to 820 NTU. Further increases of phosphoric acid rate of addition from 5 up to 15.0 min almost have a slightly negative effect on the solution turbidity. This indicates that the increase in phosphoric acid rate of addition up to 5 min has positive effect on the precipitation process. Accordingly, all experiments were carried out with 5 min phosphoric acid rate of addition.

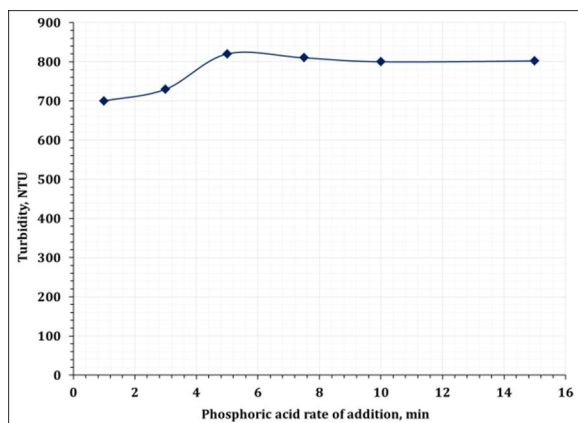


Figure (4): Effect of phosphoric acid rate of addition on titanium phosphate precipitation (phosphoric acid concentration, 35.8 % H_3PO_4 %, temperature 65 °C, with phosphoric acid/ titanium oxide mass ratio, ml/ g, 5.0, reaction time of 30 min, stirring speed of 400 rpm and nucleation time of 30 min).

e. Effect of Stirring Speed:

The effect of stirring speed on titanium phosphate precipitation process was studied with different stirring speeds from 200 to 600 rpm however; the other parameters were fixed at 35.8 % H_3PO_4 phosphoric acid concentration, reaction time of 30 min, phosphoric acid/ titanium oxide mass ratio, ml/ g, 5.0, phosphoric acid rate of addition of 5 min, temperature 65 °C and nucleation time of 30 min. The experimental results were plotted in Figure (5) as a relation between turbidity (NTU) and stirring speed clear that, as stirring speed increased from 200 to 400 rpm, the solution turbidity increased from 370 to 820 NTU. The increase in stirring speed up to 600 rpm has no effect on the solution turbidity. This means that stirring speed within the range from 200 to 400 rpm is enhancing the titanium phosphate precipitation process. Therefore, stirring speed of 400 rpm is the preferred stirring speed for the precipitation process using Abu-Tartur phosphoric acid.

Optimum titanium phosphate precipitation conditions:

From the aforementioned investigations, it is clear that, the optimum conditions for titanium phosphate precipitation using titanyl sulfate and phosphoric acid produced from Abu-Tartur phosphate concentrate are; 65 °C reaction temperature, 35.8 % H_3PO_4 phosphoric acid concentration, 30 min. reaction time, ml/ g, 5.0 phosphoric acid/ titanium oxide mass ratio, 5 min. phosphoric acid rate of addition and 400 rpm stirring speed.

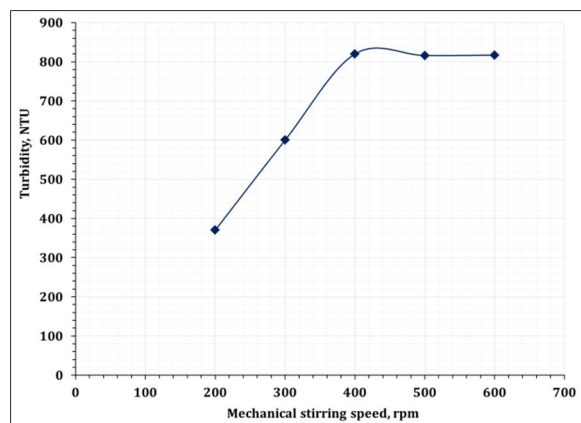


Figure (5): Effect of stirring speed on titanium phosphate precipitation (phosphoric acid concentration, 35.8 % H_3PO_4 %, temperature 65 °C, with phosphoric acid/ titanium oxide mass ratio, ml/ g, 5.0, reaction time of 30 min, phosphoric acid rate of addition of 5 min, and nucleation time of 30 min).

Characterization of the produced titanium phosphate precipitate:

Elemental analysis of the prepared titanium phosphate (TP) precipitate has been performed by ICP-OES. The obtained data shows that the ratio of Ti: P in titanium phosphate precipitate is 1: 1 with presence of small portion impurities of iron. SEM EDX analysis was used for further analysis (Wang *et al.*, 2007; Onoda and Yamaguchi, 2012; Onoda and Fujikado, 2014). As can be seen from the SEM micrographs in Figure (6), TP particles have no uniform with amorphous shape. EDX analysis graph determine the weight percent of element present in TP precipitate as shown in Figure (7).

