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Optimization of the Viscosity of Wack (Cameroon) Clay Slurry for Coating of a Filtration Layer over a Tubular Ceramic Support Layer by the Slip Casting Technique for Domestic Water Treatment

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Abstract

The objective of this work is to optimize the viscosity of Wack clay slurry for coating of a filtration layer over a tubular ceramic membrane support by the slip casting technique for application in domestic water treatment. Coating of a slurry over tubular ceramic membrane support by the slip casting technique requires a clay slurry of appropriate viscosity for it to adhere to the ceramic support by capillary suction. The clay slurry is elaborated using a mixture experimental design with constraint used to determine the optimum viscosity of the slurry, the proportions of raw materials in the slurry and the level of influence and interactions between the proportions of raw material on the viscosity of the slurry. The experimental domain used for the raw materials was: proportion of clay between 10% and 12%, proportion of binder between 11% and 22% and proportion of water between 66% and 79%. It resulted that the proportion of clay has the highest positive influence on the viscosity of the slurry and the interaction proportion of clay-proportion of binder influences positively the viscosity of the slurry. The optimal viscosity was found to be 40.4 mPa·s for optimal proportions of clay, binder and water/dispersant of 10%, 11% and 79% respectively. The prepared optimal slurry was then cast over the support for contact times of 2, 4 and 6 minutes in order to determine the effect time on the compromise homogeneity (no cracks) and thickness (5 - 15 µm) of the deposited layer. When each contact time was attended, it was dried, sintered at 900˚C and characterized. It resulted that as the contact time increases, there is an increase in the thickness of the deposited layer from 8.8 to 23 μm with the gradual appearance of cracks from 4 minutes. The tubular membrane coated for 2 minutes gave the best results in terms of homogeneity and thickness of 8.8 μm. The porosity, permeability and average pore size this multilayer membrane were determined. It resulted from this that the membrane had a porosity of 32.6%, permeability of 305.67 L/h·m^2 bar and an average pore size of 0.48 μm adapted for microfiltration.

Keywords

Slip Casting, Filtration Layer, Mixture Design with Constraint, Contact Time, Viscosity, Optimization

1. Introduction

Ceramic membrane layers have been widely used in the treatment of domestic drinking water [1] [2]. These membranes are porous support layers made from inorganic materials such as clay and capable of eliminating suspended particles and even microorganisms from water [3]. These ceramic support layers however do not eliminate these pollutants totally from water (retention capacity generally less than 85%) [4]. Recent studies have shown that coating these ceramic support layers with another fine clay layer greatly ameliorates the elimination of these pollutants from water (retention capacity greater than 95%) [5]. The coated layer consists of a filtration layer over which filtration takes place first before going through the porous support layer. The support ceramic layer and the deposited filtration layer constitute a multilayer ceramic membrane used in the treatment and amelioration of the quality of domestic drinking water. Different techniques have been used to coat these ceramic support layers they include: spray coating, dip coating, sol-gel process, tape casting and slip casting techniques. Slip casting is the most appropriate technique used in coating slurry on a tubular ceramic support layers since it is easy to realize at a relatively lower cost compared to the other techniques [6]. Tubular membranes have the advantage over the other forms of membrane (disc, pot, cylindrical) in that they offer low fouling during filtration as it is mostly used in tangential filtration compared to the frontal filtration used by the other forms [7]. Previous studies have shown that coating a filtration layer over a tubular ceramic support by the slip casting technique is influenced by the viscosity of the clay slurry which depends on the proportion of raw materials used [8] [9]. Slurries that are too viscose or not viscose at all will not adhere properly on the ceramic support layer by capillary suction producing a deformed and cracked deposited filtration layer which would not retain the pollutants efficiently from water, hence low retention [6] [8] [10]. Authors have shown that the viscosity of the slurry increases with the proportion of clay and binder but tends to decrease with an increase in the proportion of water [5] [10]. These authors generally used the one variable analysis method to study the effects of these factors on the viscosity of the clay slurry. No experimental design was used by these authors, resulting in little information known on the effect and the interaction between the clay, binder and water on the viscosity of the slurry [11]. Hence, the needs to use an experimental mixture design with constraints to determine the effect and optimal proportions of each ingredient on the viscosity of the slurry. Mixture design with constraint is a better tool used for mixing the different proportions of raw materials rather than one variable mixing as it verifies how the properties of interest (viscosity in this case) are affected by the variation of these proportions in a mixture. It also permits to determine the interactions and the optimal proportions of the raw materials in the mixture [12]. Casting or coating of slurry of optimum viscosity onto the porous support is influenced by the parameter contact time slurry/support which determines the homogeneity and thickness of the deposited layer by capillary suction of the slurry onto the porous support [5]. Authors have shown that high contact times ≥ 6 mins lead to a high suction of the slurry on the support, but have as a consequence in that it leads to the saturation of the support giving a fragile (cracked) layer after sintering and to the formation of a thick and less porous layer which decreases the flow rate [8]. The best contact time is one which will give ideally a homogenous layer with thickness comprised between 5 - 15 µm, porosity between 30% - 35% and pore size between 0.1 - 2 µm after sintering to be adapted for microfiltration for the elimination of suspended particles and other pollutants [6] [10].

