



Facile synthesis of mesoporous ZSM-5 aided by sonication and its application for VOCs capture

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ABSTRACT

Application of ultrasound power to the mother liquor is popular pretreatment for zeolite synthesis which offers a simple way of accelerating crystallization process and finetuning the properties of nanocrystalline zeolites. In this work, sonication-aided synthesis of mesoporous ZSM-5 at low temperature and ambient pressure was systematically studied, in an attempt to reach efficient and benign synthesis of zeolites with hierarchical pore structure, which has wide applications as catalysts and sorbents. The effects of sonication duration, power density, sonication temperature and seeding on the crystallization of ZSM-5 were investigated. The obtained samples were characterized by XRD, SEM, BET and VOCs capture. High quality mesoporous ZSM-5 can be obtained by a facile 5 d synthesis at 363 K, much faster than conventional hydrothermal synthesis. The reduced synthesis time was mainly attributed to the enhanced crystallization kinetics caused by the fragmentation of seeds and nuclei, while sonication radiation had little impact on the nucleation process. Compared with control sample, mesoporous ZSM-5 prepared by sonochemical method had higher surface area and mesoporosity which demonstrated improved adsorption performance for the capture of isopropanol.

1. Introduction

Zeolites are a group of microporous crystalline materials whose structures are composed of regular networks of tetrahedral SiO_4^{4-} and AlO_4^{5-} . Zeolites have gained numerous applications in many industries as catalysts, ion-exchangers and sorbents because of their regular microporous pore structure, large specific surface area and porosity, tuneable acidity and adsorption characteristics, excellent thermal and hydrothermal stability [1,2]. The formation of zeolite usually involves two steps, nucleation and crystallization, which could be time-consuming. Therefore, long hydrothermal treatment at high temperature is generally required for the synthesis of many zeolites, which leads to issues like lengthy synthesis time, high energy consumption, high equipment cost and safety issue associated with the use of high-pressure autoclave etc. It is extremely desirable to shorten synthesis time and lower synthesis temperature, which not only reduces synthesis cost (energy consumption, autoclave productivity etc), but also lowers environmental footprints and safety concerns. To reach this goal, it is of great importance to enhance nucleation and crystallization kinetics

during zeolite synthesis, which could be realized by the following approaches [2], including recipe (high concentration) [3–6], high synthesis temperature [7,8], reactive Si source [9–11] and Al source [12], fast heating (including microwave heating [13–15] and oil-bath heating [16–18]), seeding [3,5,18–21] etc. However, these methods usually associate with high equipment cost, the use of expensive and toxic chemicals, safety concerns, etc., which limit their large-scale applications.

In recent years, sonochemical method has attracted researchers' interest as the application of ultrasound energy to the reaction media can accelerate the reaction rate of many kinds reactions in a simple and benign way [22–24]. The mechanism causing this sonochemical effects can be called acoustic cavitation, where acoustic cavitations are generated in the reaction media upon the ultrasonic radiation, which lead to the formation, growth and implosive collapse of numerous tiny bubbles in the solvent. The collapse of these bubbles produces extremely high temperature (>5000 K), pressure (>20 MPa) and very high cooling rates ($>10^7$ K·s⁻¹), which can enhance the kinetics of many kinds of reactions [22,23], including zeolite crystallization [24]. Until now, ultrasound

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assisted synthesis has been used for the preparation of many zeolites, such as SAPO-34 [25–27], NaY [28], NaA [29,30], MCM-22 [31], DDR [32,33], CHA [34,35], NaP [36], EMT [37], T [38] and ZSM-5 [39–46]. In the sonochemical synthesis of zeolites, ultrasound radiation was usually applied to mother liquor during aging process, and the mother liquor was then treated by a regular high temperature hydrothermal synthesis. Sonication of mother liquor usually led to shorter synthesis time and smaller crystals, which was generally ascribed to the positive contribution of sonication energy to the nucleation and crystallization processes [24].

For example, Taghizadeh et al. studied sonochemical synthesis of nano ZSM-5. They found the crystallization time was reduced from 72 h to 60 h and the crystal size was reduced from 258 nm to 30 nm, while the catalytic performance for the dehydration of methanol to dimethyl ether was the same as sample prepared by conventional synthesis [41]. In the sonication-aided synthesis of ZSM-5, Fatemeh et al. claimed crystallization time was significantly reduced from 48 h to 4 h, while the crystal size was reduced from 890 nm to 310 nm. Compared with the parent catalyst, the catalyst prepared by sonochemical synthesis exhibited higher selectivity for propylene (C_3 , 36.8 %) and lower selectivity for total heavy hydrocarbons (C_{5+} , 16.1 %) in methanol to propylene reaction [42]. Kong et al. found similar shortening of synthesis time (from 48 h to 32 h) and crystal size reduction (from 300 nm to 50 nm) in the ultrasound-assisted synthesis of nanosized ZSM-5. In the alkylation of o-xylene with styrene, the activity of the ultrasonically treated ZSM-5 decreased only 5 % after 12 h operation, while the activity of control sample decreased 63 % [45].

However, after literature review, we find most of sonochemical zeolite syntheses followed the same pattern, room temperature sonication + conventional high temperature hydrothermal synthesis. As far as we know, the application of ultrasound pretreatment for the facile synthesis of mesoporous ZSM-5 at low temperature and ambient pressure has not been reported. Zeolite synthesis at low temperature and ambient pressure greatly reduces equipment cost and energy consumption, which is extremely desirable for commercial applications. The presence of mesopores in microporous zeolite crystals alleviates diffusional constraint of zeolite's intrinsic microporous channels, which has broad applications in catalysis and adsorption [1,2]. In this study, we systematically studied the sonochemical synthesis of mesoporous ZSM-5 at low temperature and ambient pressure. The effects of sonication duration, power density, sonication temperature and seeding on the crystallization of ZSM-5 were investigated. The obtained mesoporous ZSM-5 samples were characterized by X-ray diffraction (XRD) and scanning electron microscopy (SEM), N_2 adsorption-desorption technique (BET), Fourier transform infrared spectrometer (FT-IR), and VOCs capture performance.

2. Experimental

2.1. Chemicals

Sodium hydroxide (NaOH, 99.8 %, Shanghai Taitan Chemicals), sodium aluminate ($NaAlO_2$, Shanghai Bailingwei Chemicals), silica sol (SiO_2 , 25 wt%, Qingdao Haiyang Chemicals), tetrapropylammonium bromide (TPABr, 99 %, Anhui Nuochun Chemicals) and isopropanol (99.9 %, Shanghai Taitan Chemicals) were used as received.

2.2. Conventional hydrothermal synthesis of mesoporous ZSM-5

Mesoporous ZSM-5 was prepared in a mother liquor with molar ratio of $1SiO_2:0.004Al_2O_3:0.03TPABr:0.13NaOH:12H_2O$ [47]. Silica sol was slowly added into the mixture of sodium hydroxide, TPABr and DI water with 2 h stirring. Then sodium aluminate was added under stirring. After 2 h aging, seed crystals (1 wt% ZSM-5 crystals, relative to the amount of SiO_2 added in the mother liquor, prepared by the same recipe) was added into the mother liquor with 30 min stirring, which was transferred

into a polypropylene bottle for x days hydrothermal synthesis at 363 K. After crystallization, the product was collected by suction filtration, washed until pH = 7 and dried at 383 K. The obtained zeolites were calcined at 873 K for 5 h in air for template removal.

2.3. Sonochemical synthesis of mesoporous ZSM-5

The same mother liquor was used for the sonochemical synthesis of mesoporous ZSM-5, which was also prepared the same way as previous section. The prepared mother liquor was placed in a polypropylene bottle and an ultrasonic probe (TL-650Y Jiangsu Tianling Instrument) was directly immersed in the mother liquor. The volume of mother liquor was fixed at 20 mL. The power of sonication can be controlled at 33 W, 130 W, and 293 W respectively. The sonication treatment was operated in a non-continuous mode, 3 s sonication + 2 s interval. The temperature of mother liquor can be controlled between room temperature (~293 K) to 363 K using a water bath or an oil-bath. The mother liquor was subjected to ultrasonic radiation for certain periods of time, and then hydrothermally treated at 363 K for x h. After hydrothermal synthesis, the same filtration, washing, drying and calcination were used to collect the powder products.

