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Zeolite Y synthesis without organic template: The effect of synthesis parameters

Raja BELAABED[1*], Hakima ELKNIDRI[1], Redouane ELKHALFAOUY[1], Abdellah ADDAOU[1], Ali LAAJAB[1], Ahmed LAHSINI[1]

1 : Laboratoire de Catalyse, Matériaux et Environnement, Ecole Supérieure de Technologie, Université Sidi Mohammed Ben Abdellah, Fès

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Keywords
- ✓ zeolite Y,
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- ✓ effect,
- ✓ time,
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Abstract

In this study, the zeolite Y was synthetized without organic template at low temperature of crystallization (100°C).The effect of crystallization time, aging time and aging temperature on zeoliteY synthesis were investigated. The end products were characterized by X-ray diffraction (XRD) and Environmental Scanning Electron Microscopy (ESEM). The results revealed that all the three parameters studied in this work have a great effect on crystallinity and final product of the synthesized zeolites.

1. Introduction

Zeolites are crystalline aluminosilicate or silicate materials, which have a highly regular and open microporous structure formed by a three-dimensional network of SiO_4 and AlO_4tetrahedra.

Because of their high surface area, fast diffusion characteristics, adjustable porosity, and high mechanical strength over amorphous porous silicas they have potential applications in fields such as catalysis, adsorption, bacterial adhesion and ion exchange [1–10].

Out of the various high silica zeolites, zeolite Y from the faujasite family is one of the microporous crystalline aluminosilicate zeolites which have 0.74 nm diameter pores and a threedimensional pore structure [11].

Faujasite zeolites are separated into two categories; zeolite X and zeolite Y which are differencied by their silicon to aluminum Si/Al ratio. Zeolite X has Si/Al ratio between 1 and 1.5, and zeolite Y has a Si/Al ratio above 1.5 [12]. The Si/Al ratio of Y zeolite is strongly related to its properties namely the thermal and hydrothermal stabilities, which plays a very important in application of this zeolite type in fluidized catalytic cracking (FCC) catalysts.

It has been reported that synthesis of zeolite is a complex process influenced by a large number of parameters including crystallization conditions and compositions parameters [13]. Therefore, much works have been focused on the investigations of various factors which controlled zeolite synthesis [14–18]. S. Alfaro et al showed that the structural and morphological properties of LTA zeolites depend strongly on the synthesis aging time [16]. M. Jafari et al revealed that crystallization parameters affect zeolite synthesis [15]. Xu Zhang and coauthors' have reported that a change of the batch molar ratio SiO2/Al2O3 changes the zeolite structure and composition, they showed also that the final product depend on crystallization time and temperature [14].

The objectives of present study were the preparation of Y zeolites by hydrothermal synthesis without organic template at low temperature of crystallization (100°C) and the investigation of the effects of synthesis parameters on the final synthesis product. The effects of synthesis parameters, namely aging time (ta), aging temperature (Ta) and crystallization time (tc) were investigated. The synthesized zeolites were characterized using X Ray Diffraction XRD and Environmental Scanning Electron Microscopy ESEM.

2.Experimental details

2.1. Synthesis

The precursor gels were prepared in two steps. First, an aqueous sodium hydroxide solution (Na OH 99%, Sigma-Aldrich) was mixed with an sodium aluminate (Na2O.Al2O3 anhydrous, Sigma-Aldrich) solution using magnetic stirring until dissolving.

Then, sodium silicate (Na2O.SiO2.5H2O, Sigma-Aldrich) was added into above suspension under stirring. The gels were aged for 8h / 24 h, at room temperature/ 80°C. The overall gel is hydrothermally treated for 6, 24, 48, 72 and 168 at 100°C without stirring. Then, the solid product is filtered off, washed with deionized water until the neutral pH and dried in air at 80°C overnight. Table 1 summarize the synthesis parameters for all samples.