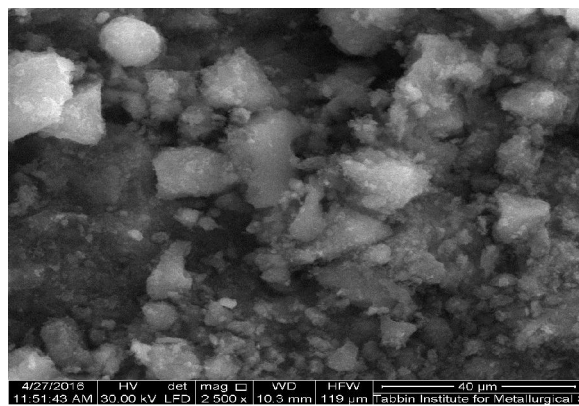


Figure (6): SEM of the produced titanium phosphate precipitate

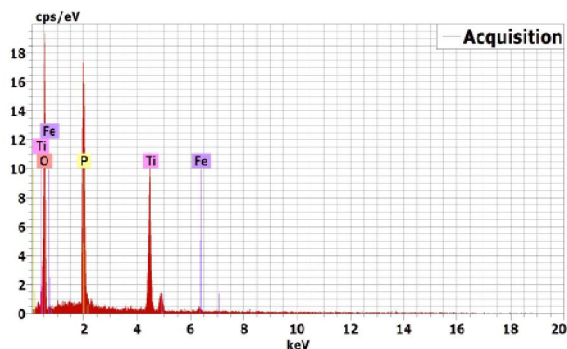


Figure (7): EDX of the produced titanium phosphate precipitate

The crystalline structure of titanium phosphate synthesized from titanyl sulfate and phosphoric acid have been estimated using XRD patterns Figure (8). As shown in XRD pattern an amorphous titanium phosphate is produced because of the absence of any sharp peaks in the X-ray diffractograms (Ludmány *et al.*, 2004; Patel and Chudasama, 2007; Jia *et al.*, 2008).

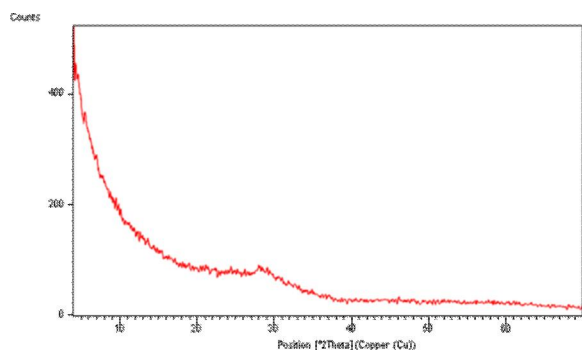


Figure (8): XRD of the produced titanium phosphate precipitate

The FTIR spectra of titanium phosphate precipitate, Figure (9), exhibits a broad band in the region $\sim 3400 \text{ cm}^{-1}$ (Region 1) which is attributed to symmetric and asymmetric $-\text{OH}$ stretching, while band at $\sim 1632 \text{ cm}^{-1}$ (Region 3), a sharp medium band is attributed to aqueous ($\text{H}-\text{O}-\text{H}$) bending. A band in the region $\sim 1026 \text{ cm}^{-1}$ (Region 5) is attributed to the presence of $\text{P}=\text{O}$ stretching. A medium intensity band at $\sim 1427 \text{ cm}^{-1}$ (Region 4) is attributed to the presence of δ (POH). These bands indicate the presence of structural hydroxyl groups/protonic sites in the material (Maheria and Chudasama, 2006b; Patel and Chudasama, 2007; Thakkar and Chudasama, 2009).

TGA, Figure (10) indicates two weight-loss regions. The first weight-loss region ($\approx 11.85\%$ up to 270°C) is attributed to loss of moisture/hydrated water. The second weight loss in the range $270\text{--}578$

$^\circ\text{C}$ is attributed to condensation of structural hydroxyl groups. The material gets converted to the pyrophosphate at temperatures greater than 700°C and the oxide at very high temperatures. The DTA shows two endothermic peaks and one exothermic as shown in Figure (11). The minor endothermic process in 117°C is related to removing of surface water. The second endothermic process in 217°C , respectively, followed by an exothermic peak at 721°C . The third endothermic process occurs in the range $873\text{--}981^\circ\text{C}$. The DTA and TG results indicate a structure similar to amorphous TiP (Suarez, 1983; Maslova *et al.*, 2012).

