



## Evaluating the Sintering Temperature Control Towards the Adsorptivity of Ammonia onto the Natural Zeolite Based Hollow Fibre Ceramic Membrane

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### A B S T R A C T

The disproportionate quantity of ammonia presence in water has led to serious drinkable water scarcity worldwide. The abundant source of mineral and superior cations selectivity has made natural zeolite as a good adsorbent for the ammonia removal. This work aims to fabricate natural zeolite based hollow fibre ceramic membrane (HFCM) via extrusion-based phase inversion and sintering techniques for the ammonia removal in wastewater. The physical properties of the fabricated HFCM were investigated through surface morphologies and pure water permeation. The performance of the HFCM for ammonia removal was studied using synthetic wastewater with HFCM prepared at different sintering temperatures. Of all studied sintering temperatures, it was found that 1050 °C was the best fabrication conditions. The membrane also revealed acceptable morphologies (roughness) and water permeation flux of 249.57 L/m<sup>2</sup>.h to which both contributed to the performance of the HFCM. The ammonia removal using the fabricated HFCM gave an outstanding performance with nearly 90% rejection, which probably is due to the synergistic effect of the two processes in the HFCM system, i.e. adsorption and separation. It was found that natural zeolite based HFCM has a great potential to be developed as a single – step ammonia removal in wastewater treatment.

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### NOMENCLATURE

HFCM	Hollow fibre ceramic membrane	AFM	Atomic Force Microscopy
PESf	Polyethersulfone	<b>Greek Symbols</b>	
NMP	N-methyl-2-pyrrolidone	$F = V / At$	Water permeation flux (L/m <sup>2</sup> .h)
SEM	Scanning Electron Spectroscopy	$\frac{(C_i - C_f)}{C_i} \times 100$	Percentage of ammonia removal (%)

## 1. INTRODUCTION<sup>1</sup>

Ammonia is regarded as one of the major contaminants present in the water bodies. This contaminant originated from both municipal and industrial wastes as the concentration varies in the range of 10 to 200 mg/L and 5 to 1000 mg/L, respectively [1]. The industrial activities contributed into the discharge of ammonia included chemical fertilizer, coking, petroleum refining

and pharmaceutical industry. Even though nitrogen compound is essential to numerous organisms, however the excessive presence of this compound can cause a significant pollution burden. The impact of disproportionate release of nitrogen compound into the water streams involved the increment of the demand of dissolved oxygen as it is needed by the aquatic lives, as well as possessed the toxic effects towards the marine species. In addition, the unnecessary nitrogen may disrupt the ecosystem by promoting the algae bloom to that eventually causing the eutrophication in rivers, lakes, estuaries as well as other surface water reservoirs

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[2, 3]. The adverse effect of ammonia has also mentioned by Daud *et al.* [4] in his work of eliminating COD and ammonia from the landfill leachate. Therefore, the removal of these nitrogenous compounds present in the wastewater is of great importance due to these unsolicited consequences.

Previously, there were numeral of approaches have been done to remove ammonia in wastewater such as biological nitrification-denitrification, air stripping, selective ion exchange, breakpoint chlorination, etc. [5-8]. Some of the methods used have offers number of advantages such as easy and continuous operation, simple equipment requirement, suitable for high feed concentration and few others. However, most of these methods suffer from drawbacks related to the high cost and challenging maintenance due to the chemicals used in oxidation and regeneration. For instance, the highly sensitive operation such as in biological treatment, requirement of large stripping tower in air-stripping process and the highly sensitive to pH in breakpoint chlorination have made these techniques of ammonia removal become less favorable [9-11]. Thus, an alternative approach for the effective removal of various volatile contaminants including ammonia has greatly investigated. To date, membrane technology is known to be as one of the best choice for such purpose [12].

Unlike the polymeric membrane counterparts, hollow fibre ceramic membrane (HFCM) has superior properties related to the persistence towards harsh physical and chemical environments such as high pressure and temperature, extreme acidic/basic solutions, and also oxidative/reductive conditions [13, 14]. However, the production of ceramic membrane usually associated with the high cost related to the expensive raw materials as well as high sintering temperature. These drawbacks have inevitably avoided the choosing of ceramic membrane in developing wastewater treatment system. Thus, the emergence of finding the low-cost ceramic material for the development of ceramic membrane has become an urge in these coming days.