Table 1: synthesis parameters

Sample	**Aging time t_a(h)**	**Aging temperature T_m(°C)**	**Crystallization time t_c(h)**
A	24	Room temperature	6
B	24	Room temperature	24
C	24	Room temperature	48
D	24	Room temperature	72
E	24	Room temperature	168
F	24	80°C	6
G	8	Room temperature	24

2.2. Characterization

Crystallization and purity of the synthesized zeolite phases was evaluated by X-ray diffraction XRD using an X-ray Diffractometer X'PERT PRO (PANalytical) with CuKα radiation, Ni filter. The reflection was scanned at 2 theta range between 4° and 50° in steps of 0.016.

Morphology is displayedusing Environmental Scanning Electron Microscopy ESEM using a Quanta-200 microscope (FEI Company).

3. Results and discussion

3.1. Effect of aging time

The effect of aging time on zeolite Y synthesis was investigated using the gel aged for 8 and 24 hour at room temperature and followed by crystallization for 24h (figure 1).

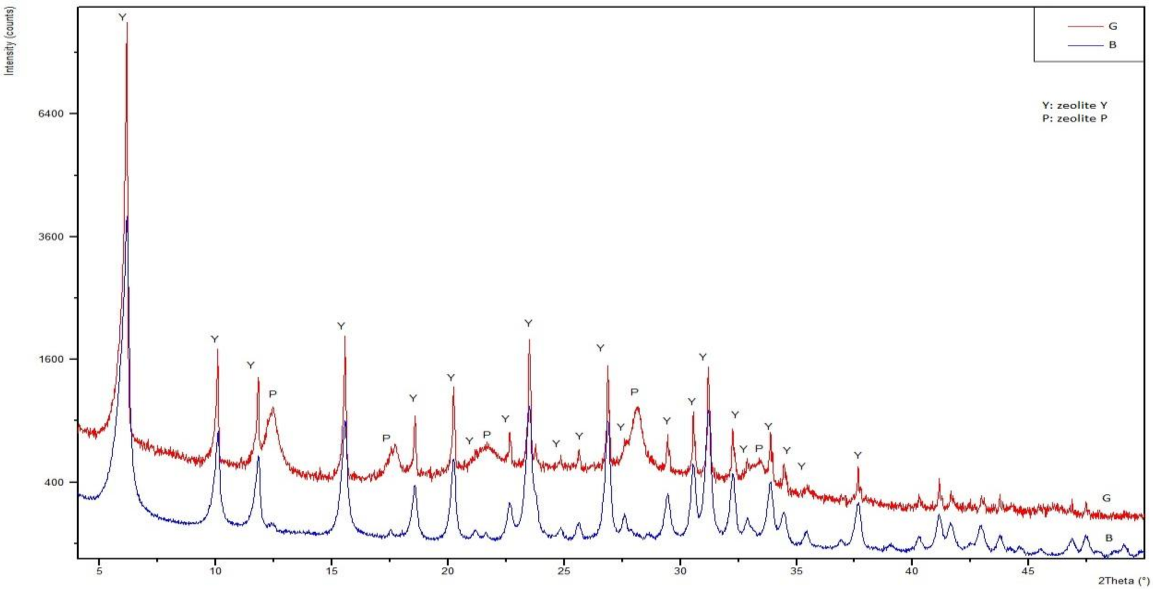

Figure 1: XRD spectrum of samples aged for 8(G) and 24h (B) at room temperature

The XRD spectrum of sample aged for 8h show zeolite Y and P with a high background noise, which may be explained by existence of amorphous phase. This result is confirmed with ESEM image, it display two different shapes and some aggregate particle (figure 2(G)). It has been reported that zeolite P appear as a second phase in zeolite Y synthesis [19]. 24h aging, gives rise to the formation of pure zeolite Y showed in figure 2(B).and displayed by homogenous shape in ESEM image.Kim and Ahn [20] pointed that aging time affect the crystallization of zeolite; they reported that increasing aging time caused a rapid increase in crystallization rate. It was reported that the crystallization depends on time-temperature treatment applied to the synthesis mixture before heating step [21].