2.4. VOCs removal

Fixed bed adsorption experiments with granular ZSM-5 were performed in custom-made adsorption system, as shown in Fig. 1. The granular ZSM-5 sorbent (~0.5 g, 40–60 mesh) was loaded into a U-shaped quartz tube (6 mm inner diameter) and activated at 473 K in a 30 mL/min N_2 flow for 4 h before VOC adsorption at ambient pressure and room temperature. The VOC containing stream was prepared by bubbling nitrogen stream into VOC liquid (isobutanol in this study, three bubblers in cascade mode to ensure the saturation of VOC in the nitrogen stream), then mixed with another pure nitrogen stream. The VOC concentration in the final nitrogen stream can be controlled in a broad range by manipulating the ratio of two nitrogen streams and the temperature of bubbling process (controlled in a thermostat bath). The final nitrogen stream with hundreds ppm VOC was sent to the adsorption column and the VOC concentration in the effluent stream was monitored by a gas chromatograph (Shanghai Fuli 2060 with a FID detector, on-line monitoring). After adsorption, the adsorbed VOC was desorbed by heating at 483 K for 4 h under 50 mL/min flowing nitrogen. The calculation of adsorption capacity (q, the weight of adsorbed VOC/the weight of sorbent, $g \cdot g^{-1}$) was defined by the equation below.

$$q = \frac{M \times F \times C_0 \times 10^{-9}}{22.4W} \left[t_s - \int_0^{t_s} \frac{C_t}{C_0} dt \right]$$

M is the molecular weight of adsorbate (g/mol) and W is the weight of sorbent (g). F is the volumetric flow rate of feed stream ($ml \cdot min^{-1}$) and C_i and C_0 are the VOCs concentrations (ppm) of the feed stream and effluent stream respectively. T_i and t_s are the adsorption time (min) and the adsorption equilibrium time (min), respectively.

2.5. Characterization

A Bruker X-ray powder diffractometer (D2 PHASER) was used for phase identification. The calculation of relative crystallinity was based on the sum of seven peaks of zeolite ZSM-5 ($2\theta = 7.9, 8.9, 22.9, 23.3, 23.6, 23.9$ and 24.3°). The sample with the highest sum of peak intensities is considered as 100 % crystalline. Crystal morphology and size were studied by a Hitachi scanning electron microscope (SU8010). The surface area analysis was performed by an automatic micropore physorption analyzer (Tristar 3020, USA). The Fourier-transform infrared spectra (FT-IR) of the ZSM-5 samples were recorded on a Nicolet Nexus 470 FT-IR spectrometer. Ultrasonic radiation is generated by a probe sonicator (TL-650Y, Jiangsu Tianling Instrument).

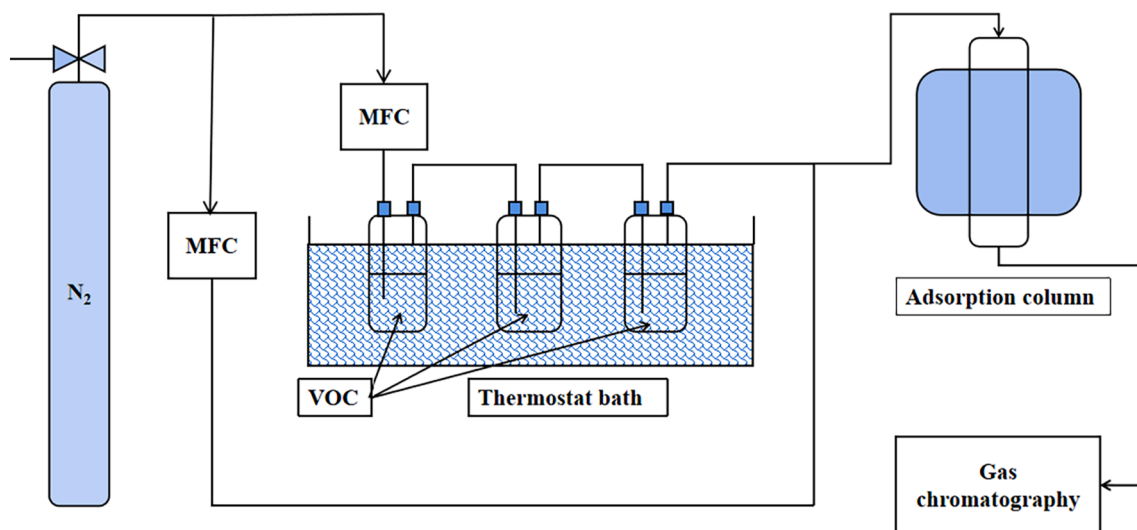


Fig. 1. Flow chart of VOCs adsorption-desorption system.

3. Results and discussion

3.1. Effect of sonication time

In this study, only the following recipe was used for the synthesis of

mesoporous ZSM-5 (1SiO_2 : $0.005\text{Al}_2\text{O}_3$: 0.03TPABr : 0.13NaOH : $12\text{H}_2\text{O}$). First, the effect of sonication time on the synthesis of zeolite ZSM-5 was studied, as shown in Fig. 2. The sonication power was fixed at 33 W and sonication temperature was fixed at room temperature (in a water bath). Fig. 2(a) shows the effect of hydrothermal synthesis time on

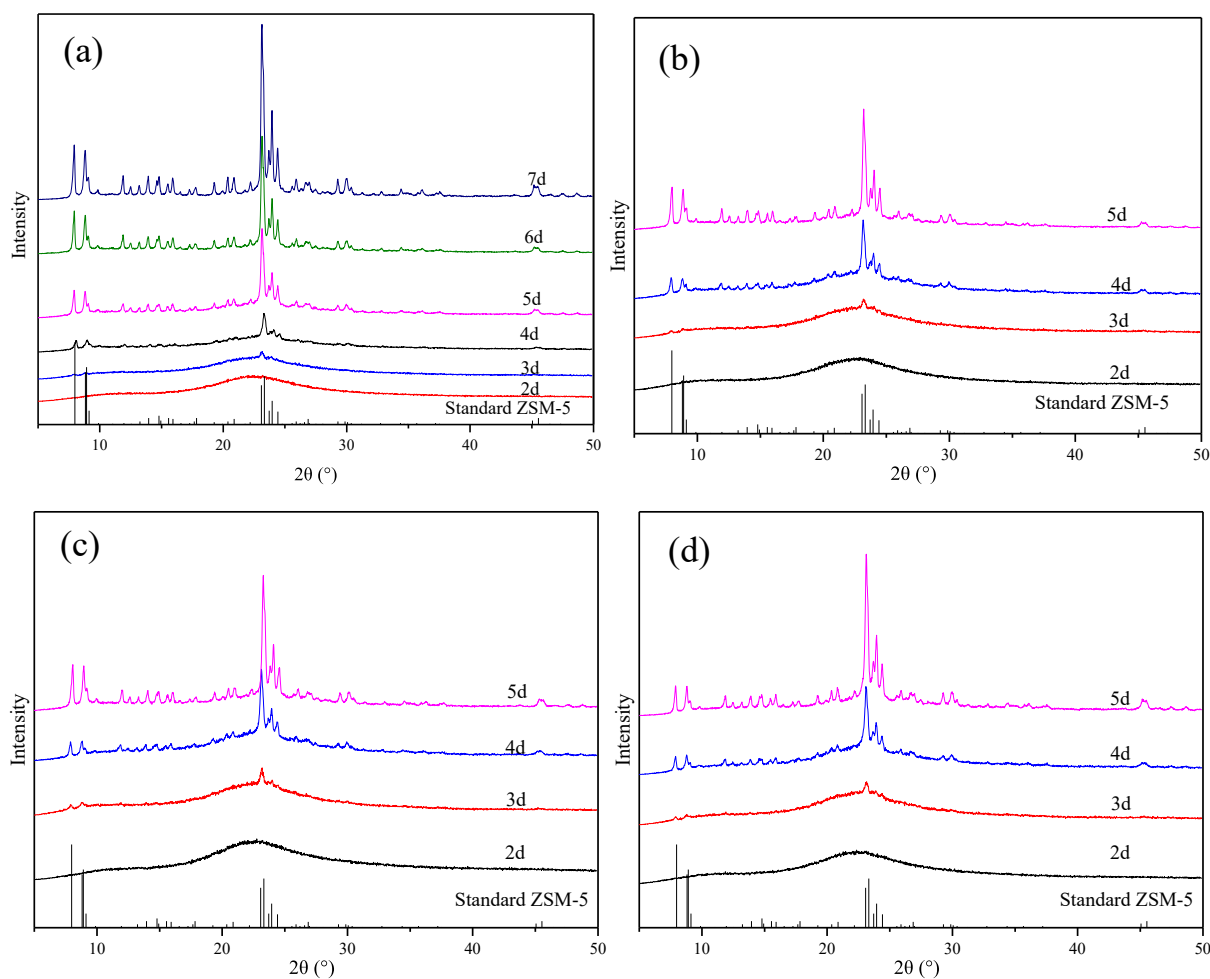


Fig. 2. The effect of sonication time on the crystallization of ZSM-5 samples, (a) 0 min, (b) 10 min, (c) 15 min and (d) 20 min (no seeding, HT synthesis at 363 K, sonication power: 33 W).

