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Use of Organic Binders to Enhance Defluoridation and Pathogen Removal Efficiency of Diatomaceous Earth-Based Ceramic Filters

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ABSTRACT

The use of diatomaceous earth, DE, ceramic membranes in water purification has been in existence for centuries. However, the DE-based membranes are brittle, ineffective in the defluoridation, and disinfection of water. The aim of this work is to improve the mechanical strength, water defluoridation, and filtration efficiency of DE-based ceramic membrane using organic binders; *Abelmoschus esculentus, Aloe vera,* and *Basella alba*. The ceramic membranes were fabricated from DE-powder and plant-based organic binders with a ratio of 2:1 by mass. The dried samples were fired at 700.0 to 1150.0 °C. The fabricated membranes were then made to filter water contaminated with *Escherichia coli*, Rotavirus, and sodium fluoride. The results showed the DE-powder was characterized by 87.5%; 4.0% and 89.6; 2.9% silica and aluminum oxides for DE-A and DE-B respectively. *Basella alba* binder showed the highest content of organic matter and formed the strongest membranes with the highest efficiency. *Basella alba* was able to improve the modulus of rupture, defluoridation, and virus removal efficiency of the DE-B membranes by 84.8%, 30.9%, and 40.3% respectively. DE-B-powder plus *Basella alba* are potential materials in ceramic membranes as they were able to defluoridate by 89.2% and remove Rotavirus by 98.3% from water.

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1. Introduction

The use of organic binders to enhance the mechanical properties of ceramics has been reported by many authors. Baklouti and his colleagues used polyethylene glycol and polyvinyl alcohol organic binders to enhance the adhesive and mechanical properties of the polymer-rich external layer of spray-dried granules [1]. Aduda and his colleagues in their work on ceramic charcoal stoves showed improved bulk density and modulus of rupture (MOR) in green and fired ceramics when the *Corchorus olitorius* binder was used as a binder [2]. Other researchers found that chemically modified starch enhanced mechanical property and could be applied in the manufacture of filter membranes [2]. The effects of natural organic binders extracted from *Abelmoschus esculentus* and *Corchorus olitorius* on the fracture toughness or strength were found to improve the toughness of the fired ceramic membranes [3]. The thermal shock of kaolin-based refractories was also improved by the use of organic binders [3,4]. In 2020, Simiyu and co-workers [5] used *Corchorus olitorius* and showed that the mechanical strength of DE-waste membranes improved by 152.0%. There are, however, no studies on the behaviour of pure diatomaceous earth based-ceramics plasticized with natural organic binders. This work investigates the effects of three natural organic binders extracted from *Abelmoschus esculentus*, *Aloe vera,* and *Basella alba* on the mechanical strength, defluoridation, and disinfection efficiency of DE-based ceramic membranes.