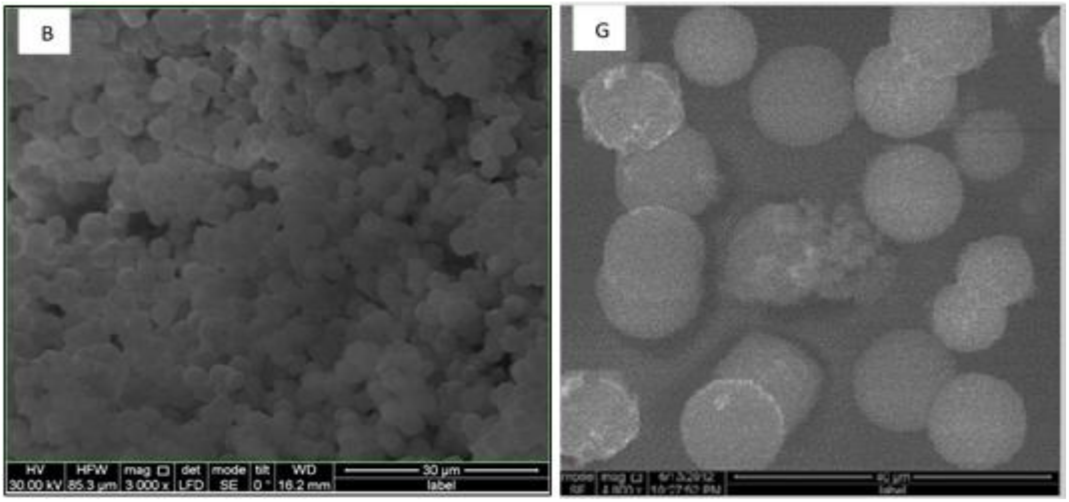

Figure 2: ESEM images of samples aged for 8h (G) and 24h (B) at room temperature

3.2. Effect of aging temperature

Figure 3 shows the XRD patterns of samples aged for 24h at room temperature and 80° and with crystallization for 6h. XRD patterns clearly revealed that the crystallinity depends on aging temperature. Sample aged at room temperature leads an amorphous phase whereas, when the aging temperature is increased, zeolite P is formed with a high background noise indicating that the sample still contained an amount of amorphous material. Figure4 (A) reveals that sample aged at room temperature consists of aggregated amorphous aluminosilicate gel However, the ESEM image of sample aged at 80° (figure 4(F)) displays a few particles embedded in amorphous aggregates. This result shows that increasing aging temperature accelerate the crystallization process. Cizimek et al. studied the effect of aging time on the synthesis of silicalite-1 at 170°C. They found that gels were aged from 0 to 192 h at ambient temperature, whereas, the ageing period was for 12 and 24 h when the ageing temperatures were 20, 35, 50, 65 and 80°C [22].

