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Effect of Sintering Process and Starch Amount on the Porosity and Permeability of Al_2O_3-ZrO_2 High Porous Oil Filters

Abstract- Alumina-Zirconia is prepared for oil filter. Oil filters are used to separate oil derivatives from each other using separation principles depending on the size of the holes. These filters work on the principle of separation depending on the density and viscosity of liquid or on the difference in impurity crystals size. This work involves preparation of filters from Alumina-Zirconia powder materials by a hybrid freeze casting and space-holder method. These filters have excellent properties such as stability at high temperature, excellent corrosion resistance; withstand static stresses, and other unique thermal properties. Alumina-Zirconia powder is mixed with different amounts of (zero, 15, 25, and 35 vol. %) of starch powder. Powder mixture has been blending with water using an electric mixer to obtain homogeneous slurry. Solid: liquid ratio of slurry of 30:70 is poured in a cylindrical metal molds and freezed by liquid nitrogen chamber. The solidified material was heat treated at 300 °C for 60 min then sintered at 1550 °C, 1600 °C and 1700 °C under vacuum for 120 min. Hot samples were cooled inside furnace until room temperature. The sintered materials were examined to show the effect of starch adding and sintering process on the porous structure and permeability ratio of the fluid using Archimedes method and SEM image analyzed by J-image program. Best permeability and homogeneous porous structure are obtained at 1600 °C sintering temperature with 24 vol. % of starch powder.

Keywords-*Oil derivatives ceramic filters, Al2O3-ZrO2 porous structure, hybrid freeze casting and space-holder method.*

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1. Introduction

Ceramic filters are used as emissions reductionism of electric power stations [1], in gas fuels filters and bio-implants, for the recovery and refining gases process, for incarnation of low-level radioactive wastes, for compressed system of air, for water filters and plugs of lead acid batteries [2,3]. Modern ceramic-filters methods are provided a high porous ceramic which is sintered at an elevated temperature more than 1000 °C [4,5]. The ceramic filter is chemically inert and can be stored without any effectively lose. Although, the ceramic filters have a collection efficiency down to particle size of 25 to 5 microns and more than 90 to 99% [6].
Freeze-casting method, in recent decades, the increasing interest to develop of pores-ceramic parts with hierarchical porous structure with maintaining good mechanical characteristics led the scientific to spot-focus on other shaping techniques to substitute the traditional methods [1, 7]. Freeze-casting also called ice-templating, is a unique fabrication technique for the prepared of porous with hierarchical ordinary ceramic microstructures. It consists of freezing the slurry of ceramic followed by the sublimation of the slurry by drying at both low temperature and pressure [8-10]. The freezing of the slurry created a repetitive pattern, the growing of perpendicular ice crystals along the freezing path and the associated denial of ceramic particles between these crystals. Latest, the green body after solvent deletion by freeze-drying is sintered for consolidation and the final freeze-cast sample exhibitions hierarchically and perpendicularly associated porosity, which is the model of the original solvent crystals [11]. Sacrificial Fugitives "Space-Holding Fillers" Solid material in powdered form can be used for creation cellular ceramic structures [12]. The powder still at solid state during this process and simply goes finished a sintering action or other solid state processes **[13,**14]. Space holders' materials contents ceramic particles or high porous spheres, polymer and organic grains, salts or even low melt point metals. The packed bulk is then both simply compressed at room temperature and, if the space-holder is heat resistant, so can be pressed at elevated temperatures to develop compaction and sintering processes tack place between the powder particles [15].
Ranier et al. [16] was created porous Fe_2O_3 scaffolds by freeze-casting and sintering processes, this scaffold used as artificial implants. New multifunctional porous Yb2SiO5 ceramics prepared using freeze-casting this study is

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published by Rubing et al. [17] they were prepared porous silicon, mullite-nitride ceramics via pressure less freeze-casting method.
In this work, a hybrid freeze-casting and space-holder presses is used to prepared a porous ceramic structure with high amount of porosity used as a perpendicular straight oil filters. Prepared filters are tested by Archimedes method to measure the permeability them.

2. Experimental Work

I. Materials

Al_2O_3 powder was mixed with 10 vol. % ZrO_2 (Japan, TZ-3YE). The density and particle size of the Al_2O_3 green compacted were 4.053 g/cm^3 and 2.7 µm. The particle size and density of the ZrO_2 green compact were 4.5 µm and 5.894 g/cm^3, respectively. Belgian starch powder type with 2.6 µm particle size was used as sacrificial materials or "space-holder material". Polyvinyl alcohol PVA powder with 4.2 µm particle size used as the compulsory matters. Table (1) shows the main used materials and their specifications.

