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# Effect of phase composition of natural quartz raw material on characterization of microfiltration ceramic membranes

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ABSTRACT

The results of development of multi-layer ceramic membranes on the basis of natural quartz raw material from Mongolia are presented. The influence of the phase composition and temperature of calcination on the porosity, morphology and mechanical strength of large-porous ceramic support obtained by the method of isostatic pressing was studied. It was established that multi-layer ceramic membranes obtained by the application of water suspension of high-disperse quartz sand of Mongolia and alumosilicate binder with the addition of 15–35 wt% of quartz are characterized by optimal properties. The developed tubular ceramic membranes with the average pore size 5.3 μm, coefficient of air permeability $(4.17–4.41) \times 10^{-13}$ $m^2$, productivity by water 46.3–48.0 $m^3/(h \times m^2 \times bar)$ and mechanical strength 2.27–2.53 MPa are perspective for wide use in microfiltration processes.



## 1. Introduction

Micro and ultra-filtration membranes find wide use in the processes of water purification, separation and concentration of multi-component liquid mixtures [1–3]. Polymer membranes are most frequently used for such a purpose, whereas ceramic membranes have several advantages upon them: the resistance to increased temperatures and chemically aggressive media, high mechanical strength, long term of exploitation [4]. The main factors that hold wide practical use of ceramic membrane are their high cost and low density of packing of membrane elements compared to roll or polyfibrous polymer membranes [5]. Due to this the growth of the number of publications dedicated to the study of cheap ceramic membranes on the basis of natural raw materials (clay minerals, natural zeolites and etc.) and their use in the processes of purification of natural and industrial sewage waters is observed [6–9]. Also, thanks to the development of tubular multichannel and poly-fiber ceramic membranes substantial success is achieved while solving the task of increasing the density of their packing [10–12].

Natural quartz sands that are a widely used raw material around the world predominantly consist of crystal silicon dioxide, which makes them attractive for the creation of ceramic membranes. The developed technology of obtaining ceramic membranes on the basis of quartz sand [13–15] includes the stages of formation of porous tubular support by the method of radial isostatic pressing and its subsequent application on the outer surface of membrane layers. Such an approach is successfully realized at obtaining tubular ceramic membranes from natural quartz sand of Belarus.

It is shown in [16] that at the use of quartz sand of different deposits it is important to consider the influence of size and shape of the particles, pressurizing pressure, conditions of drying and thermal treatment on exploitation properties of membranes but also to take into account phase and chemical transformations occurring at thermal treatment of porous support and fixing of membrane layers. This is reasoned by substantial differences in phase and granulometric composition of quartz sand, structure of mineral mixture (presence of dust-like fraction) and separate particles (crust formations, surface formations of new phase) caused by natural conditions of their formation.

The purpose of this work is to develop and study the properties of multi-layer ceramic membranes on the basis of natural quartz

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raw material from Mongolia for microfiltration.

## 2. Materials and methods

### 2.1. Membranes

Natural quartz raw material from Mongolia (Hustain nuruu area, Lun sumon, Tove aimag) of fraction 200–400 μm was used to obtain ceramic membranes. Quartz sand of Belarus of analogous fraction was used for comparison. Inorganic binder (water solution of sodium alumosilicate), organic burning additive (flower), plasticizer (clay mineral material, non-ionogenic surface active substance (1% water solution of OS-20) were used as components of ceramic mixture. In order to optimize the component composition of the formed ceramic mixture, the samples of tablets (diameter 19 mm, height 10–13 mm) pressurized on hydraulic laboratory press were used for optimization of the component composition of the formed ceramic mixture.

The sintering of the samples was performed in a laboratory oven SNOL 7.2/1100 in an air medium at 830, 850, 870 °C, speed of heating −1 °C/min. The formation of porous ceramic supports in the shape of tubes (diameter 65 mm, thickness of the wall 5 mm, length 500 mm) was performed by the method of radial isostatic pressing on the machine URP 02.00.

Microfiltration membrane layers were applied on the outer surface of tubular supports using the suspension of quartz sand from Mongolia with the additives of crushed quartz (size of the particles 40–70 μm and < 40 μm) in water solution of alumosilicate binder (10 wt%). The contents of disperse phase in the suspension was 30–50 wt%. The fixing of the membrane layers was performed by drying at a room temperature during 24 h with subsequent thermal treatment during 5 h at 600 °C, speed of heating 5°/min.