the synthesis of ZSM-5 without the aid of sonication (control experiment). No crystalline material was obtained after 2 days synthesis at 363 K, demonstrated by the amorphous XRD patterns. After 3 days synthesis, characteristic peaks of zeolite ZSM-5 started to appear at 2θ between 7 and 9° and 22 – 25° . However, these peaks were very weak, suggesting the early stage of crystallization. After that, the peak intensity increased very fast with synthesis time and decent ZSM-5 can be obtained in 7 days. Compared with conventional high temperature hydrothermal synthesis of zeolite ZSM-5 (usually 2 days at ~ 453 K), our synthesis at 363 K is much slower, which can be attributed to the slow crystallization kinetics at such low synthesis temperature, as crystallization rate decrease exponentially with temperature. However, zeolite synthesis at low temperature and ambient pressure is highly desirable due to the low energy consumption and equipment cost, but the sacrifice of synthesis time reduces synthesis efficiency. Therefore, sonication treatment of mother liquor was carried out to increase crystallization kinetics and shorten synthesis time.

Fig. 2b, c and d show the effect of sonication time (10, 15 and 20 min) on the synthesis of ZSM-5. As shown by the XRD patterns in Fig. 2, sonication treatment of mother liquor did have some impact on the overall crystallization kinetics. After 2–3 days synthesis, the products were still amorphous, like sample prepared without sonication treatment. However, after 4 days synthesis, the difference on crystallinity started to show up and increased with synthesis time. The trend is clearly shown by the crystallinity \sim synthesis time curves in Fig. 3. Judged by crystallinity data, we can say the impact of sonication treatment was negligible in the first 3 days, demonstrated by almost the same sample crystallinities. However, the difference on crystallinity became obvious after 3 days and the final product crystallinity had a positive relationship with sonication time. The first 3 days can be considered as the induction period of zeolite synthesis, when the nucleation process occurred. After nucleation, zeolite crystallization proceeded on the surface of these tiny nuclei. This suggests the sonication treatment of mother liquor in this study had some impact on the crystallization process while the nucleation process was not accelerated. Similarly, Mu et al. studied the sonochemical synthesis of zeolite SSZ-13 and found the shortening of synthesis time mainly came from crystallization process rather than nucleation process [35].

Fig. 4 shows the SEM image of mesoporous ZSM-5 samples prepared with different sonication durations. Fig. 4(a–c) show the SEM images of ZSM-5 samples prepared without sonication treatment between 5 and 7 days at 363 K. After 5 days synthesis, a mixture of spherical particles (particle size $\sim 1.8 \mu\text{m}$) and substantial amount of amorphous material was observed, which indicates 5 days synthesis might not be enough for

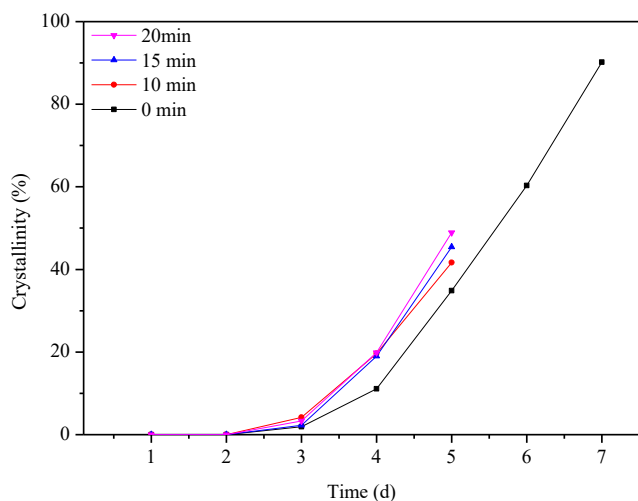


Fig. 3. The effect of sonication time on the crystallinity-synthesis time curves (no seeding, HT synthesis at 363 K, sonication power: 33 W).

the crystallization of ZSM-5. Further extension of synthesis time to 6 and 7 days led to fully developed spherical crystals with little amorphous materials. With the aid of sonication radiation, the percentage of amorphous materials decreased substantially with the increase of sonication time, suggesting positive impact of sonication treatment on the crystallization of zeolite ZSM-5. However, compared with the fully crystallized ZSM-5 sample prepared with 7 days synthesis, the presence of amorphous materials suggests incomplete crystallization. This result indicates that sonication treatment of mother liquor did enhance the crystallization of zeolite ZSM-5 even at such low synthesis temperature. However, the improvement on crystallization kinetics is not significant, which indicates the sonication intensity (duration or power density) is not enough.

3.2. Effect of seeding and sonication power

In this section, we studied the impact of seeding and sonication energy density on the crystallization of zeolite ZSM-5. The reason we use seeding is that sonication treatment in the previous section didn't shorten the induction period, which was also observed by other researchers [35]. Certainly, the addition of seeds into the mother liquor would eliminate the lengthy induction period and therefore shorten synthesis time significantly. In this section, sonication time and sonication temperature were fixed at 10 min and room temperature respectively, while the seeding was fixed at 1 wt% (relative to SiO_2 added in the mother liquor).

Fig. 5(a) shows the effect of hydrothermal synthesis time on the seeded synthesis of ZSM-5 without sonication treatment. Similar to the previous results, no crystallized product was obtained in the first two days, and weak ZSM-5 diffraction peaks appeared on the third day. After 5 d synthesis, the ZSM-5 peaks are marginally higher than the sample prepared without seeding (see Fig. 1). This result is a surprise to us as seeding didn't eliminate the lengthy induction period, which might be attributed to the low synthesis temperature used in this study. Then, sonication power was combined with seeding to further accelerate nucleation and crystallization processes.

Fig. 5(b), (c) and (d) show the effect of sonication power on the synthesis of zeolite ZSM-5. The sonication power was increased from 33 W to 130 W and 293 W (fixed 20 mL mother liquor). It is worthy to point out that the use of high sonication power can produce substantial amount of heat in the system, which would lead to a temperature increase up to 30 K in the mother liquor. This temperature jump may create an unfair comparison considering the low synthesis temperature used in this study. Therefore, a water-bath was used to control the temperature of mother liquor within 5 K of target temperature. As shown by the XRD patterns in Fig. 5, the trend is similar to previous section. No crystallization occurred in the first 2 days and weak MFI peaks started to appear in the 3rd day. After that, crystallization rate apparently picked up and well-developed XRD patterns can be obtained after 5 days synthesis. Judged by crystallization curves shown in Fig. 6, the trend is almost the same as Fig. 4. Both seeding and sonication power can increase the crystallization rate substantially, judged by the higher crystallinity of the final products after 5 days synthesis. However, seeding and sonication treatment had little impact on the nucleation process, which still has the same lengthy 2 d induction period as control experiment.

Fig. 7 shows the effect of sonication power on the SEM images of ZSM-5 samples (5 days synthesis at 363 K). Fig. 6(a) is the SEM image of seeded synthesis without sonication treatment, which is not fully crystallized as shown by the mixture of spherical particles (zeolite crystals $\sim 2 \mu\text{m}$) with some amorphous materials. Compared with non-seeded synthesis in Fig. 3, the percentage of amorphous material is lower, in line with its XRD pattern with higher peak intensity. With the aid of sonication radiation, zeolite crystallization was enhanced substantially and amorphous materials decreased with the increase of sonication power. At 130 W sonication power, fully developed spherical particles