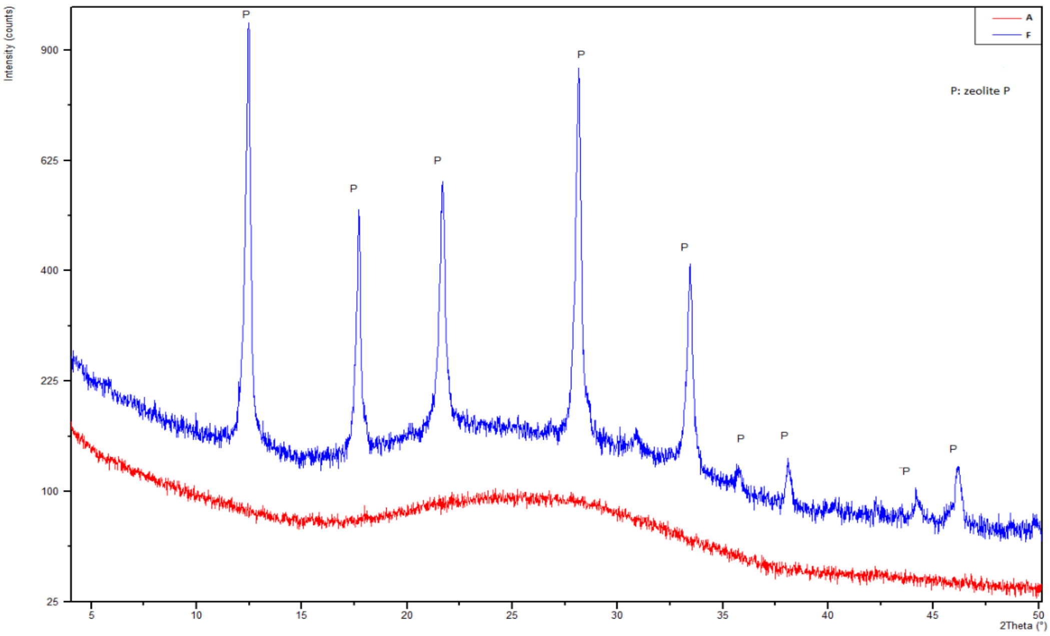

Figure 3:XRD spectrum of samples aged for 24h at room temperature (A) and 80°(F) with crystallization for 6h.

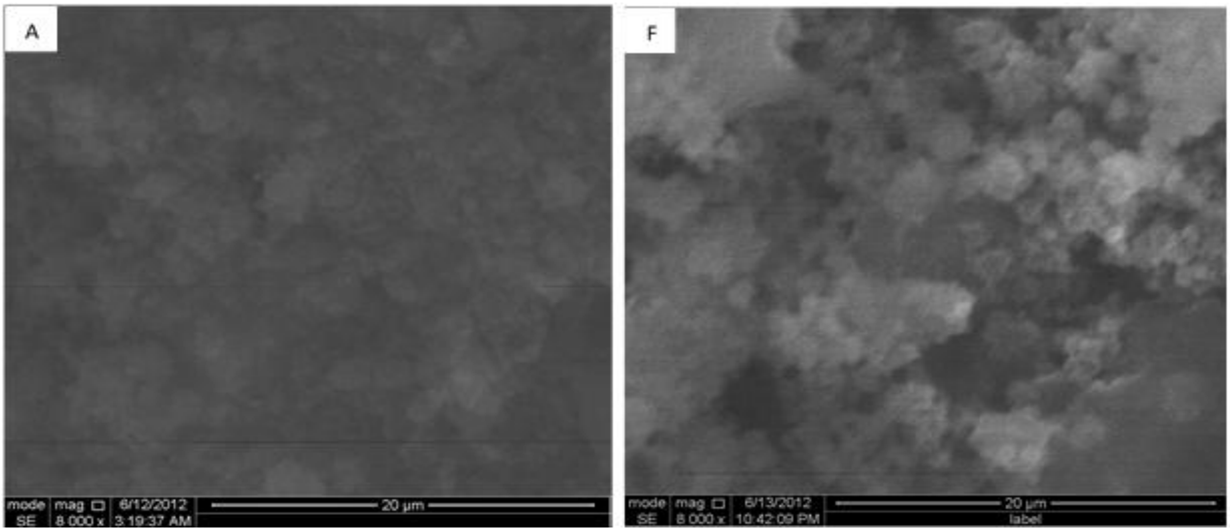

Figure 4: ESEM images of samples aged for 24h at room temperature (A) and 80° (F) with crystallization for 6h.

3.3. Effect of crystallization time

The sample aged for 24h at room temperature was used to study the effect of crystallization time; all samples were crystallized at 100°C. Figure 5 shows the XRD patterns of samples crystallized for 6(A) 24(B) 48(C) 72(D) 168(E). It can be seen that the XRD did not detect any crystalline structure after 6h hydrothermal treatment. Pure Y zeolite is obtained after 24h crystallization, further after prolonging crystallization time to 48h and 72h , another phase; zeolite P appear. After 168h of crystallization zeolite Y disappears and we have only zeolite P.