2. Materials and Methods

The two final products of diatomaceous earth (Kensil 90 and Kensil SSF) of African Diatom Industries Limited (ADIL) in Kariandusi, Kenya were investigated in this work. The DE samples were supplied by ADIL in powder form. In this work, Kensil 90 was identified as diatomaceous earth A (DE-A) and Kensil SSF as diatomaceous earth B (DE-B). The samples were subjected to particle analysis using Malvern Panalytical Mastersizer 3000 and DE-A and DE-B ware found to have a mean particle size of 15.3 µm and 12.1 µm respectively. The fresh leaves of, *Abelmoschus esculentus*, *Aloe vera*, and *Basella alba*, were sourced from the Muthurwa Open Market, Nairobi, Kenya. The leaves were washed, drained, and boiled for fifteen minutes in an equal amount of distilled water by mass. The boiled leaves were left to cool and sieved. The filtrate obtained before any further dilution was taken as a 100.0% binder. Subsequent 50.0 % dilution with ionized water was done to get a 1:1 water: binder ratio. All the binders were stored in the fridge at 6.0 ºC to prevent degradation. The organic matter in the binder was determined by the use of a UV spectrophotometer and other techniques like the Kjeldahl technique for proteins, and the use of Benedict solution for carbohydrates [6]. The DE sample and binder were uniformly mixed and cast using a hydraulic press. A pressure of 2.04 MPa was applied to cast discs of 0.5 cm (thickness) by 2.5 cm (diameter) of DE-A and DE-B samples. The discs were dried at room temperature for 72.0 hours. The mass and other dimensions of the drying samples were taken after every 8.0 hours. The dried discs were further heat-treated for 30.0 minutes at 110.0 °C to get rid of trapped water vapor. The dried discs were then fired in an LH 15/14 1706997 Nabortherm furnace at a 3.2 °K/min heating rate to varying maximum firing temperature of 700.0 to 1150.0 °C. The samples were soaked at these temperatures for 2.0 hours. The fired discs were left to cool in ambient air for 48.0 hours. The samples for modulus of rupture test were cast into 15.0 cm (length) by 2.5 cm (diameter) rolls, dried, fired, and classified similarly as the discs. The dimensions of all samples (discs and rolls) were taken after firing. The elemental analysis of the DE-powder, binder, and fired membranes was carried out using an x-ray fluorescence spectrometer (ED XRF CG Spectrometer Rigaku and Empyrean). A three-point bend test was done on five rolls from each sample to determine the modulus of rupture and the porosity of the samples was determined by the Archimedes immersion technique. To determine the efficiency of membranes, 50.0 ml of each contaminated water with 10^8 CFU/ml *E. coli*, 10^8 viral particle/ml Rotavirus, and 50mg/l NaF were filtered and the level of the pathogen in the filtrate was determined by an enzyme-linked immunosorbent assay (ELISA). The pH of NaF and the content of pathogens before and after filtration was used to calculate filter efficiency (x) using Equation 1, where x_i, and x_f are the initial and final levels of pathogens or pH of NaF.

$$x = \frac{x_f - x_i}{x_i} \times 100 \tag{1}$$

3. Results and Interpretation

Characterization of raw materials. Table 1 shows the elemental composition and organic matter content of the DE-A, DE-B, and binders. The silica content was 87.5% and 89.6% for DE-A and DE-B respectively. Aluminum oxide was around 4.0% in DE-A and 2.9% in DE-B and the difference was attributed to the clay minerals present in the DE [7]. The other oxides present in both DE-A and DE-B were CaO, Na_2O, K_2O, and MgO. The presence of Fe_2O_3 was attributed to hematite present in clay minerals. The slight variation in the content of the two DE could be attributed to the age of the fossil and location [8]. The binders showed the presence of Si, P, S, K, and Ca ions which are essential elements in the plants' well-being. Among the three binders, *Abelmoschus esculentus* showed the highest content of silicon ions and carbohydrates. The binder from *Aloe vera* had the least organic matter and low content of carbohydrates and proteins which could be attributed to hydrolysis that occurred during heating as the binders were being prepared. It is thus recommended that the *Aloe vera* binder be used without boiling to maintain its polymeric behaviour.

Table 1: The elemental composition and organic matter content of the DE-A, DE-B, and binders (Abelmoschus esculentus, Aloe vera, and Basella alba)

Oxide	*Diatomaceous earth, DE-A*	*Diatomaceous earth, DE-B*	*Abelmoschus esculentus*	*Aloe vera*	*Basella alba*
Si^{+4}	89.6	87.5	21.7	4.3	18.0
Al^{+3}	4.0	2.9	15.1	4.7	15.2
Fe^{+3}	2.3	2.1	0.1		0.1
Ca^{+2}	1.2	2.6	10.2	5.4	26.0
K^{+}	0. 8	0.7	47.4	22.0	55.9
Na^{+}	1.9	4.2	0.1	0.1	0.1
Mg^{+2}	1.5	1.4	0.5	0.2	0.6
Ti^{+4}	0.3	0.3			
Zn^{+2}	0.1	0.2			
P_2O_5	0.4	0.5	2.5	2.5	2.2
SO_3			0.5	0.4	0.5
MnO			2.1	2.0	6.3
flux			5.5	3.1	11.9
Carbohydrates			31.9	15.8	21.9
Proteins			16.5	6.3	21.9
Fats			6.3	4.8	1.1
Ash			3.1	11.9	5.5

Figure 1 shows the variation of linear drying shrinkage with binder content in dried diatomaceous earth discs. The linear shrinkage increases with an increase in binder concentration. A similar trend, not reported, here was also observed in MOR of the green samples. Samples fabricated with *Basella alba* had a high rate of shrinkage and MOR attributed to high levels of polysaccharides. However, *Aloe vera* had a high content of monosaccharides which denatured upon boiling thus losing its binding ability. Also, samples fabricated with DE-B had a higher shrinkage and MOR than DE-A because of the high content of silica and smaller particle size than DE-A. Shrinkage, generally, was caused by loss of moisture due to the capillary pressure created by a capillary gradient between solid-liquid and solid-vapor interfaces.