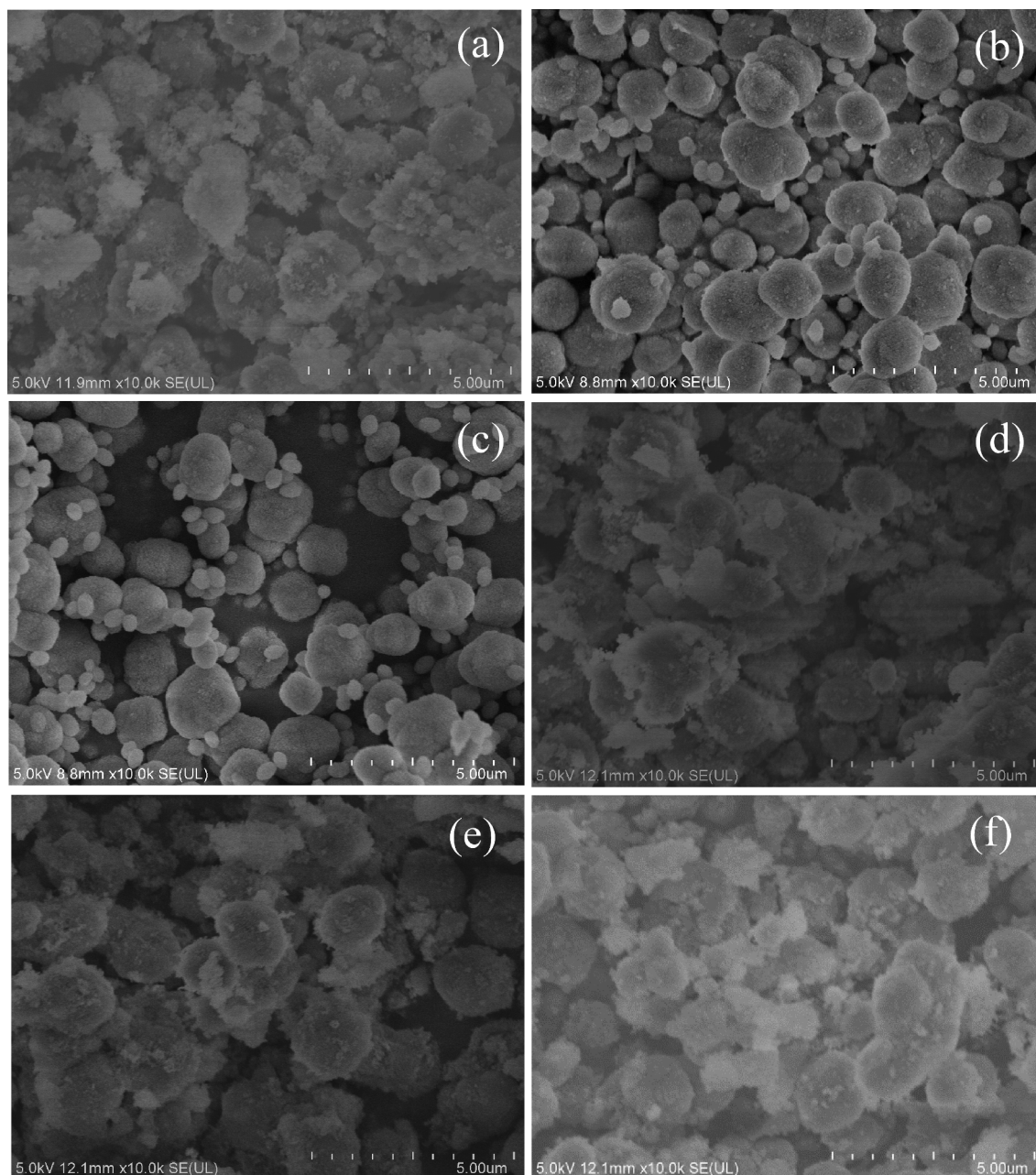


Fig. 4. The effect of sonication time on the synthesis of ZSM-5, (a) no sonication + 5 d HT, (b) no sonication + 6 d HT, (c) no sonication + 7 d HT, (d) 10 min sonication + 5 d HT, (e) 15 min sonication + 5 d HT and (f) 20 min sonication + 5 d HT (no seeding, HT synthesis at 363 K, sonication power: 33 W).

~1.8 μm were obtained in 5 d and there was no sign of un-converted amorphous materials. Further increase of sonication power to 290 W has no obvious impact on crystal morphology, but the crystallinity was improved by ~15 %. Higher sonication power was not tried considering the small volume of mother liquor ~20 mL used in this study. The obtained result suggests that the combination of sonication + seeding has pronounced impact on zeolite synthesis, mainly on crystallization rather than nucleation. Highly crystalline ZSM-5 sample can be obtained in 5 d synthesis at 363 K, which cannot be realized by sole sonication treatment or sole seeding treatment. It seems that both seeding and high energy sonication cannot reduce the induction period, which is different with our expectation and previous publications [45]. We hypothesize the low sonication temperature (~room temperature) used in this study cannot provide enough energy required for nucleation and crystallization. Therefore, we investigated high temperature sonication treatment at synthesis temperature, which might contribute positively to

nucleation and crystallization processes. We didn't intend to increase synthesis temperature to ~453 K (conventional synthesis temperature of ZSM-5) as our interest is low temperature synthesis at ambient pressure. Sonication of mother liquor at temperature higher than 373 K creates sealing issue, which increases system cost significantly.

3.3. Effect of sonication temperature

In previous sections, sonication treatment of mother liquor was performed at room temperature. In this section, we increased sonication temperature from room temperature to 333 K and 363 K (synthesis temperature) with a purpose of further reducing synthesis time. The sonication power was fixed at 293 W, and the sonication time was fixed at 10 min.

Fig. 8 shows the effect of sonication temperature on the XRD patterns of ZSM-5 samples, while Fig. 9 shows the crystallinity data calculated

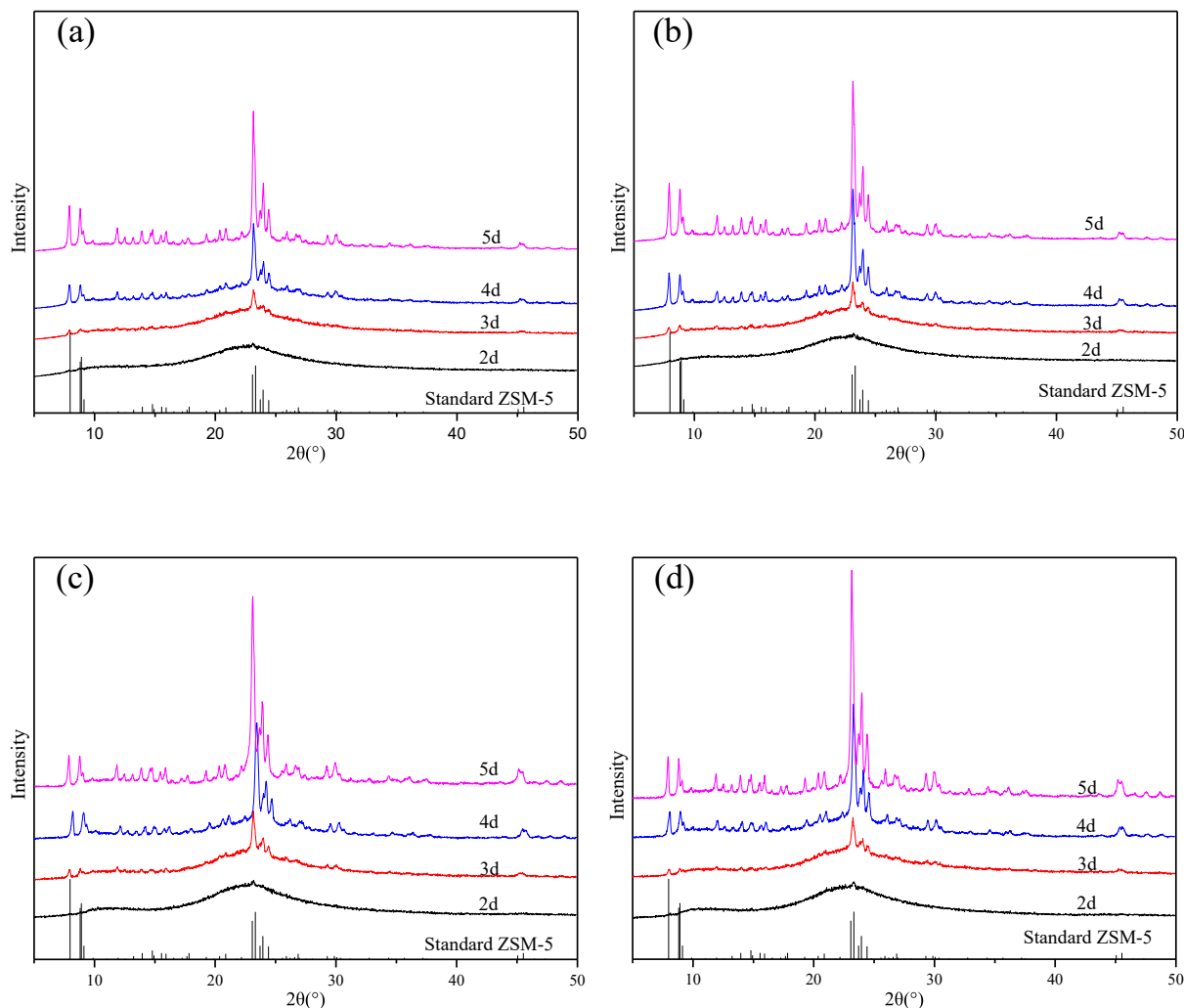


Fig. 5. The effect of seeding and sonication power on the XRD patterns of ZSM-5 samples, (a) seeding + no sonication, (b) 33 W, (c) 130 W and (d) 293 W (HT synthesis at 363 K, sonication time: 10 min).