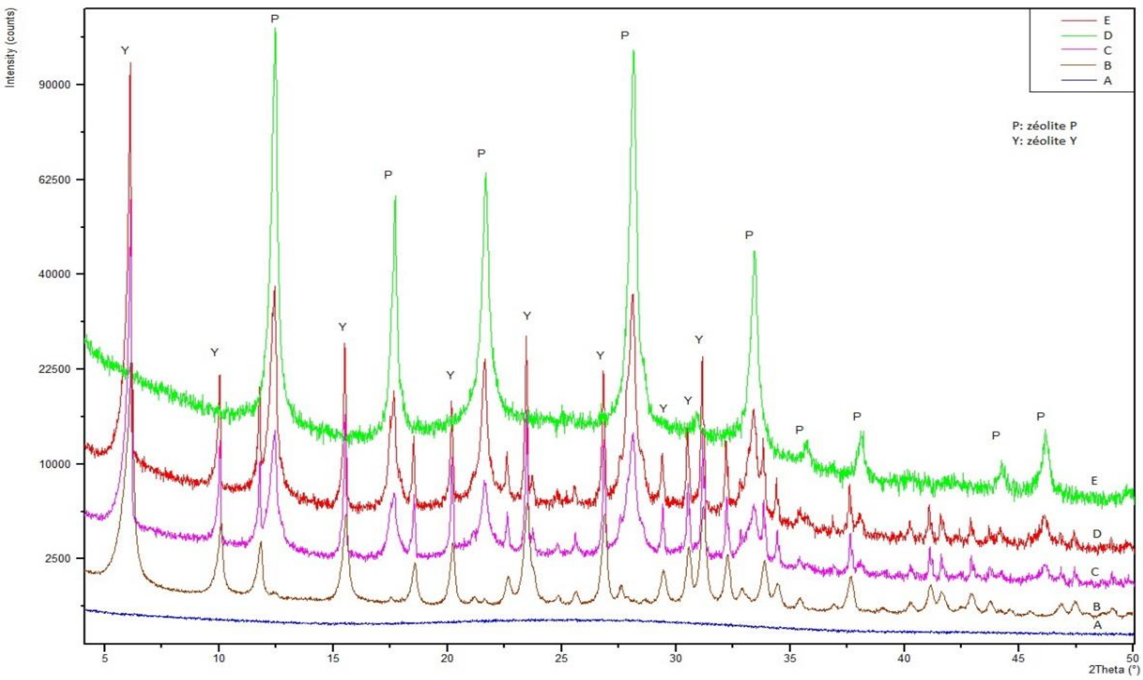

Figure 5 : XRD spectrum of samples aged for 24h at room temperature with crystallization at 100°C for 6h (A) 24h (B) 48h (C) 72h (D) and 168h (E)

The crystallization products obtained at 24 h matched the characteristic peaks of zeolite Y at d values of 14.29- 8.75- 7.48- 5.69- 5.06- 4.78- 4.38- 3.93- 3.78- 3.48- 3.32- 3.23- 3.03- 2.93- 2.87- 2.77- 2.72- 2.65- 2.60- 2.53- 2.44- 2.39- 2.24- 2.20- 2.17- 2.11- 2.07- 1.94- 1.92 that were reported by Donald W Breck [23] suggesting successful synthesis of zeolite Y with good crystallinity.

Nada Salman proved that the crystallization of zeolite P starts in the late step of synthesis of Y zeolite and its level increase when the crystallization time increase [19].

ESEM images of the samples treated for 6h (figure 6(A)) consist of amorphous aggregates. Well-developed zeolite crystals are formed after 24h of crystallization (figure 6(B)). However, when the crystallization time is increased to 48 and 72 h (figure 6 (C) and (D)), two different shape witch are defined by XRD as Y and P zeolite. After 168h we observe that the structure of crystals is damaged (figure 6(E)). The ESEM image show that crystallization time was found to have a substantial effect on the crystal morphology. These results are in good agreement with the XRD results.