II. Preparation of Porous Ceramics

Set powders with a 30 vol. % of solid load were mixed by ball-mill and using a bottle which made by polyethylene with distilled water and alumina balls for 90 minutes. Changed vol. % of starch was used as a space-holder material to increase of porosity amount. While the binder is PVA, ethoxylated acetylenic with 0.35 vol. % as surfactant was used to improve the surfaces of powder particles to enhance attachments. 1 vol. % Citric acid was used to disperse the aggregation of particles. Table 1 shows the chemical composition of slurry. Each sample sits were mixed in electrical mixture at 60 °C. The warm solution poured inside a cylinder-shaped mold made by epoxy (20 mm diameter) endangered by a heat isolating layer that had the underside of the mold strongly crowned. Later, the mold located on a stainless-steel dish, which controlled the low temperature in less than 0 °C, used liquid nitrogen. In this case, the freezing orderly water progressively occurs from the lowest to the highest of the sample leading to the formation of linked open pores because of the adding to space exporter materials. This was affected in unidirectional networks of pores concluded the entire cast form after desertion. The green samples were sensibly impassive from the solid mold; the solution intermediate suspended in a freezing. After the green systems were prepared at 600 °C per hour in the atmosphere without any inert gas or vacuum (at a heating system rate of 2 per min) to release the carbon-based material, they sintered in an electronic quartz-tube furnace in 1550 °C, 1600 °C and 1700 °C for 2 hours. Figure 1 shows the heat-treated and sintering temperature processes.

3. Testing of Porous Ceramics

I. SEM imaging and XRD

SEM sample dimensions were 5mm × 3mm × 3mm SEM Images. Scanning electron microscopy type (Hitachi S-570 SEM, Materials Science and Technology Department- B18-F Room-Mc Nutt Hal-Bishop Rd., Rolla, MO 65401), at an accelerating voltage of 5kV and a working distance of 5 mm. Silicon carbide papers were used with grids of 600, 800 and 1000 within surface grinding steps. After surface preparation process, the samples are secured onto specimen holders by conductive carbon cement and coated with 5 nm layer of gold (20:80) by spattering technique using sputtering device type (Denton Vacuum-model DS 3II U.S). The imaging process is controlled using Smart SEM software. SEM images analyzed by using image-J program to measure the pores size and the distribution of pores.
Crystalline solid phases are branded by X-ray diffractometry using (Philips-model X-Pert, Materials Science and Technology Department, 6B room- straw man James Hall-Bishop Rd., Rolla, MO 6540) with CuKα radiation, Voltage: 40.0 kV, Current: 30.0 mA. The finding is normally from 10º to 80º and a stage size of 0.02 º with a rapidity of 1.2 º per min. The specimen dimensions are 3x3x3 mm, which used for 3 samples for each set.

II. Apparent Open-Porosity and Density

Porous ceramics' densities and porosities were measured by method of water immersion or "Archimedes' process method" agreeing to ASTM C20-00 [18]. The process of submerging technique is to: i) weight of dry samples, ii) weight the immersed samples in the water for 25 h, and iii) weight of saturated samples. The calculations equation of specious density (AD) and proportion of specious open porosity (AP) are [19]:

$$\mathrm{AP} = \frac{W_s - W_d}{W_s - W_i} \quad (1)$$

$$\mathrm{AD} = \frac{W_d}{W_d - W_i} \quad (2)$$

where W_d is the dried weight, W_i is the submerged weight and W_s is the soaked weight.

Table 1: Main used materials and their specifications.

Materials	Chemical Formula	Resource	Thermal Conductivity $W.m^{-1}.K^{-1}$	Size µm	Density g/cm^3	Melting Point °C	Molecular Weight
Main Material							
Alumina	α-Al_2O_3	CT-3000-SG and CT-3000-SDP (Almatis, Germany) Ungranulated powder	18-30	50-30	4.053	2977	101.96
Zirconia	t-ZrO_2	TZ-3YE (Tosoh, Japan) powder	1.7-2.7	35-20	5.894	2715	123.22
Space-Holder							
Belgian Starch Corn	---	Tingkat Perusahaan, 2472 Dua, Prai Industrial Estate (Malaysia) powder		< 3	---	---	---
Gel Material							
Polyvinyl Alcohol Gel	$(C_2H_3C1)_x$	Shanghai Kaidu Industrial Development Co., Ltd. China, white color		< 3	1.45	250	---
Binding Material							
Polyvinyl Chloride PVC	$(C_2H_4O)_x$	Shanghai Kaidu Industrial Development Co., Ltd. China, white color, Gel		---	1.31	228 boiling	---
Other Materials							
Tarttiara-butyl alcohol TBA	Tert-Butanol or 2-Methylpropan-2-ol (2M2P), (CH3)3COH BuOH	Britannica, 234 Building 102, Mofen st. (China) liquid		---	---	25	---
Citric acid	$C_6H_8O_7$	Britannica, 234 Building 102, Mofen st. (China) liquid		---	---	---	---