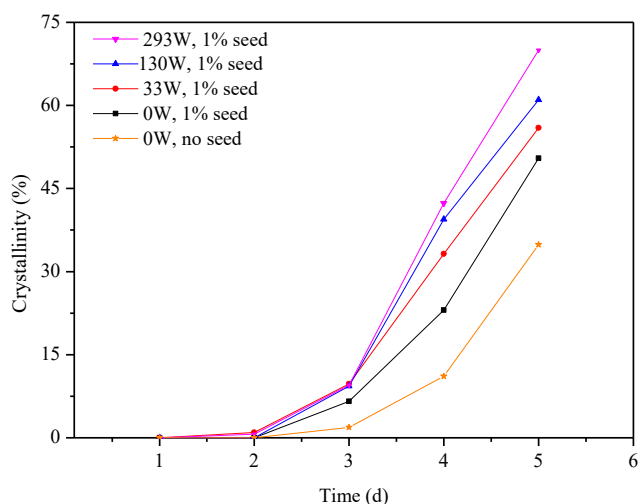


Fig. 6. The effect of sonication power density on the crystallinity ~ synthesis time curves (HT synthesis at 363 K, sonication time: 10 min).

from XRD patterns. Similar to the trends observed in previous section, it seems that almost nothing happened in the first 2d and the characteristic peak of ZSM-5 appeared after 3 d synthesis. Then, crystallinity increased rapidly with synthesis time. The increase of sonication temperature resulted into higher crystallinity. The sample sonicated at 363 K with the highest sonication power 293 W had the highest crystallinity. This indicates that sonication treatment at higher temperature contributed positively to the overall zeolite crystallization process. Generally, as the temperature increases, the surface tension and viscosity of the mother liquor decrease, which could favor the cavitation effect and create positive impact on the nucleation and crystallization processes [22–24]. Similar to previous section, the increase of sonication temperature had more impact on the crystallization kinetics rather than the nucleation process, as shown in Fig. 9. SEM images in Fig. 10 show highly crystalline spherical samples without any sign of amorphous materials. The crystal sizes are pretty uniform, centered at $\sim 2 \mu\text{m}$. Compared with control experiment (synthesis without seeding and sonication), high-temperature sonication can improve crystallization kinetics substantially and synthesis time can be shortened from 7 d to 5 d.

Based on the obtained result, high sonication power delivered to the seeded mother liquor at synthesis temperature actually combine all regular weapons we can use to accelerate zeolite formation process. However, it seems that sonochemical effect in this study was not powerful enough, especially on the nucleation process, which stimulates us to investigate literature results.

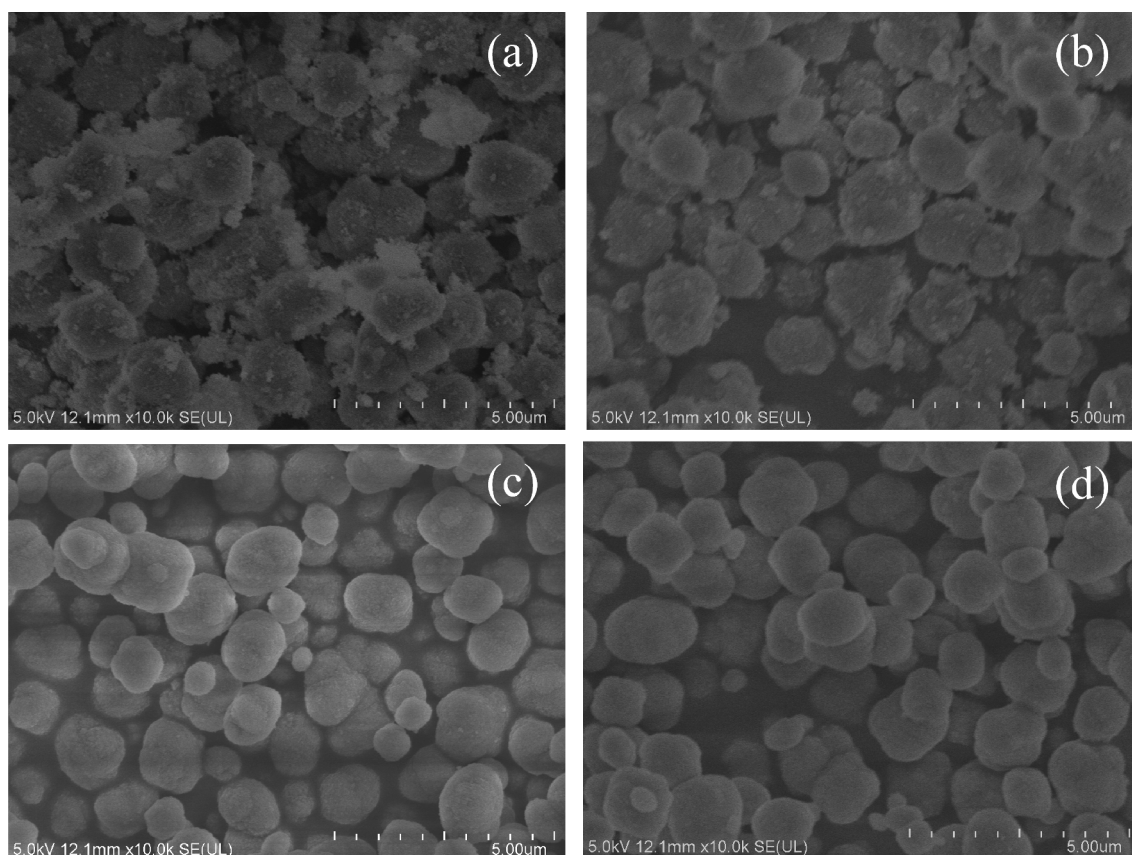


Fig. 7. The effect of sonication power on the SEM images of ZSM-5 samples, (a) 1 % seeding but no sonication, (b) 33 W, (c) 130 W and (d) 293 W (1 % seed, 5 d HT synthesis at 363 K, sonication time: 10 min).

3.4. Mechanism of sonication

It is generally accepted that sonication of mother liquor before hydrothermal synthesis has substantial impact on the formation of zeolite crystals, including nucleation, crystallization, crystal size and morphology etc. The powerful cavitation effect during sonication treatment can create high temperature and high pressure in the mother liquor, which can affect the formation of nuclei and crystallization kinetics by breaking chemical bonds, initiating reactions and preventing agglomeration. Despite the numerous publications about sonication-aided synthesis, the real impact of sonication energy on zeolite nucleation and crystallization is still under debate [22–24].

Table 1 compared sonochemical syntheses of various zeolites, especially the impact of sonication treatment on the induction time, overall synthesis time (induction + crystallization) and crystallization kinetics (slope of crystallization curve, not considering the induction period). As shown in Table 1, sonication of mother liquor led to substantially reduced overall synthesis time in most cases (including zeolite SSZ-13 [35], EMT [37], T [38], A [39], ZSM-5 [41,42], Silicalite-1 [48], SAPO-34 [49]), Y [50], and MCM-22 [31,51], which demonstrated the effectiveness of sonication power on the acceleration of zeolite formation process. However, after further scrutinizing the data (the crystallization curve), we found the effect of sonication treatment can be classified into three categories, A: acceleration of both nucleation and crystallization kinetics; B: acceleration of nucleation only (demonstrated by shorter induction period), and C: acceleration of crystallization kinetics only (demonstrated by the increased slope of crystallization curve). The syntheses of zeolite EMT [37], T [38], A [39], ZSM-5 [42], Silicalite-1 [48] and SAPO-34 [49] fall into category A, where sonication treatment had positive impact on both nucleation and crystallization, as demonstrated by substantially reduced induction period and enhanced

crystallization kinetics. For example, the induction time of zeolite T was significantly shortened from 5 d to 0.5 d, while the crystallization kinetics was increased marginally by ~25 % [38]. For the synthesis of zeolite EMT at 298 K, the reduction of induction period was not huge, down from 18 h to 14 h, while the crystallization kinetics was increased by ~40 % [37]. Regarding the synthesis of ZSM-5, both nucleation (4 h to 0.5 h) and crystallization (+486 %) were significantly improved [42].