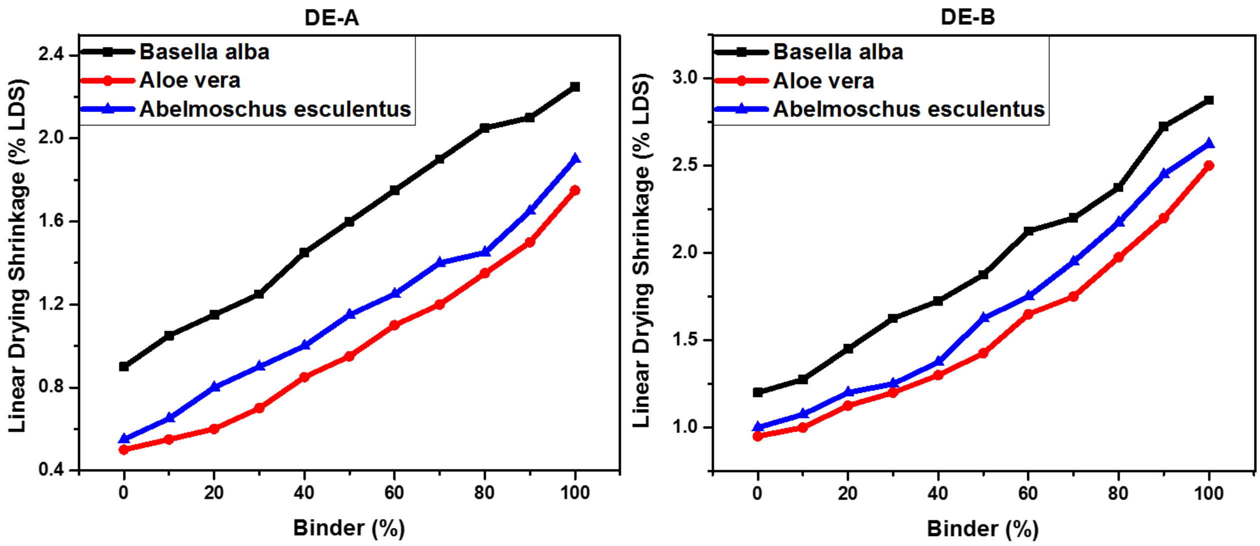

Figure 1: Variation of linear drying shrinkage with binder content in dried DE-A and DE-B discs

Mechanical Strength. Fig. 2 shows the plot of the modulus of rupture (MOR) and porosity against the firing temperature used to fabricate DE-A and DE-B membranes fabricated by *Basella alba, Aloe vera, and Abelmoschus Esculentus* binders. It was observed that the MOR increased with increasing firing temperature. The samples fired at the lowest firing temperature (700.0°C) showed the least MOR whereas those fired at the highest firing temperature showed the highest MOR in all samples. DE-B samples showed higher values of MOR than DE-A which can be attributed to the higher content of silica in the powder. The two samples show a steady increase in MOR as the firing temperature increased up to around 900.0 °C when the MOR increases rapidly up to the maximum firing temperature. This can be attributed to decreased porosity due to increased particle-particle interaction caused by necking processes. Beyond 900.0 °C α-cristobalite starts to turn to β-cristobalite which is more stable and crystalline hence the increased MOR. The average optimal firing temperature for the three binders was found as 964.3 °C, and 919.0 °C for DE-A and DE-B respectively. The average MOR for the membranes at these optimal temperatures were 32.2 MPa and 89.0 MPa with a fraction porosity of 4.1 and 0.38 respectively. The difference in the MOR was a result of the higher content of aluminum in DE-A than in DE-B. Furthermore, the green samples of DE-B were stronger than DE-A hence their higher strength in the fired state. *Basella alba* formed the samples with the highest MOR and least porosity because these samples equally had the highest rate of shrinkage and MOR in their green state hence the tendency to form strong membranes. Furthermore, *Basella alba* had the highest content of protein which has binding surfaces with silica [9].