The development of hollow fibre membrane provides attractive alternatives for various wastewater treatments. The porous membrane has a wide range of separation/purification applications namely water desalination, drinking water purification, wastewater treatment as well as contaminant removal [15-17]. The usage of the hollow fibre membrane configuration has eventually increased the efficiency of the membrane. The effectiveness was attributed by the high area/volume ratio as well as high packing density as well as the presence of unique asymmetric structure of sponge-like and finger-like separation layer [18]. This unique structure offers a significant decrease in mass transfer resistance, to which particularly advantageous for the separation and purification applications.

Natural zeolites are porous aluminosilicate with valuable physico-chemical properties namely high cation exchange capacity, cation selectivity and high void volume [19]. The primary zeolite framework is the tetrahedron structure. The centre of this structure is occupied by an aluminium or silicon atom to which is covered by four oxygen atoms at the vertices. The substitution of each aluminium ( $Al^{3+}$ ) atom over the silicon ( $Si^{4+}$ ) atoms in zeolite framework creates one negative charge on the framework. Therefore, the higher the substitution of the aluminium atom, the higher the negative charge of the zeolite [20]. The negative charge within the pores is balanced by the cations (positively charge) existing on the surface of the zeolite such as  $Na^+$ ,  $K^+$ ,  $Ca^{2+}$  and  $Mg^{2+}$  [21]. These cations are bonded to the aluminosilicate by a weaker electrostatic interaction that causes the ability of exchange with certain cations presence in the solution. This significant property of natural zeolites makes it capable of exchanging ions with external medium. In addition, the ion exchange performance of natural zeolite is dependable on several factors such as charge density of the anionic framework, ionic charge, framework structure, ion size and shape and the concentration of the external electrolyte solution [22]. In addition, these natural zeolites can have varying chemical composition and cation-exchange capacity (CEC) due to the formation environment. These CEC values lies between 0.6 and 2.3 meq/g. Moreover, ammonia is known as one of compound that has higher affinity towards natural zeolite to which in turn is favorable to be adsorbed by this zeolite.

Among the zeolites, clinoptilolite is the most abundant natural zeolite and is widely used across the globe. The use of natural zeolites for environmental applications is gaining new research interest mainly due to their properties. In addition, the application of natural zeolites for water and wastewater treatment has been realized and still a promising technique in environmental cleaning processes. In the past decades, natural zeolites have been utilized and focused for the removal of ammonium and heavy metals due to the nature of its ion-exchange properties.

This present paper intends to fabricate the alternative inexpensive adsorptive HFCM from natural zeolite and aiming for the removal of ammonia from wastewater. The effect of sintering temperature onto the surface properties of the HFCM and the adsorption of ammonia on top of it were also experimentally tested.

## 2. MATERIALS AND METHODS

**2.1. Materials** The starting materials of natural zeolite (clinoptilolite) powder (with average particle size of 70  $\mu m$ ) was purchased from Liaoning Province,

China (Shijiazhuang Mining Trade Co. Ltd.). Polyethersulfone, PESf (Radal A300, Ameco Performance, USA), N-methyl-2-pyrrolidone, NMP (AR grade, Qrec™) and Arlancel P135 (Polyethyleneglycol 30-dipolyhydroxystearate, Uniqema) were used as polymer binder, solvent and dispersant, respectively. 25% ammonia solution (AR grade, Qrec™) was used to prepare the synthetic ammoniacal wastewater in membrane performance study.

## 2. 2. Fabrication of Natural Zeolite Based HFCM

The low-cost zeolite based HFCM was prepared by phase inversion-based extrusion/sintering technique using the composition shown in Table 1. Firstly, the dispersant, Arlancel P135 was dissolved in NMP under vigorous stirring. Once a homogeneous solution was formed, the pre-dried zeolite powder (45 wt.%) was slowly added into the solution based upon the weight ratio specified in Table 1 and the suspension was subsequently subjected to ball milling using NQM-2 planetary ball mill. After 48 hours of milling, PESf of the desired weight was added into the dope and milling continued for another 48 hours. The resulted suspension was then degassed for 30 minutes at room temperature to eliminate the air bubbles.