The synthesis of MCM-22 (no seeding) [51] and MCM-22 (seeded synthesis) [31] can be grouped into category B, as sonication treatment shortened induction period substantially while the crystallization kinetics was almost the same. The syntheses of zeolite SSZ-13 [35], ZSM-5 [41] and Y [50] belong to category C, as only the crystallization kinetics was improved between 23 % and 72 %, while the induction period was almost the same. Based on the available literature results, we have to say that there is no simple conclusion regarding the real impact of sonication radiation on zeolite crystallization.

Our results on the synthesis of zeolite ZSM-5 suggest sonication treatment mainly affect crystallization kinetics, consistent with category C and ref 41. Certainly, for the same MFI zeolite (including ZSM-5 and Silicalite-1), some researchers' results (ZSM-5 [42] and Silicalite-1 [48]) belong to category A. This discrepancy can only be attributed to the following factors, such as different recipe, synthesis and sonication parameters used in these studies. It should be noted that our synthesis temperature is much lower than conventional synthesis temperature, which may cause different result. More fundamental study is required to elucidate the underlying mechanism of sonication power on zeolite synthesis.

Based on our results, we propose a mechanism of sonication treatment on the crystallization of ZSM-5, as shown in Fig. 11. We believe that sonication energy might not be powerful enough to facilitate the nucleation process, as demonstrated by the same induction period

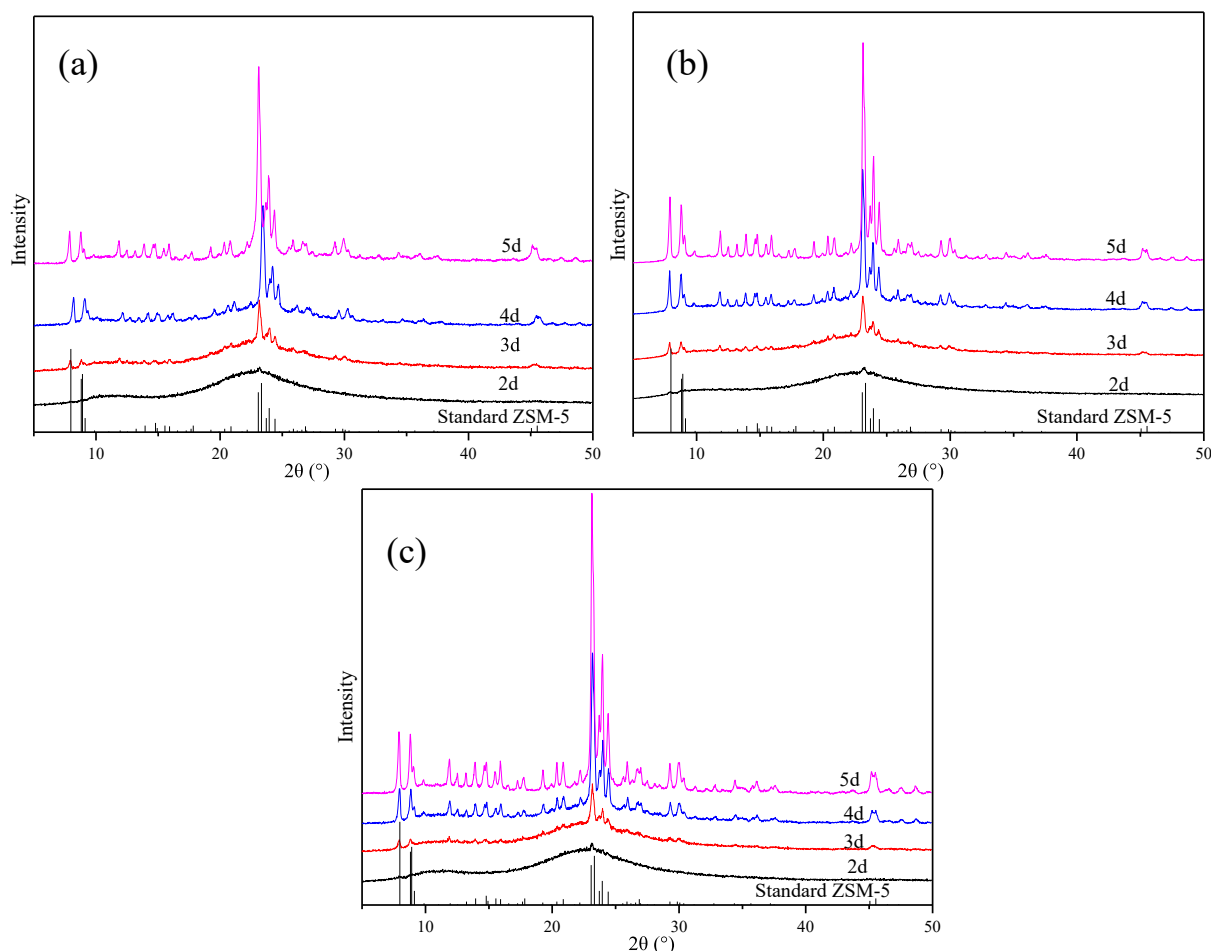


Fig. 8. The effect of sonication time on the XRD patterns of ZSM-5 samples, (a) 298 K, (b) 333 K and (c) 363 K (1 % seed, HT synthesis at 363 K, sonication power: 293 W, sonication time: 10 min).

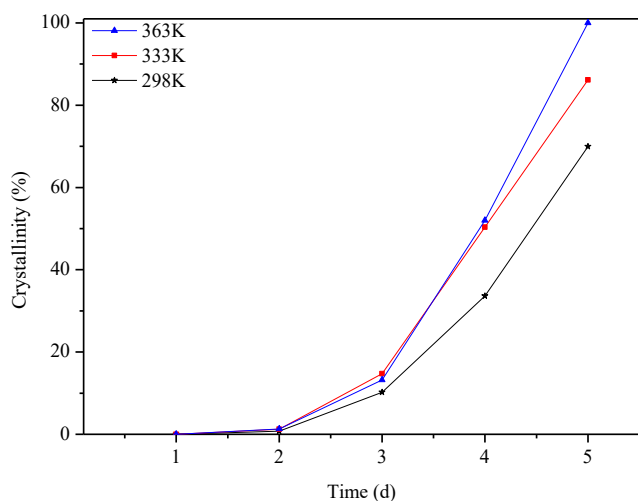


Fig. 9. The effect of sonication power density on the crystallinity ~ synthesis time curves (1 % seed, HT synthesis at 363 K, sonication power: 293 W, sonication time: 10 min).

observed in our study [35]. But sonication can break the added seeds (secondary seeds) and those naturally formed nuclei (primary nucleation in the mother liquor) easily, which creates more tiny nuclei (or even smaller secondary and tertiary building unit) and much higher

crystal surface area. So, in the following hydrothermal synthesis, higher crystallization kinetics was obtained due to the presence of more nuclei and crystal surface in the mother liquor. Most of sonochemical syntheses (if not all) start with a room temperature sonication treatment + a high temperature hydrothermal synthesis without sonication. This indicates that sonication should have little direct impact on the crystallization kinetics. Otherwise, researchers would apply sonication radiation during hydrothermal synthesis. The enhanced crystallization kinetics observed in many studies, including our study, can only be attributed to the de-aggregation and defragmentation of nuclei and seeds, which provide more nuclei for faster crystallization. In our study, we even tried sonication at synthesis temperature (~363 K), however, no proof of enhanced nucleation was observed, as demonstrated by the same induction period. Therefore, conventional sonication at room temperature should have much weaker effect on the nucleation process, which means the real contribution of sonication treatment is downsizing the primary nuclei and secondary seeds rather than enhancing nucleation or crystallization directly. This mechanism can explain our result and many literatures results [35,41,50].