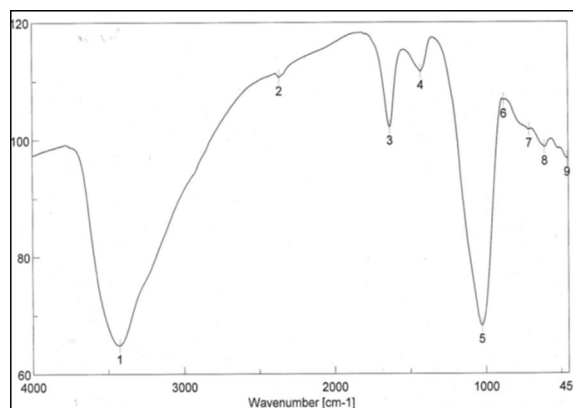


Figure (9): FTIR of the produced titanium phosphate precipitate.

From all previous characterizations and by using Alberti Torracca formula (1968), the TiP formula is $\text{TiO}(\text{OH})(\text{H}_2\text{PO}_4) \cdot 1.3\text{H}_2\text{O}$ (Maheria and Chudasama, 2007). Table (2) shows a comparison between the theoretical weight percent and the experimental weight percent in EDX which confirm the chemical formula of produced titanium phosphate precipitate.

Chemical resistivity:

Chemically resistivity/stability of prepared titanium phosphate in different solutions media; mineral acids, bases and organic solvent is useful and important while using the material for various applications in varied environments (Naushad, 2009; Thakkar and Chudasama, 2009). Titanium phosphate is found to be stable in acidic medium with maximum limits being (15 N HNO_3 , 10 N HCl and 15 N H_2SO_4) and also stable in organic solvent medium (benzene, acetone and ethanol) but not so stable in basic medium with maximum limits being (0.5 N KOH , 5 N NaOH).

Ion exchange capacity:

The ion-exchange capacity was measured by adapting the method used by Helen *et al.* (2007) 0.5 g of titanium phosphate sample was dispersed in 100 cm^3 of 0.1 M NaCl , kept under magnetic stirring for 6 hours. The supernatant liquid was then titrated with

0.15 M NaOH and the ion exchange capacity (IEC) of Na⁺ was calculated from the volume of consumed titrant required to reach pH 7 from the equation:

$$IEC = \frac{V \cdot C}{W}$$

Where IEC is the ion exchange capacity (meq/g), V the added NaOH volume when the solution pH was 7 (cm³), C the molar concentration of NaOH (mol/L), and W the test sample mass (g). IEC can be determined from pH titration curve in Figure (12) (Clearfield, 1988).