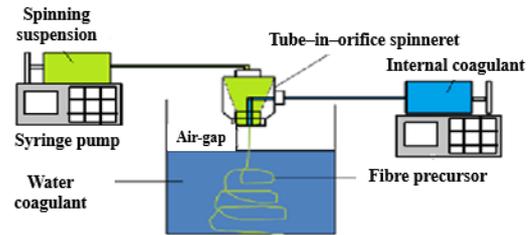
After degassing, the spinning suspension was introduced into a syringe pump and extruded through a tube-in-orifice spinneret using spinning parameter as shown in Table 2. Tap water was used as the internal coagulant. After travelling a certain air gap distance, the fibre membrane green bodies were immersed in a water coagulant bath for 24 hours to allow the completion of the phase inversion process (Figure 1). Afterwards, they were dried at room temperature. Finally, the fibre precursors were calcined in air for 4 hours at various targeted temperatures in the range of 900 - 1100 °C.

**TABLE 1.** Dope suspension composition of the HFCM

Component	Mass % (g)
Zeolite	45
Arlancel	1
PESf	5
NMP	49

**TABLE 2.** HFCM spinning parameter

Component	Value
Ceramic composition (g)	45
Suspension extrusion rate (mL/min)	10
Bore fluid flowrate (mL/min)	15
Air gap (cm)	5



**Figure 1.** Extrusion based phase inversion process of the fabrication of natural zeolite based HFCM [23]

The heating rate was 2 °C min<sup>-1</sup> throughout the heating and cooling process of calcination. The fabrication of the zeolite based HFCM was exactly followed the steps done in previous study with slightly modification [23].

## 2. 3. Characterization of HFCM

The surface morphology of the HFCM was examined using scanning electron microscopy (SEM, Hitachi Model TM 3000). Prior to the SEM analysis, each membrane was coated with silver for 3 minutes under the vacuum condition. These analyses consisted of cross – sectional anatomy of the membranes.

The surface roughness of the HFCM was measured using atomic force microscopy (AFM) (model: PARK XE-100, SCHAEFER Technologies GmbH). The surface roughness was obtained by tip scanning with the dimension of 10 µm x 10 µm.

The water permeation flux ( $F$ , L./m<sup>2</sup>.h<sup>1</sup>) was calculated using Equation (1) where  $V$ ,  $A$  and  $t$  are the volume of the permeate (L), membrane area (m<sup>2</sup>), and time (h), respectively.

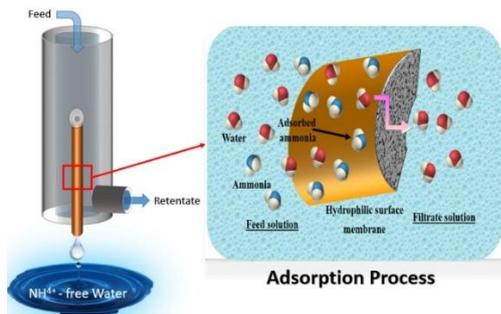
$$F = V / At \quad (1)$$

## 2. 4. Performance of HFCM

The removal of ammonia from the synthetic wastewater was examined via a laboratory made cross - flow permeation cell. The feed solution, prepared by diluting 25% ammonia stock solution to 40 mg/L, was passed through the dead – end epoxy coated HFCMs under the pressure of 1 bar at room temperature. The cumulative permeate volume was measured from time to time and the ammonium concentration in the cumulative permeate was determined by UV–Visible spectrophotometry (DR5000). The adsorption/filtration process was illustrated as in Figure 2. The process was repeated using HFCM sintered at different temperatures. The percentage of ammonia removal was calculated using Equation (2) as follow;

$$\text{Percentage of removal, (\%)} = \frac{(C_i - C_f)}{C_i} \times 100 \quad (2)$$

where,  $C_i$  and  $C_f$  are the initial (feed) and final (permeate) concentrations of ammonia.