3.5. Other characterizations

Table 2 lists nitrogen adsorption (BET surface area and pore size distribution) of final ZSM-5 samples treated at different conditions. It is worth mentioning that only samples prepared at 363 K for 5 days were listed in Table 2 (S1 to S7). Sample S1 was prepared by a 5 days conventional hydrothermal synthesis at 363 K without the aid of seeding and sonication. It has a low BET surface area of 239 m²/g, which is

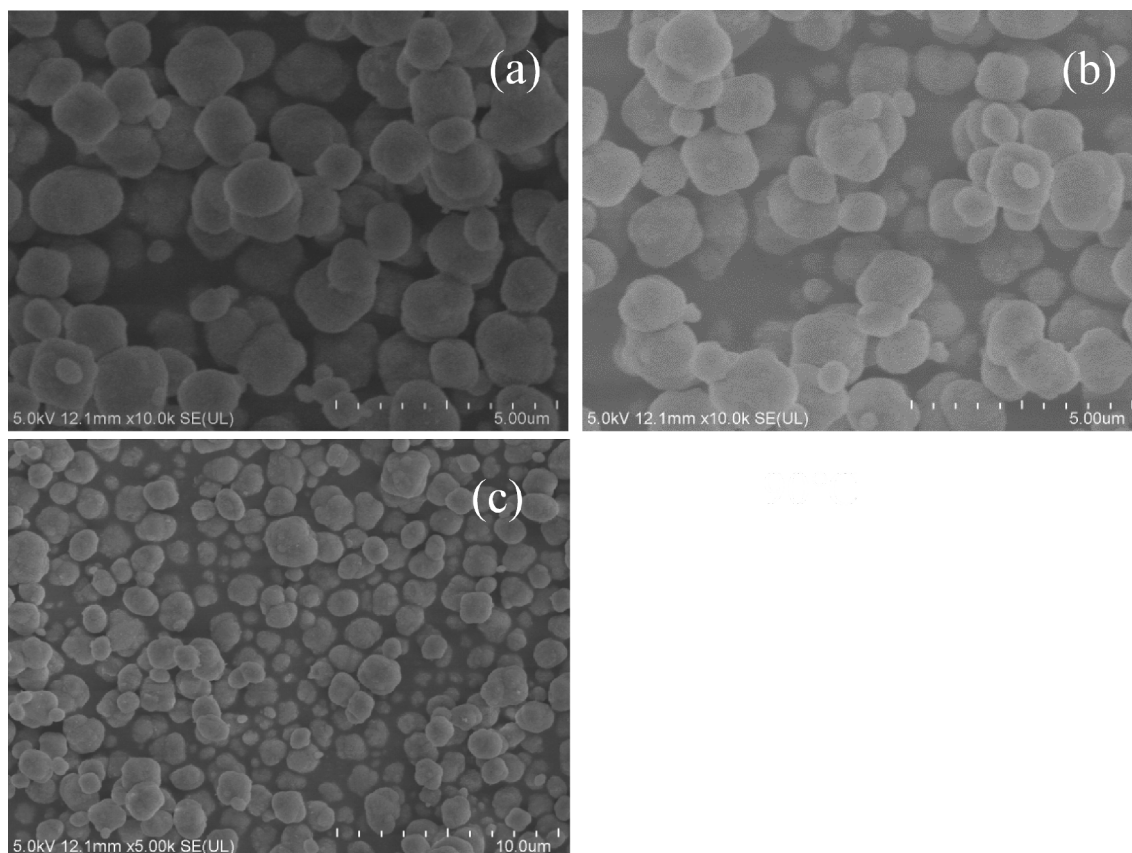


Fig. 10. The effect of sonication time on the SEM images of ZSM-5 samples, (a) 298 K, (b) 333 K and (c) 363 K (1 % seed, 5 d HT synthesis at 363 K, sonication power: 293 W, sonication time: 10 min.

Table 1

Comparison of zeolite synthesis by sonochemical method.

Zeolite structure	Syn temperature	Seeding	Change of synthesis time [#]	Change of induction period	Change of crystallization kinetics [*]	Ref
EMT	298 K	No	35 h → 20 h	18 h → 14 h	+183 %	[37]
T	393 K	No	7d → 2d	5d → 0.5d	+33 %	[38]
A	373 K	No	105 min → 50 min	15 min → 5 min	+100 %	[39]
SAPO-34	473 K	No	2.5 h → 1.5 h	1 h → 0.5 h	+50 %	[49]
ZSM-5	460 K	No	48 h → 8 h	4 h → 0.5 h	486 %	[42]
Silicalite-1	348 K	No	74 h → 55 h	21 h → 15 h	33 %	[48]
MCM-22	423 K	Yes	72 h → 3 h	64 h → 30 h	No change	[31]
MCM-22	431 K	No	10d → 5.5d	4d → 0.1d	10 %	[51]
Y	353 K	Yes	90 min → 70 min	30 min → 30 min	+50 %	[50]
SSZ-13	433 K	No	5d → 3d	6 h → 6 h	+72 %	[35]
ZSM-5	453 K	No	72 h → 60 h	8 h → 8 h	+23 %	[41]
ZSM-5	363 K	Yes	7d → 5d	3d → 3d	+40 %	This study

^{*}crystallization kinetics based on the slope of crystallization curve; [#]synthesis time defined by overall synthesis time (including induction period).

consistent with its poor crystallinity. Increasing synthesis time to 7 days led to a highly crystalline sample with high surface area $\sim 324 \text{ m}^2/\text{g}$ (control experiment). As shown in Table 2, the use of seeding and sonication (sonication power, time and temperature) all had positive contribution to zeolite crystallization, demonstrated by higher surface area compared with sample S1 and control sample. Sample S6 and S7, treated with high sonication power and high temperature sonication, had higher surface area than control sample prepared with 7 d synthesis.

Fig. 12 shows the nitrogen adsorption–desorption isotherm of sample S7, which exhibited mesopores $\sim 4.0 \text{ nm}$ in diameter, calculated by the BJH desorption branch. The introduction of mesopores into microporous zeolitic materials is desirable as the presence of mesopores can facilitate the diffusion of guest molecules into the intrinsic microporosity in zeolite crystals and alleviate diffusion constraint, which could enhance reaction and adsorption kinetics [47,52].

3.6. VOC capture

Volatile organic compounds (VOCs) are common gaseous pollutants in the atmosphere, which create serious threats and damages to our environment, eco-system and human health. It is very important to remove VOC for controlling air quality [53–55]. Adsorption process is a simple and effective method to remove VOCs from air. Zeolites are commonly used as sorbents for VOCs capture due to their tunable hydrophobicity&hydrophilicity, high adsorption capacity and selectivity, outstanding stability and safety etc [53–58]. VOC-laden air has characteristics of large volume (up to $10^5 \text{ m}^3/\text{h}$) and low concentration (usually in the range 300–500 ppm), which requires pre-concentration to reduce the burden of final destruction units. VOCs preconcentrators are usually based on adsorption by various porous sorbents, like activated carbon and zeolites. The adsorbents must be assembled in the form

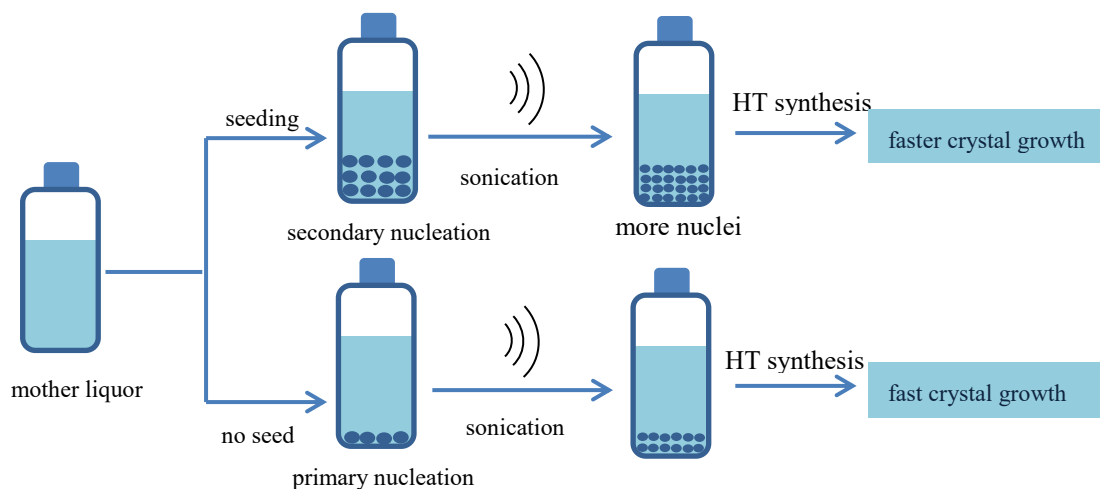


Fig. 11. Reaction mechanism of the increase of crystallization kinetics by sonication.

Table 2

Nitrogen adsorption data of ZSM-5 samples.