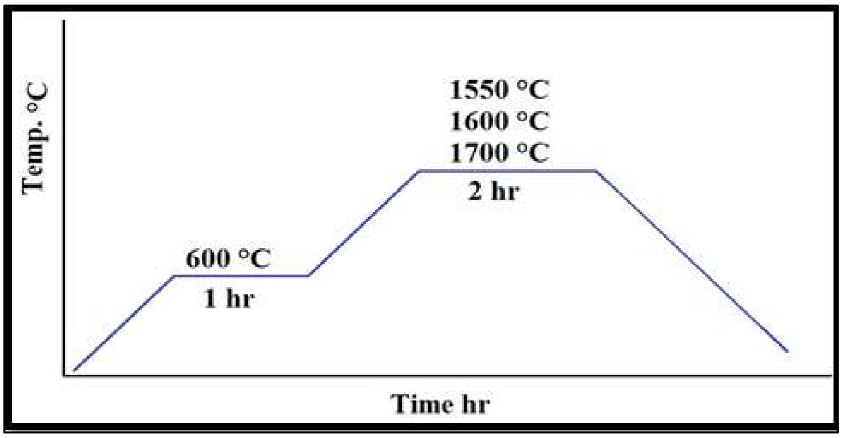

Figure 1: Heat-treated and sintering temperature processes

III. Permeability

Permeability is measured ability of a material to transmit fluids. It is a test of the venting characteristics of rammed foundry sand.

Darcy's law is used to measure the permeability in water, and could be lengthy to extra liquids [18] and which the proportionality endless K could be swapped by k / µ, where k is the permeability of the stone, and µ is the viscosity of the liquid curving through the stone. With this alteration, Darcy's law can be printed in a form appropriate for investigate as [20]:

$$Q = -\frac{kA}{k\mu}\Delta P \quad (3)$$

Where Q is certain in cm^3 /s, k is permeability in Darcy, µ is total viscosity in cP, A in cm^2, L is length in cm, and ΔP is pressure alteration alongside the samples in atm. When a 1 cP viscosity liquid flows at 1 cm^3 /s through samples of cross sectional zone of 1 cm^2 and distance of 1 cm under a compression change of 1 atm [21].

4. Results and Discussion

Figure 2 show outcome of space-holder materials content on the open-porosity with different sintering temperature. In this figure observed that the best porosity result obtained at 25 vol.% of starch. In other side, best sintering temperature is 1600 °C. The main reasons for these results are increased of starch amount lead to increase of open porous percentage [16], that occur at 15 vol.% to 25 vol.% of space-holder material, because of release this material during sintering process and left voids [11]. At 35 vol.% of starch porosity is decreased may be due to agglomerate the starch particles and create a large block [2]. These blocks are broken down, that led to create distorted pores and decrease porosity amount [7]. While the sintering temperature, at 1550 °C is not enough temperature to release all space-holder and other volatile materials. Because of a high amount of starch amount and bonding material, they need elevated sintering temperature to ensure removed all these materials, so 1600 °C is a good sintering temperature. It is removed a largest amount of space-holder and other volatile materials from ceramic samples. However, at 1700 °C this temperature may be over-heating temperature that led to quick swelling of space-holder blocks and form a nonuniform pores [12].

Figures 3 to 5 show the permeability values with different amount of space-holder and sintering temperatures. It is observed the permeability increase with increasing of space-holder material, until 25 vol.% of starch. At 35 vol.% space-holder, the permeability value decrease because of agglomerate of starch particles [15]. The best permeability is recorded at 1600 °C, this temperature provides enough energy to remove all volatile materials.

This variation in the oil permeability values, may be due to the clearly difference in the percentage of connected open-pores [4]. Increases of separation (non-connected) closed-pores mainly were lead to reduced permeability as evident in Figures 4 and 5.