Considering such research trends, the objective of this work is to elaborate and coat the filtration layer (tubular multilayer) from Wack (Cameroon) clay by the slip casting technique. Specifically, to determine the optimal formulation of raw materials that gives the best viscosity of the slurry using a mixture design with constraint and to determine the best contact time to have the required homogeneity and thickness of the filtration layer.

2. Material and Methods

2.1. Composition of the Wack Clay Slurry for the Filtration Layer

A slurry is a stable suspension of mineral particles and is generally composed of mineral powder, solvent, the dispersant and organic additives. Indeed, organic additives such as binders, surfactants or thickeners make it possible to control the rheology of the slurry and to ensure the adhesion of the layer after evaporation of solvent during first stages of sintering and the creation of pores on the layer after thermal treatment. The slurry was composed of the following:

- Wack Clay (50 µm);
- The solvent (alkali water of pH 8.2 that acts as both a solvent and a dispersant);
- The binder (PVA prepared at 12%).

The combination of a well-defined percentage of the different elements making up the slurry, leads to a stable, not flocculated suspension that can be deposited on the support to form layers without defects (homogenous).

2.1.1. Preparation of the Wack Clay

The choice of the granulometry of the clay powder to be deposited will depend on final morphological characteristics desired for the membrane. Indeed, the principal parameter that should be well mastered and known is the size (granulometry) of the raw material. This parameter will influence in a direct way the final characteristics of the membrane after sintering, particularly the porosity and the diameter of the pores. The presence of heterogeneous particles size in the powder can lead to the reduction in the porosity and the homogeneity on the surface of the layer [8]. A monodisperse granulometric distribution is thus desirable for any raw material.

Clay mineral was collected at a depth of about 1.5 m from a mining site located in Wack village, in the Adamawa region, at 50 km SW of Ngaoundere, Cameroon at an altitude of 708 m, latitude 07˚40.685 N and longitude 013˚33.026 E. Generally, it is known as Wack clay. The Wack clay is predominantly composed of silica and alumina with a small amount of Fe_2O_3, CaO, K_2O, MgO and Na_2O. The percentage of Al_2O_3 corresponds to non-refractory clay since it does not exceed 45% [1]. The presence of a high level K_2O confirms that it can be used as a melting material in our formulation [1].

The Wack clay powder was crushed using a ceramic mortar and pistol and then sieved using a 50 μm sieve to obtain a homogenous clay powder. The final powder obtained from the sieved powder clay crushed above, has a granulometry which do not exceed the 50 μm. The percentage or proportion of clay used in the preparation of the slurry varies and it depends on the thickness of the deposited layer to be attained. To elaborate a microfiltration membrane of a few tenths of μm, the order of magnitude of mineral materials (clay) to be used is 10% [5]. In the case of this study we used a proportion of clay in the range 10% - 12 %.

- **The solvent:**

Water is generally used as the solvent. This is because water has a low viscosity and allows the solubilisation of the organic additives. The water used in the preparation of the slurry is usually basic in the range of pH of 8 - 9, this in order to enhance repulsion between the powder particles and have a stable suspension. In our case we used water of pH 8.2 which was prepared by adjusting the pH with NaOH. The solvent here that acts as both a solvent and a dispersant.

- **Organic polymer (Binder):**

It can be used both as a dispersant and binder while contributing to modify or adjust viscosity. It protects the membrane from cracks which generally appear during the drying and at the beginning of re-sintered. In our case we used Polyvinyl Alcohol (PVA). The PVA has a role of binder but also of plasticizer since it forms a thin film very elastic after drying. PVA is a hydrosoluble polymer of structure—$(CH_2\text{-}CHOH)_n$, with degree of polymerisation of 2000 and degree of hydrolysation of 86 - 89 mol%. It was prepared at 12% concentration [10]. The PVA was supplied by the department of Applied Chemistry of the University of Ngaoundere.

2.1.2. Tubular Ceramic Membrane Support Layer

The tubular ceramic membrane support layer shown in **Figure 1** was elaborated by Yanu *et al.* in 2020 and consist of optimal size and amount of raw materials of 66% Wack clay, 30% red sawdust and 4% binder for particle size of 100 µm elaborated by the extrusion technique. The tubular ceramic membrane support layer has geometrical configuration such as outer and inner diameters and length of the tube were 4 cm, 2 cm, and 19 cm, respectively. The sintered membrane possessed a porosity of 43.5%, water permeability of 244.9 L/h·m^2 bar, an average pore size of 2.4 µm and mechanical strength of 9.2 MPa with very good corrosion resistance in acidic and basic conditions [1].