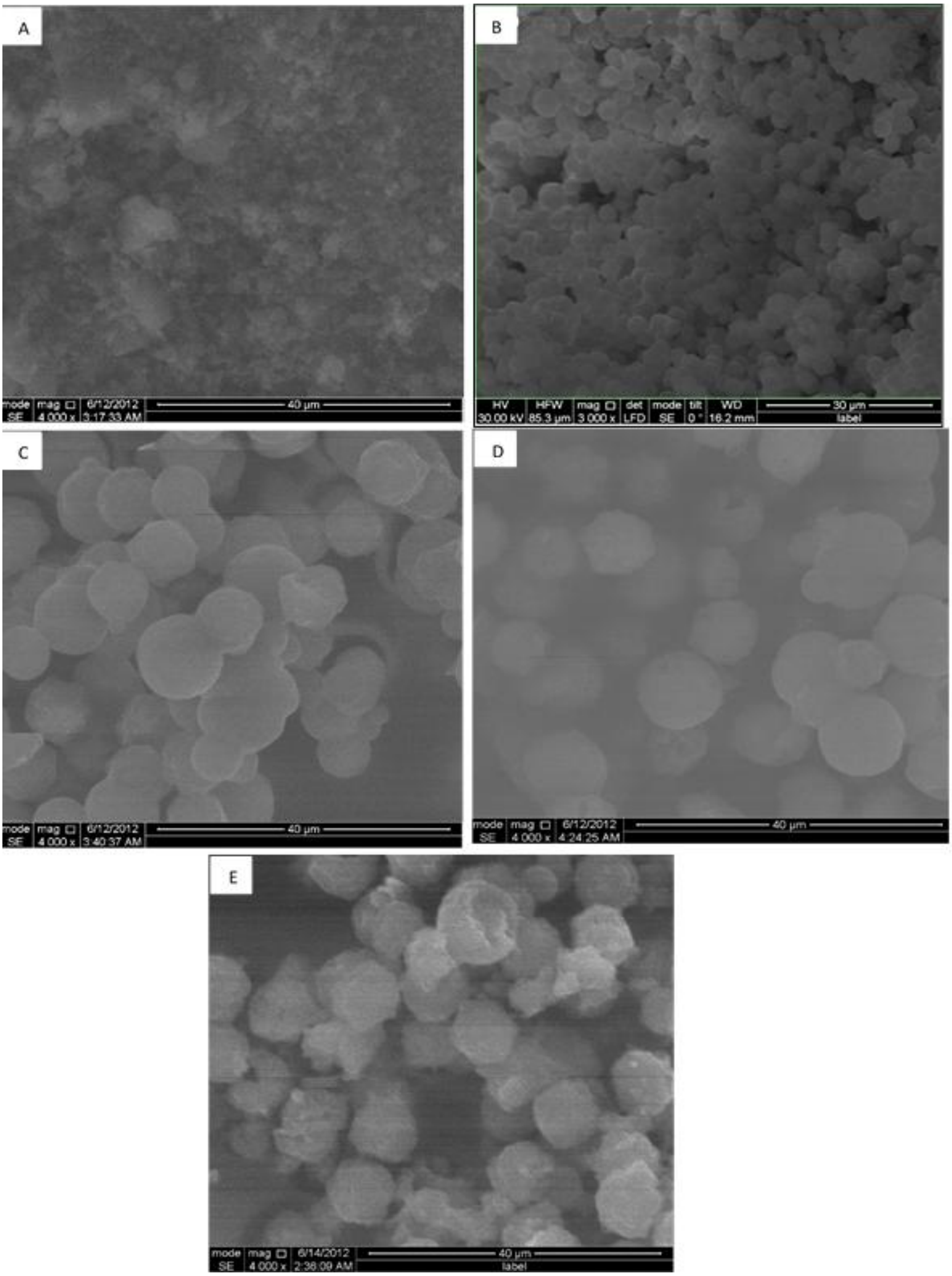

Figure 6 :ESEM Images of samples aged for 24h at room temperature with crystallization at 100°C for 6h (A) 24h (B) 48h (C) 72h (D) and 168h (E)

Conclusion

Pure zeolite Y crystallite was successfully synthetized without organic template, at 100°C for 24h, from the amorphous gel of aluminisilicate aged at room temperature for 24h. The aging time and temperature was also studied in this study. The sample aged for 8h gives rise to the formation of zeolite P and zeolite Y when the sample aged for 24h gives zeolite Y. The increasing of aging temperature seems accelerate the crystallization process. Using the same composition and the same aging condition, the variation of crystallization time give different zeolite (or amorphous) phases. The impact of synthesis parameters is seen for both crystallite phase and morphology of samples.

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