**Figure 2.** Schematic diagram for adsorption/filtration process of ammonia by natural zeolite based HFCM.

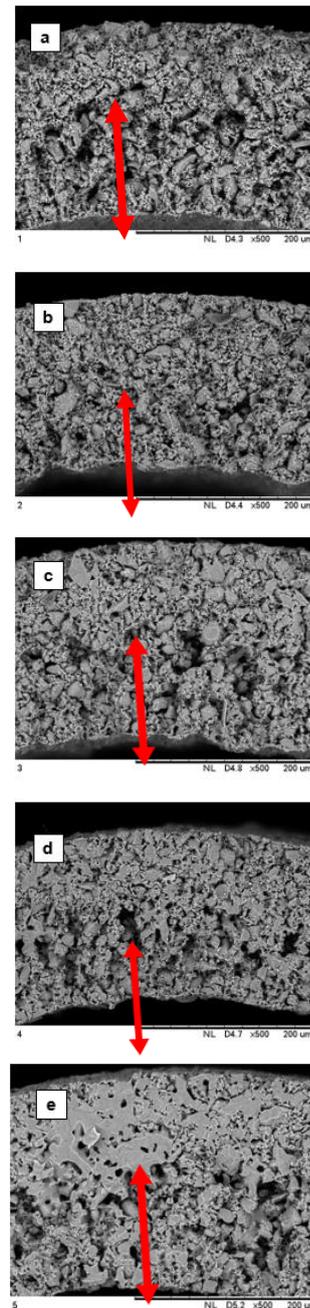
### 3. RESULTS AND DISCUSSION

#### 3. 1. Physicochemical Properties of Natural Zeolite Based HFCM

The microstructure of the HFCMs sintered at different temperatures ranging from 900 – 1100 °C is shown in Figure 3. From the SEM images, it can be obviously seen that all fibres are composed of asymmetric structure with both sponge and finger-like (voids) structures presented in the fibre morphologies. The formation of the finger-like structure (voids) could be attributed by several factors. The formation of finger-like structure is caused by the demixing process of the phase-inversion technique. The interchange between solvent (NMP) with nonsolvent (water coagulant) during the phase-inversion process will not only solidifies the membrane precursor (from ceramic dope suspension), but it rather forms the membrane structural morphology. The high miscibility of solvent-nonsolvent will be favored in forming the finger-like structure whereas the low miscibility of solvent-nonsolvent will be preferred in the sponge-like structure formation [24]. However, it is difficult to achieve finger-like structure in ceramic membrane due to several factors. One of the major factor is due to the irregular shape and size of the particle. Unlike the polymeric membrane counterpart, the ceramic suspension normally consisted of bigger particle (36 µm in this study). This will be eventually affecting the formation of finger-like structure. However, the trend of finger-like structure formation could be observed and denoted as arrow in SEM micrographs in Fig. 3. This finger-like structure or voids could have attributed to the high flux of the membranes. However, this structure is considered as defect by the mean that it sacrifices the mechanical strength of the ceramic membrane [25].

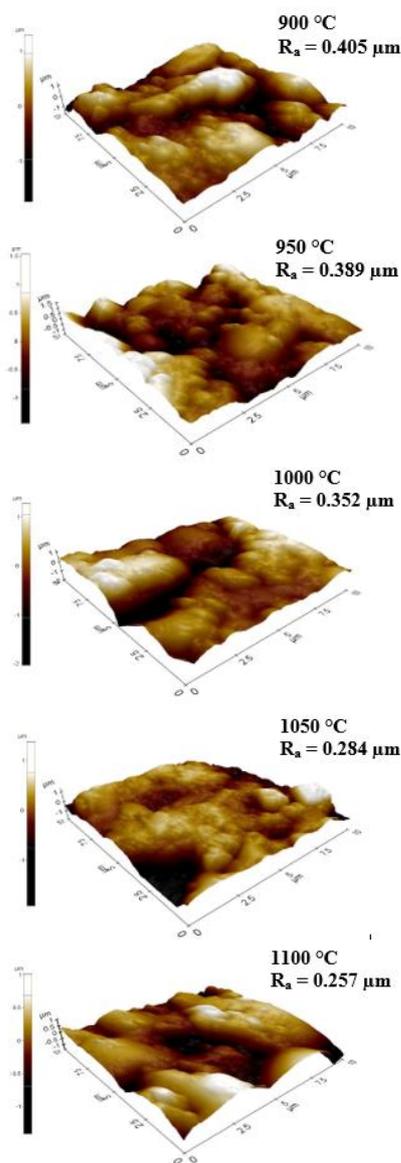
In addition, the HFCM sintered at the lowermost temperature gives the most porous structure with the most loosely packed fine – grains as compared to that of other sintering temperatures. Upon the increment of the sintering temperature, the HFCM became less porous with more densely packed grains. At the highest sintering temperature of 1100°C, the grains started to