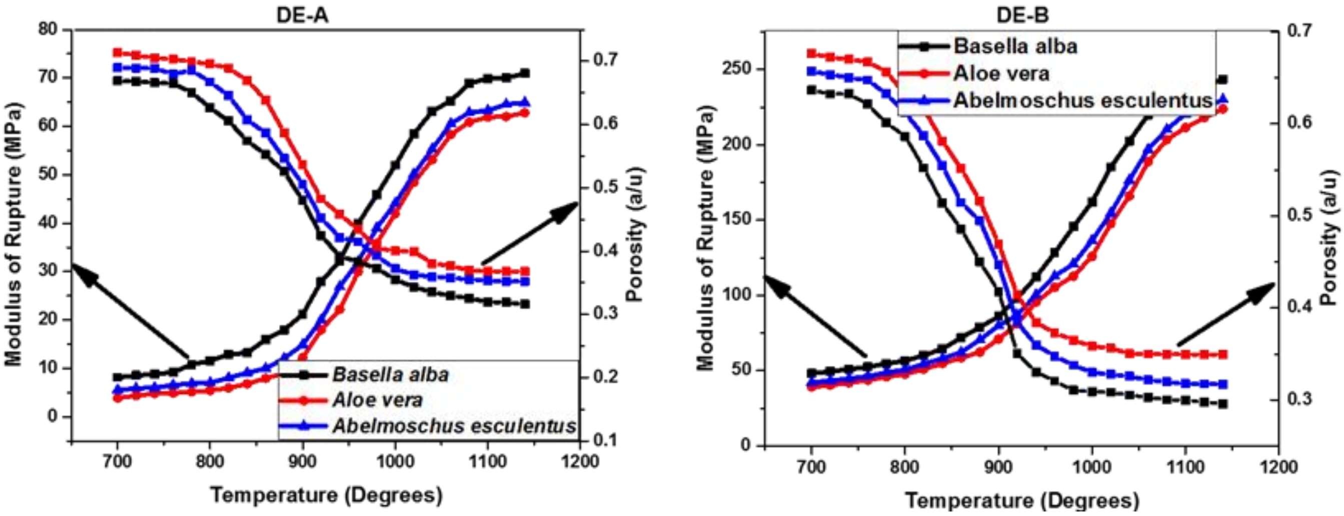

Figure 2: The plot of modulus of rapture and porosity against firing temperature of diatomaceous earth A (DE-A) and diatomaceous earth B samples (DE-B)

Figure 3 (a) and (b) show the plots of modulus of rupture against binder concentration used to fabricate DE-A and DE-B membranes fired at 950.0 °C respectively. In all the binders there was an increase in MOR as binder concentration increased up to 50.0%. The increase in MOR was attributed to an increase in particle-particle interaction enhanced by the polymeric properties of the binders. Once again samples made from *Basella alba* showed the highest MOR than membranes made from the other binders with a 59.5% and 84.8% increase in MOR for DE-A and DE-B respectively. Affirming that the high content of carbohydrates and proteins in organic binders enhances the mechanical properties of ceramics just as it was found in the literature [3,4,5,10]. The plot of porosity against binder concentration initially, shows a decrease in porosity as the concentration increases up to 50.0%, beyond which a further increase in binder concentration results in increased porosity. The decrease in MOR and increase in porosity for binder concentration beyond 50.0% was attributed to too much flux which burn-out creating more voids. The micrographs of DE-A and DE-B membranes fabricated with 50.0% binder and fired at 950.0 °C are shown in Figure 3 (c) and (d). The micrographs in Figure 3 (c) show patterns of hollow cylindrical diatoms of the DE-A membrane. At the same time, the membranes show evidence of high porosity with the pore size of the hollow diatoms being 4.2 µm and 4.6 µm. On the body of the hollow diatom, there are tiny pores of 2.1 nm to 14.4 nm. On the other hand, the micrograph in Figure 3 (d) from the DE-B membrane fired at similar conditions does not show any cylindrical diatom but a broken basket-like one with pores of 2.2 to 11.4 nm and did not exhibit any crystal growth. Thus, at 950.0 °C firing temperature, the membranes did not form crystals.