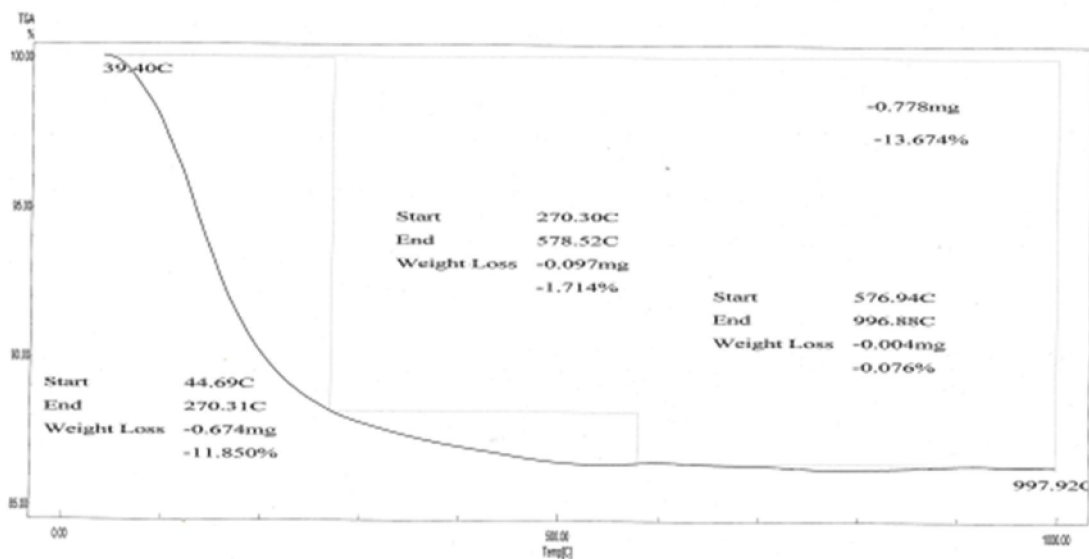


Figure (10): TGA the produced titanium phosphate precipitate

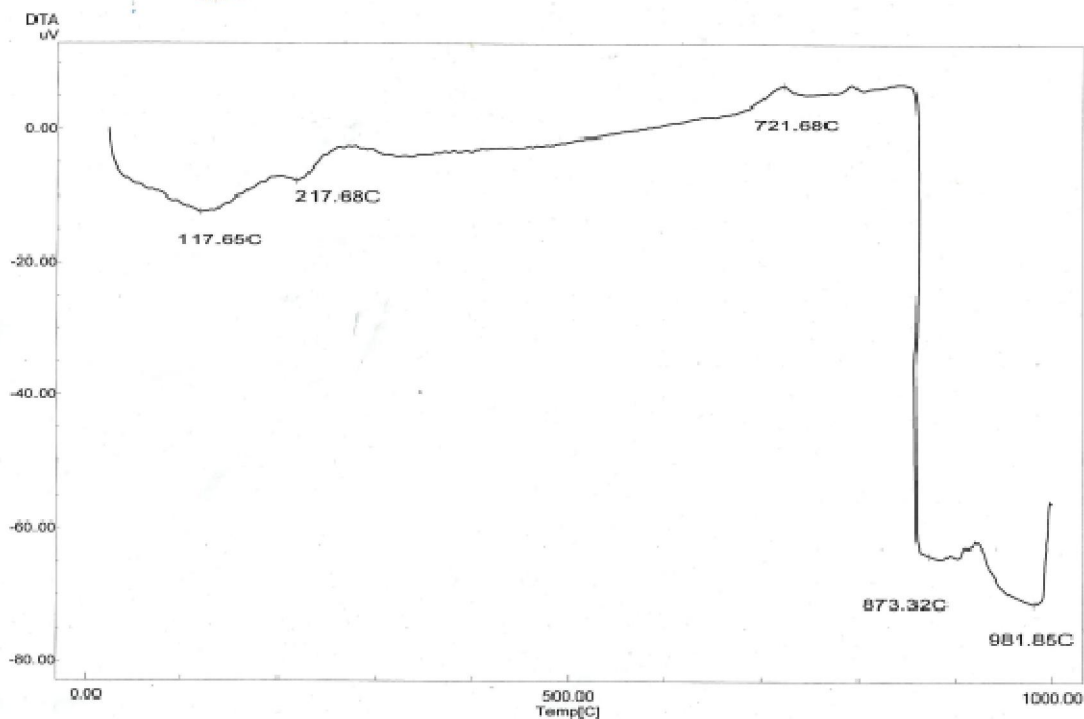


Figure (11): DTA the produced titanium phosphate precipitate

Table (2): Comparison between theoretical weight percent and EDX weight percent of titanium phosphate

Element	Theoretical Wt.%	Experimental Wt.%	Wt.% difference
Ti	23.79	24.73	0.94
P	15.39	15.66	0.27
O	58.03	58.69	0.66
Fe	0	0.92	0.92

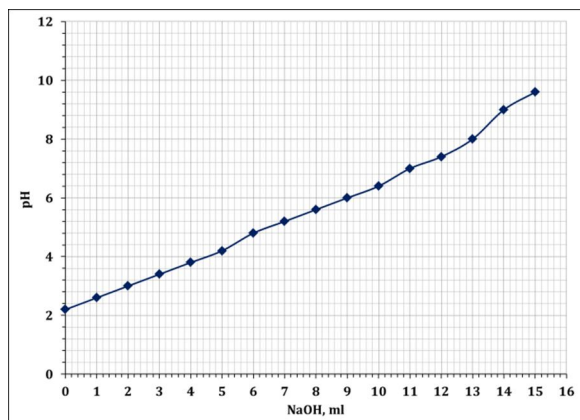


Figure (12): pH titration curve of 0.5 g TiP in 0.1 N NaCl (6 hr) with 0.15 N NaOH

Conclusion

Titanium phosphate particles were synthesized from titanyl sulfate and phosphoric acid produced from Abu-Tartur phosphate concentrate. The XRD analysis clarify that the obtained precipitate had amorphous phase. The chemical formula of the formed titanium phosphate is $\text{TiO}(\text{OH})(\text{H}_2\text{PO}_4) \cdot 1.3\text{H}_2\text{O}$. The prepared titanium phosphate exhibits good ion exchange capacity (3.3meq/g), thermal stability and chemical resistivity, which means that the produced titanium phosphate exhibits the characteristics of promising ion exchanger in the area of separation science.

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