Sample	Seeding	Sonication treatment			BET surface area(m ² /g)	Pore size (nm)
		Temp (K)	Time (min)	Power (W)		
S1	No	No	No	No	239	NA
S2	No	298	15	30	244	NA
S3	No	298	20	30	247	NA
S4	Yes	No	No	No	248	NA
S5	Yes	298	10	30	278	NA
S6	Yes	298	10	293	327	3.92
S7	Yes	363	10	293	351	3.95
Control*	No	No	No	No	324	3.87

5 d HT synthesis at 363 K. *Control sample was prepared at 363 K for 7 d without seeding and sonication.

of monolith or honeycomb structure instead of the conventional fixed bed using particles or pellets for the purpose of reducing pressure drop and enhancing rapid response to temperature swing. The unique microporous channel of zeolite is ideal for VOCs adsorption, but it also create significantly mass transfer barrier as the diffusion of VOCs molecules in the tight micropores is extremely slow compared with the unrestricted free diffusion in bulk media. It is well-known that diffusion limitations may substantially limit the reaction and adsorption in zeolite

channels to the external surface of zeolite crystals, which significantly reduce the utilization efficiency of zeolite crystals [47,52,58]. Introducing secondary mesopores into the intrinsic microporous zeolite crystals can significantly increase the number of accessible micropores and also reduce the diffusion length, thereby leading to enhanced catalytic or adsorptive performance.

Fig. 13 shows the break-through curves of isopropanol on mesoporous ZSM-5 samples prepared by conventional hydrothermal synthesis and sonochemical synthesis. Clearly shown in Fig. 13, both samples have similar break-through curves. It should be noted that the adsorption is considered as break-through when VOC concentration in the effluent stream is higher than 40 ppm (10 % of feed concentration = 90 % capture). Compared with conventional ZSM-5 samples without mesopores (commercial sample with crystals size $\sim 3 \mu\text{m}$), the adsorption capacity and break-through time were increased by 30 % and 40 % respectively. This result demonstrates the introduction of mesopores in zeolite crystals alleviates diffusion limitation successfully and leads to better adsorption performance. The adsorption capacity of isopropanol on mesoporous ZSM-5 prepared by sonochemical synthesis was $0.124 \pm 0.02 \text{ mg/g}$, consistent with literature result [54,58]. Table 3 listed the cyclic adsorption capacity of isopropanol on mesoporous ZSM-5 prepared by sonochemical synthesis. The fresh sorbent had adsorption capacity of 0.126 g/g. The adsorption capacity remained constant in 10 adsorption–desorption cycles and the final adsorption capacity was 0.125 g/g, almost the same as original value. This result demonstrated the great stability of our sorbent which is beneficial for industrial

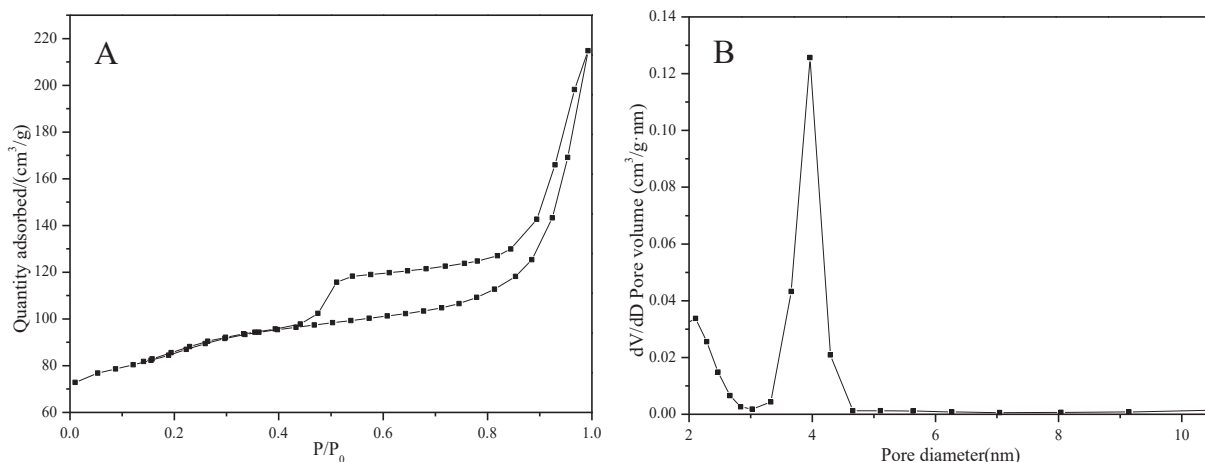


Fig. 12. N₂ adsorption–desorption data of sample S7, (a) isotherm and (b) pore size distribution.

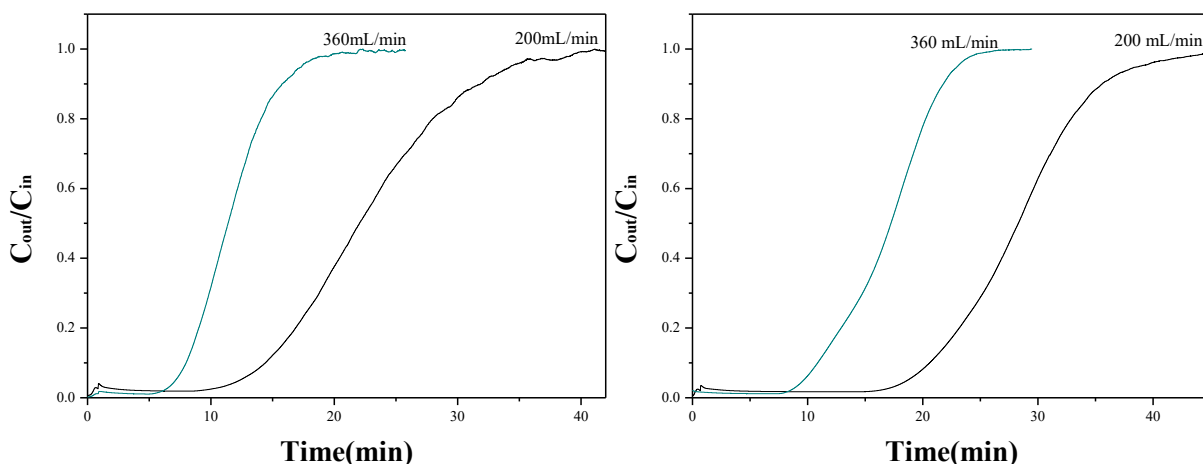


Fig. 13. Effect of isopropanol feed concentration and flow rate on adsorption performance, (a) ZSM-5 prepared by conventional hydrothermal synthesis and (b) mesoporous ZSM-5 prepared by sonochemical synthesis).

Table 3

Cyclic adsorption capacity of isopropanol on mesoporous ZSM-5 prepared by sonochemical synthesis.

Adsorption cycles	1	2	3	4	5	6	7	8	9	10
Adsorption capacity(g/g)	0.126	0.119	0.123	0.121	0.125	0.123	0.122	0.125	0.127	0.125

Feed conc = 400 ppm, flow rate = 200 mL/min, room temperature, desorption at 473 K by nitrogen purge.

applications.

4. Conclusions

In summary, facile synthesis of mesoporous ZSM-5 was successfully achieved at low temperature and ambient pressure by sonochemical synthesis. The combination of seeding, sonication radiation and high-temperature sonication was used to accelerate zeolite formation process. The use of sonication radiation in the synthesis of ZSM-5 increased crystallization kinetics substantially, but exhibited little impact on the nucleation process. We hypothesize that sonication of mother liquor breaks the added seeds or primary nuclei into smaller pieces and increased surface area of seed crystals which leads to faster crystallization kinetics. Various characterization techniques demonstrate that the mesoporous ZSM-5 prepared by sonochemical method has similar physical properties as conventional mesoporous ZSM-5. The introduction of mesopores in microporous ZSM-5 resulted into higher adsorption capacity and longer break-through time, which can be attributed to the alleviated diffusion limitation.

CRediT authorship contribution statement

Longfei Zhou: Investigation, Methodology, Data curation, Writing – original draft. **Mingquan Wang:** Investigation, Methodology, Data curation, Writing – original draft. **Senlin Yang:** Validation. **Wanying Guo:** Validation. **Xiangkai Pu:** Validation. **Yibin He:** Validation. **Jian Zhu:** Validation. **Bin Wang:** Validation. **Meiling Zheng:** Validation. **Song Liu:** Validation. **Yanfeng Zhang:** Conceptualization, Project administration, Supervision, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Author contributions

All authors approved the final version of the manuscript.

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