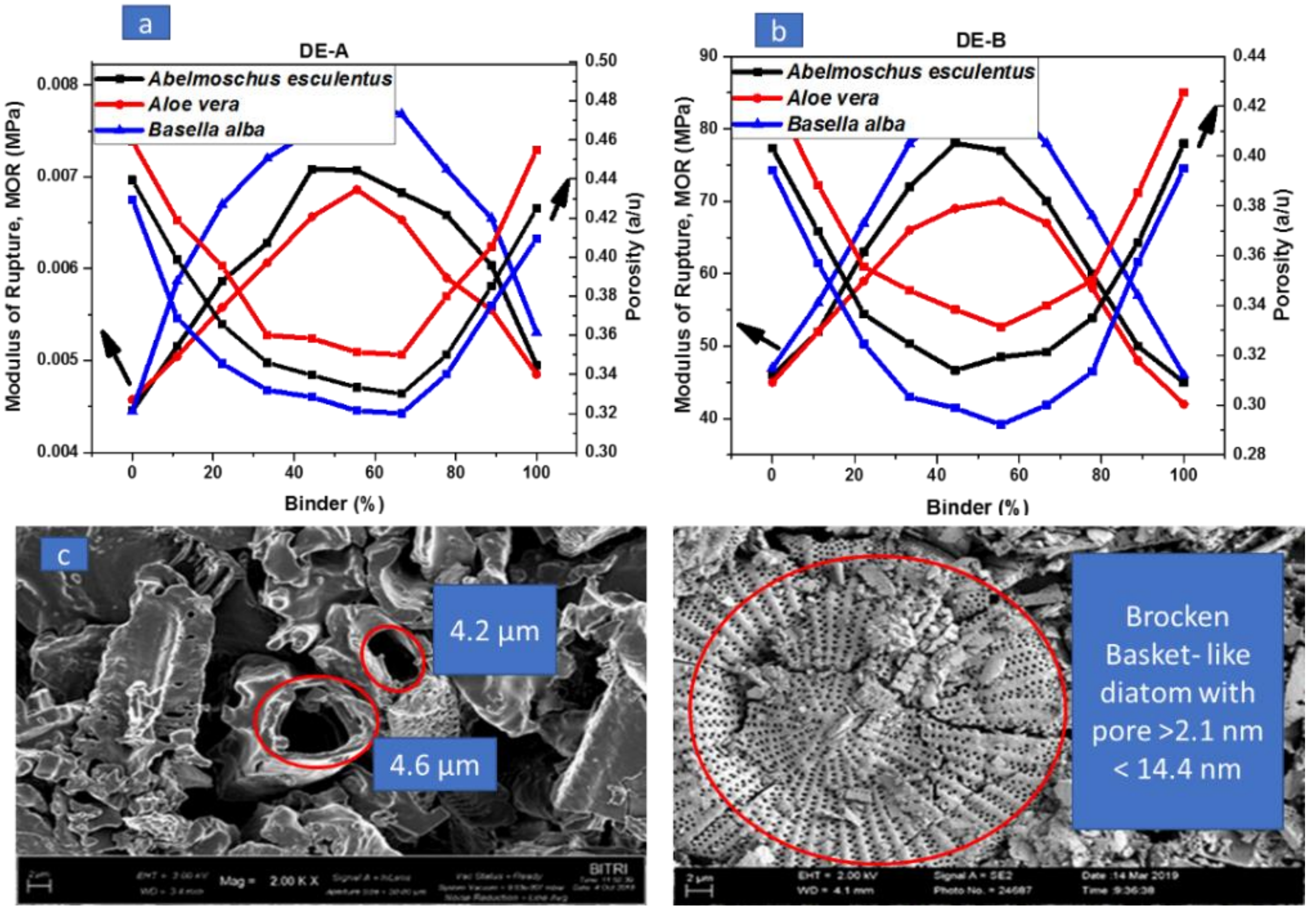

Figure 3: The plot of modulus of rupture against binder concentration used to fabricate diatomaceous earth A (a) and B (b) membranes fired at 950.0 °C and their micrograph