melt and were merged. This will eventually form the defect pores due to the spaces in between the particle left. This phenomenon suggesting the increment in sintering neck formation by the diffusion between ceramic particles or the grain growth of the particles in the hollow fibre membrane. A similar trend in membrane morphology was reported elsewhere [26].



**Figure 3.** Cross-sectional morphologies of the HFCM sintered at a) 900 °C, b) 950 °C, c) 1000 °C, d) 1050 °C and e) 1100 °C, respectively. *Note:* The finger-like structure is denoted as arrow in the HFCM cross-sectional micrographs

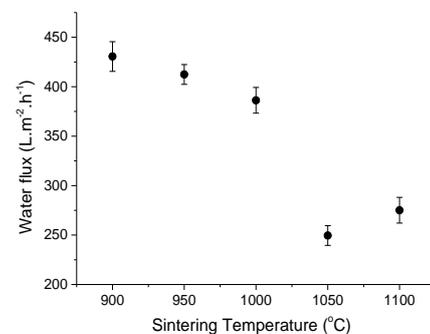
The AFM images of the HFCM sintered at different temperatures are shown in Figure 4. It can be clearly seen from the image that the 3D images and average roughness ( $R_a$ ) were varied upon the increment in the sintering temperature. The increment in the sintering temperature has eventually smooths the surface of the HFCM. This phenomenon could also attributed by the grain growth process during the sintering progression has taken place. The similar phenomenon was also reported in previous study whereby the grain growth of the pure alumina ( $Al_2O_3$ ) was aligned with the increment of the sintering temperature [27].



**Figure 4.** Membrane surface roughness of the HFCMs sintered at different sintering temperatures

This phenomenon has not only increase the particle size of the  $Al_2O_3$  due to necking formation between particles, but it would rather increased the density of the membrane due to the densification occurrence along with the grain growth process. The densification of the ceramic particle in this membrane has eventually smooths the surface of the membrane. In addition, the degradation trend of the surface roughness could have affected the adsorption process of the ammonia onto the HFCM. The smoother surface may offer limited surface area/volume as compared to that of rougher surface and expected to give better adsorption towards the ammonia. However, the adsorption/filtration process merely not only rely on the surface area for the better adsorption.

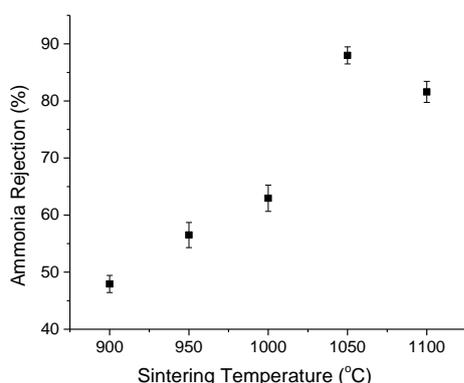
On the other hand, the water permeation of the HFCM has also significantly affected by the different sintering temperature. Figure 5 shows the water permeation profile of the HFCMs sintered at different temperature. From the figure, it can be noticeably seen that the water permeation of the HFCMs sintered at different temperature was mainly controlled by the microstructure of the membrane. The highest water permeation was given by the most porous HFCM (sintered at 900 °C) and the level off trend was observed upon the HFCMs sintered at higher temperatures. Most of the ceramic materials are hydrophilic in nature. However, the high temperature of the sintering process could possibly eliminated all the hydroxyl ( $-OH$ ) groups of the ceramic material [28]. This therefore reduces the degree of hydrophilicity of the membrane. Thus, the permeation of the water onto the membrane could be attributed by this factor. In addition, the density of the membrane structure has also affected its water permeability. The water was easily permeates the membrane that are loosely-packed as compared to the denser membrane (sintered at higher temperature). The increase trend of water permeation for the HFCM sintered 1050 °C to HFCM sintered 1100 °C is likely due to the defect pores formed, as shown in Figure 3. The slow water permeation will afterward leads to the best adsorption and removal of ammonia.