### 2.2. Investigation of the properties of ceramic membranes

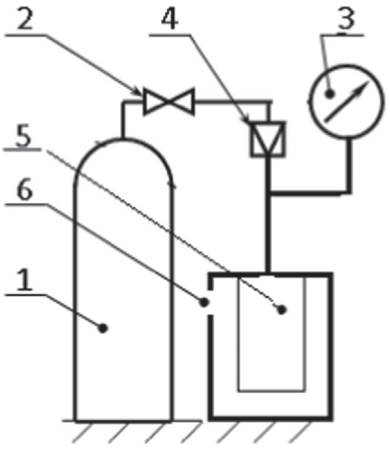
X-ray analysis of natural quartz sands and ceramic samples was performed on the diffractometer DRONE-3 in Cu-$K_\alpha$-monochromatized radiation at reflection angles 2Θ from 20° to 80°. The initial processing of obtained diffraction data and phase identification of the mixture of crystal compounds of the investigated samples was performed using software packages «Powder X» and «WinXpow»(Version 1.04, Jan.–1999) and the basis of x-ray powder standards «JCPDS PDF2» (Version 1.21, May-1999).

The structure of the surface, morphology of breaks of the ceramics and initial powders of crystal silicon dioxide and its chemical composition were studied on a scanning electron microscope JSM-5610 LV with the system of point chemical analysis EDX JED-2201 JEOL (Japan).

The estimation of the compressive mechanical strength of the samples in tablet form (diameter 19 mm, height 10–13 mm) was performed on laboratory hydraulic press by measuring the value of pressure, destroying the unity of sample during the process of testing. The evaluation of mechanical strength of tubular samples was determined on the experimental unit (Fig. 1) by measuring the pressure when the destruction of the sample occurs while applying compressed nitrogen to the interior surface of the sample. Given the high porosity and permeability of the samples, the inner surface of pores of the sample was blocked with a thin plastic film. The length of the samples in all the tests was 70 mm. The average values of mechanical strength were determined using 5 parallel tests. Taking into account that 5 samples are insufficient to obtain an acceptable error for ceramics we need to take into account existing considerable measurement errors.

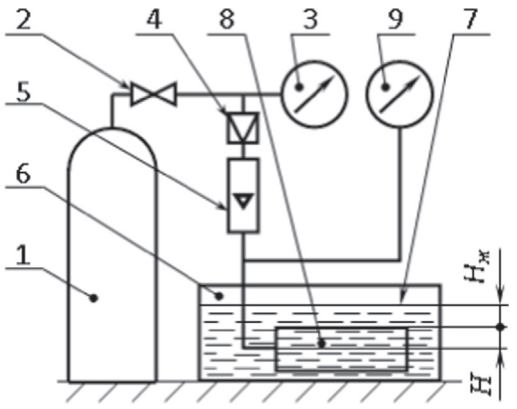
The open porosity (P) and water adsorption (W) of the samples were determined by the method of hydrostatic weighing [17]. The size of the pores of ceramic membranes was determined by the method of bubble in water ("bubble point") on the experimental unit (Fig. 2).

The sample 8 was impregnated with water and placed into the vessel 6 filled with water 7 on the depth 7–10 cm. Afterwards the gas was supplied to the inside cavity of the sample 8 from the




1 – vessel with nitrogen, 2 – tap, 3 – manometer, 4 – reductor, 5 – sample, 6 – vessel with sample

**Fig. 1.** The scheme of experimental unit for the determination of mechanical strength of tubular samples.




1 – vessel with nitrogen, 2 – tap, 3 – manometer, 4 – reductor, 5 – consumption meter, 6 – vessel for liquid, 7 – liquid, 8 – sample, 9 – manometer

**Fig. 2.** The scheme of the experimental unit for the determination of the pore size.

**Table 1**
Phase composition of initial quartz sands and ceramic materials on their basis (temperature of calcination – 850 °C).

| Sample | | Phase composition, wt% Quartz | Tridimite | Albite | Microclean | Flogopite |
|---|---|---|---|---|---|---|
| Belarus | Sand | 100 | – | – | – | – |
| | Material | 95 | 5 | – | – | – |
| Mongolia | Sand | 72 | – | 22 | 3 | 3 |
| | Material[a] | 58 | – | 30[b] | 12[c] | – |

[a] Table 2, series 2.
[b] $Na_{0.986}Al_{1.005}Si_{2.995}O_8$.
[c] $K_{0.94}Na_{0.06}Al_{0.95}Si_{3.05}O_8$.

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