Figure 6 illustrate microstructure of sintered samples. Dimensional macro-pores are detected prepared by sublimation process. Other non-dimensional micro-pores are prepared by removed space-holder material during heating process. X-ray diffraction chart for sintered samples are shown in Figure 7. In this figure three peaks of Al2O3-ZrO2 phase for each sample set according to card no. (04- 003-0649) [22].

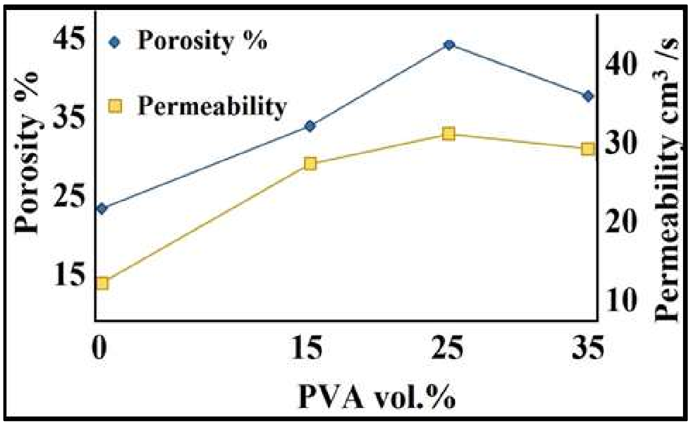

Figure 2: Effect of sintering temperature and PVA vol.% on porosity%.

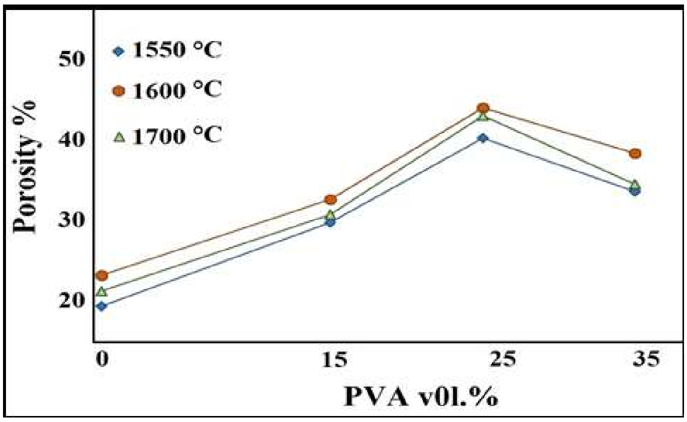

Figure 3: effect of PVA vol.% on both porosity% and permeability at 1550 °C sintering temp

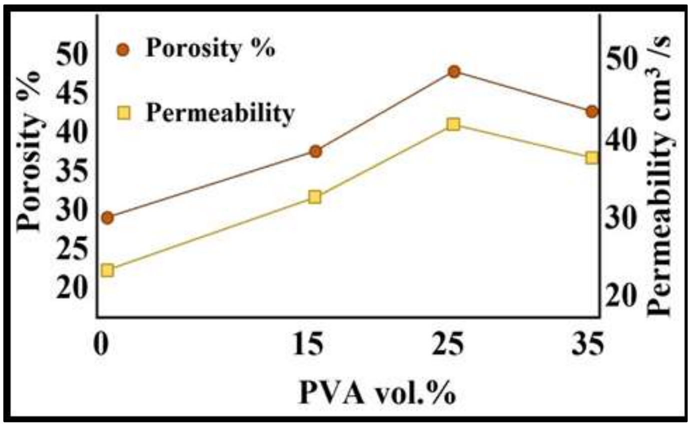

Figure 4: effect of PVA vol.% on both porosity% and permeability at 1600 °C sintering temp.

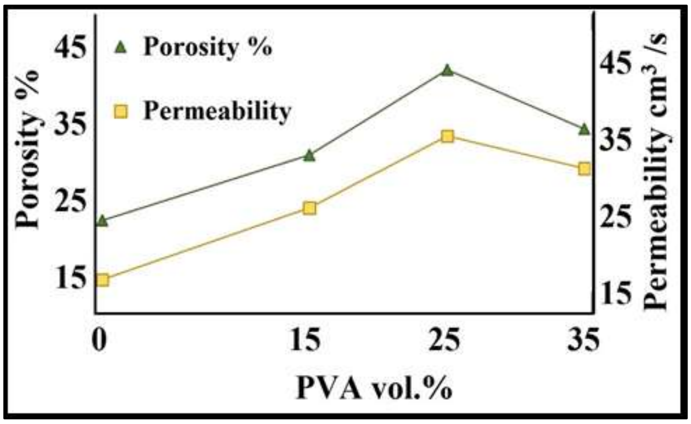

Figure 5: effect of PVA vol.% on both porosity% and permeability at 1700 °C sintering temp.