**Figure 5.** Water permeation profile of the HFCMs sintered at different temperature (n=3)

### 3. 2. Ammonia Removal Performance of the HFCM

The removal of ammonia from the wastewater was performed using HFCM sintered at different temperatures. Figure 6 showed the percentage of ammonia removal after 3 hours of continuous adsorption/filtration process whereby the concentration of ammonia in the feed solution as well as permeate solution were measured. The performance of the HFCM was calculated using Equation (2). The highest removal of ammonia was obtained from HFCM sintered at 1050 °C. Referring to the water permeation data, it was shown that HFCM sintered at 1050 °C possessed the lowest permeability. Therefore, this achievement could be attributed by few assumptions. The higher sintering temperature was attained to produced higher densification phenomenon and smaller pore size formation. Thus, there was decreases in the permeation rate and resulting in the higher ammonia removal. Similar trend of study was achieved by Ahmadiannamini et al. [29]. The removal of total ammoniacal nitrogen (TAN) out of the wastewater was about 90 % of the feed solution. However, this approach of mixed-matrix membrane could have some limitations namely the possibility of polymer matrix membrane collapse during harsh operational conditions (high pressure and temperature), the leach out of incorporated inorganic material (natural zeolite) of the membrane matrices as well as low water permeability of the polymer membrane matrices. Therefore, it could be concisely concluded that the ion-exchange properties of natural zeolite also believed to be the major factor towards the adsorptivity of the ammonia and merely not caused by the adsorption of ammonia via -OH group which likely be dehydrated during the sintering technique.



**Figure 6.** Ammonia rejection with adsorbent of HFCMs sintered at varied temperatures (n = 3).

### 4. CONCLUSION

A novel hybrid adsorptive zeolite based HFCM and its potential application in removing ammonia from wastewater was successfully proposed. The fabricated HFCM sintered at 1050 °C, achieved the best performance in terms of both permeation flux and ammonia removal. This assuring accomplishment of hybrid adsorptive HFCM indicates a remarkable capability of this membrane to be used in other water treatment membrane systems such as membrane distillation, membrane contactor and others in the future.

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Phase Inversion

Sintering, Ammonia Removal

مقدار نامتناسبی از حضور آمونیاک در آب منجر به کمبود آب آشامیدنی شدید در سراسر جهان شده است. فراوانی منبع و جذب فوق العاده کاتیون های زئولیت طبیعی از این ماده یک جاذب خوب برای حذف آمونیاک ساخته است. هدف این کار ساخت فیبر توخالی با غشای سرامیکی مبتنی بر تکنیک های اکستروژن از جنس زئولیت طبیعی و حذف آمونیاک فاضلاب با استفاده از روش وارونگی فاز و گرما دادن است. خواص فیزیکی نمونه های ساخته شده با استفاده از مورفولوژی سطح و نفوذ آب خالص مورد بررسی قرار گرفته است. عملکرد نمونه های متفاوت گرما داده شده در دماهای مختلف برای جذب آمونیاک روی فاضلاب صنعتی مطالعه شده است. بین همه دماهای مختلف ۱۰۵۰ درجه سانتیگراد بهترین دمای ساخت است. غشاء همچنین مورفولوژی قابل قبول (زبری) و شار نفوذ آب ۲۴۹،۵۷ لیتر بر متر مربع را نشان میدهد که هر دو نشان دهنده عملکرد نمونه ها می باشد. حذف آمونیاک با استفاده از نمونه های ساخته شده با عملکرد عالی تقریباً ۹۰ درصد احتمالاً به دلیل اثر متقابل دو فرآیند جذب و جداسازی در نمونه ها می باشد. مشخص شد که نمونه های ساخته شده مبتنی بر زئولیت طبیعی دارای یک پتانسیل عالی برای حذف تک مرحله ای آمونیاک از فاضلاب است.

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