Filtration and Defluoridation. Figure 4 shows the bar chart of the efficiency of the fabricated membrane in filtering water contaminated with *E. coli*, NaF, and Rotavirus. It was found that all DE membranes fabricated were able to decontaminate water due to the electrostatic forces that exist between the filter and the pathogen [11] [21]. All membranes were able to remove more than 99.9% of *E. coli* due to its cylindrical size of 1.0 -2.0 µm that could not pass through the nanopores of the membranes. It was also observed that membranes fabricated from ionized water as a binder were only able to adsorb 56.8 to 58.3% of NaF and Rotavirus respectively. However, when organic binders were used, the defluoridation and virus removal efficiency improved by 28.8; 38.7, and 30.9%; 40.3% for DE-A and DE-B membranes respectively. The improved efficiency was attributed to reduced porosity and improved MOR. Samples fabricated with DE-B showed higher efficiency than DE-A because they formed stronger membranes. Membranes made from *Basella alba* and DE-B were able to defluoridate and remove Rotavirus from water by the efficiency of 89.4 and 98.3%, respectively, which is higher than what was reported by Sobsey and co-workers in 2008 [12].

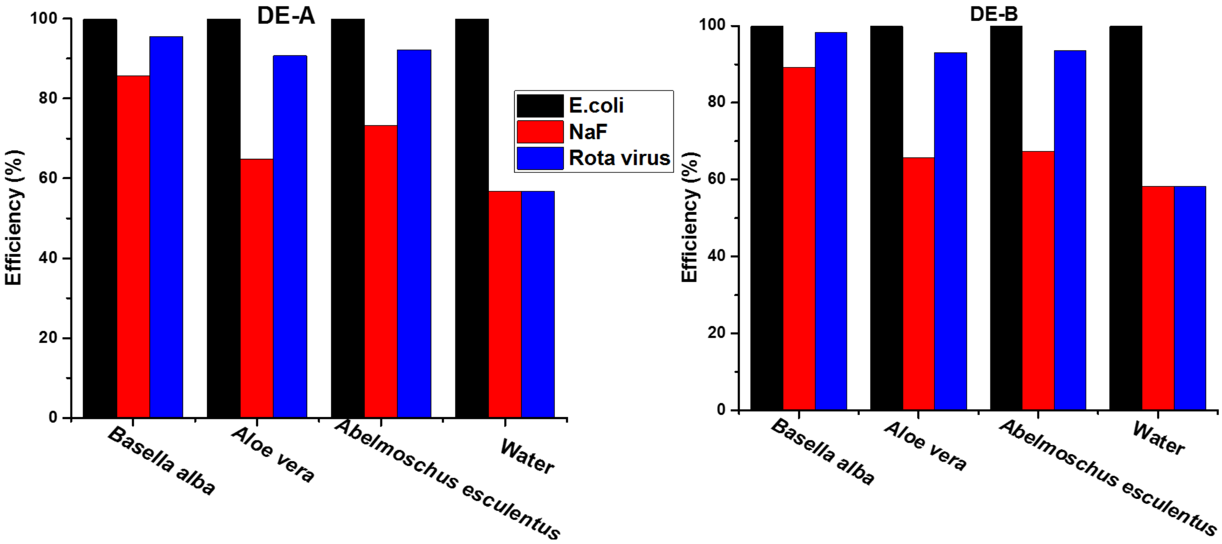

Figure 4: The bar chart of the percentage efficiency of the membrane fabricated with diatomaceous earth (DE-A and DE-B) and different binders: Abelmoschus esculentus, Aloe vera, and Basella alba in filtering water contaminated with E.coli, NaF, and Rotavirus

The adsorption of contaminants was possible because of the cristobalite and diatom structures of the membranes. The cristobalite octahedra structure has a capacity for cation exchange, chemical stability, and it has a relatively minimal expansion coefficient enhancing contaminant adsorption [13]. The same structure enhances contaminant adsorption through intra-particle diffusion. Apart from the lattice structures diatom structure have increased surface area, large adsorption capacities, and ion exchange abilities with the contaminants [11]. The firing of the membrane raptures the Si-O-Al bond leaving Al^{3+} to attract contaminant to its site. It was also noted that NaF had the lowest purification efficiency because of its ability to change the pH of water which affected its adsorption capabilities [14].

5. Conclusions

This work has shown that the strength of diatomaceous based membranes can be improved by plant-based organic binders. The most effective binder investigated in this work was *Basella alba*. This work has further confirmed that a high concentration of plant-based binder in ceramics reduces its mechanical strength. The strength of the membrane was found to have a positive correlation with the contaminant filtration efficiency.

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