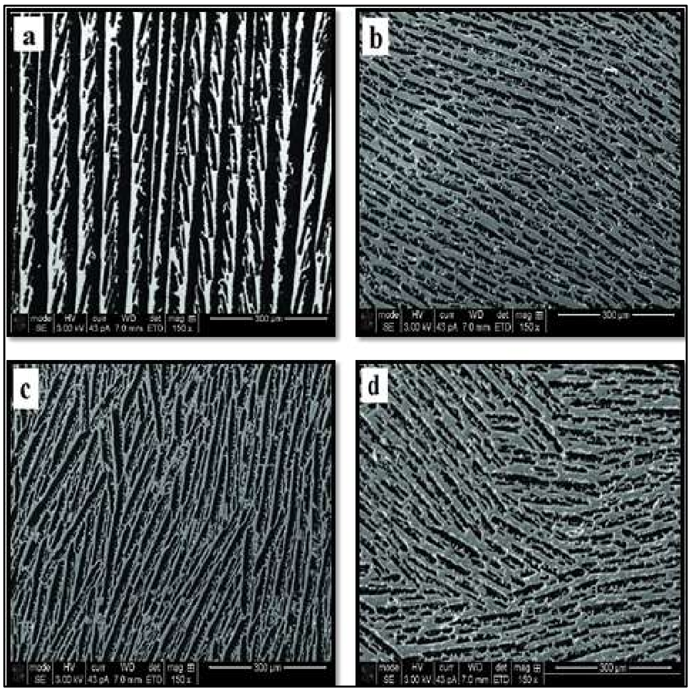

Figure 6: Porous Al_2O_3-ZrO_2 cross section microstructure analyzed by image-J program for: a) zero vol.% space-holder material; b) 15 vol.% space-holder material; c) 25 vol.% space-holder material and; d) 35 vol.% space-holder material.

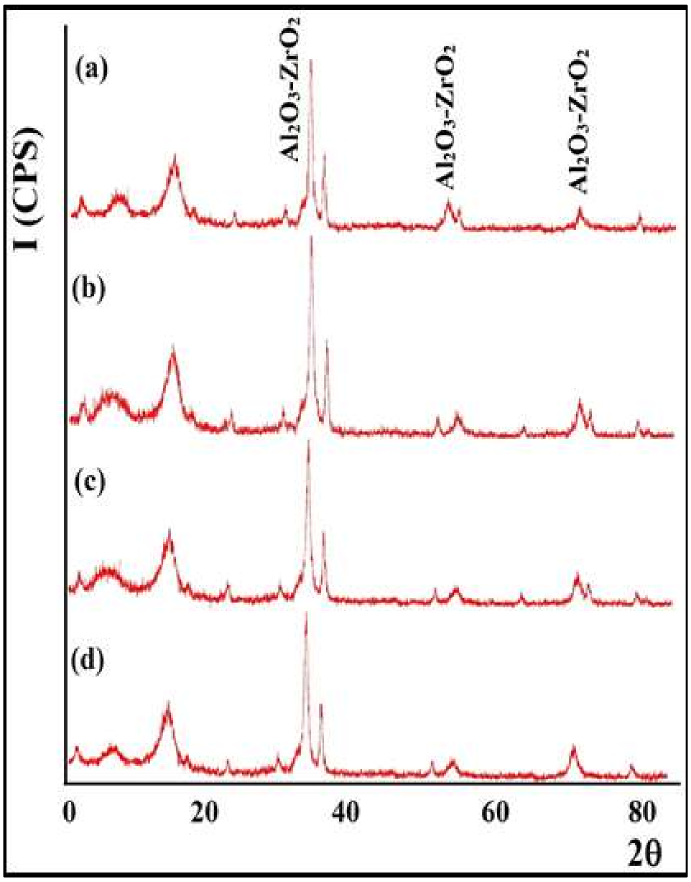

Figure 7: X-ray diffraction peaks of sintered samples; a) zero vol.% space-holder material; b) 15 vol.% space-holder material; c) 25 vol.% space-holder material; d) 35 vol.% space-holder material.

5. Conclusion

The results showed increased permeability with increased porosity. Increasing the amount of starch leads to increase of porosity, therefore increased of permeability. The increase in permeability begins significantly, but the amount of increase decreases with the large increase for starch. Because the agglomeration of starch particles. The best result of permeability was achieved at the temperature of sintering because it is the best grade for the removal of volatiles almost completely.

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