Figure 1. Tubular ceramic membrane support layer [1].

2.2. Mixture Design with Constraint (Extreme Vertex Design)

The synergetic effect of a combination of two or more components on a property of interest can be easily identified by means of a mixture design approach. In a mixture experimental design, the total amount is held constant and a measured property of the mixture changes when the proportions of the components of the mixture are changed. Therefore, the main purpose of using this methodology is to verify how the properties of interest are affected by the variation of the proportions of the mixture components [13]. The mixture experimental design generates a map of the response over a specified region of formulation. It is possible to discover the critical variables, to define mathematical models and by them, to optimize the process [14].

Recall that the purpose of this work is to study the effect of three components, namely proportion of clay, proportion of binder and proportion of water on the viscosity of the clay slurry (suspensions). A mixture design with constraints (Extreme vertex designs) is used when there is a constraint on both the lower and upper bound of the proportions of raw materials present, or when linear constraints are added to several components. **Table 1** below gives the experimental domain of the components (raw materials) with their lower and higher bounds obtained from literature [6] [10]. The constraint in our mixture design was at the level of the proportions of clay and binder which has to be maintained between 10% and 12% and between 11% and 22% respectively. This is because literature shows that proportions lower that the above values give thin and cracked layers while higher proportions values give thick non porous layers when the slurry is deposited on the ceramic support layer [5] [10].

In the case of three components, the factorial space constituted by all the possible fractions of the components is a triangle whose vertices correspond to pure components. In this design, the constraints define the area of the factor space that can be used as shown in **Figure 2**. The relationship between the viscosity of the slurry and the components of the mixture can be represented using a polynomial mathematical model, as the sum of the components $\sum X_j = X_1 + X_2 + \cdots + X_q$ must be 1 or equivalent to 100%. For three components, the Scheffe's canonical special cubic model takes the following form [15]:

$$Yi\ \left(cal\right) = b_1X_1 + b_2X_2 + b_3X_3 + b_{12}X_1X_2 + b_{13}X_1X_3 + b_{23}X_2X_3 + b_{123}X_1X_2X_3 \quad (1)$$

where $Yi\,(cal)$ is the calculated response value at the ith experiment, bi are the model coefficients, X_1, X_2 and X_3 represent the proportions of raw materials (wack clay, binder (PVA) and water) respectively. In the present research, a mixture design with 13 experiments was generated using the software Minitab version 2017 and Statgraphics plus software. The statistical experimental design as specified by the software as shown in **Table 2**.

Table 1. Experimental domain for the raw materials.

Components	Variables	Levels/Variations	
		Lower (−1)	Higher (+1)
Clay (%)	X_1	10	12
Binder (PVA) (%)	X_2	11	22
Water (%)	X_3	66	79

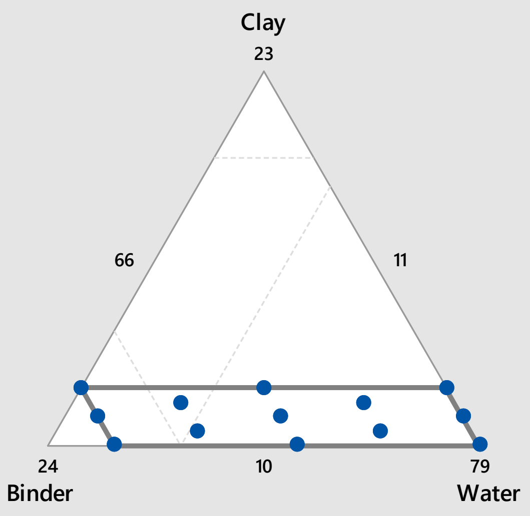

Figure 2. Distribution of the experimental points in the studied domain.

Thus a mixture design with constraint with 3 factors gives a total number of experiments of 13 experiments. The total number of experience was determined using the equation below:

$$N = \left(2^p + 2\right) + No \quad (2)$$

where, N = Number of experiments, p = Number of factors and No = Number of centre points.

Table 2. Experimental matrix with the 13 formulations of the clay slurry.

S/N	Real Values			Viscosity (Vep) mPa·s
	Clay (%)	Binder (%)	Water (%)	
1	11.0	22.0	67.0	
2	11.5	13.75	74.75	
3	12.0	11.0	77.0	
4	10.0	11.0	79.00	
5	12.0	16.50	71.5	
6	11.5	19.25	69.25	
7	10.5	13.75	75.75	
8	12.0	22.0	66.0	
9	11.0	11.00	78.00	
10	10.5	13.75	75.75	
11	10.0	22.00	68.0	
12	11.0	16.50	72.5	
13	10.5	13.75	75.75	

Validation of the Models

The empirical model obtained is validated by comparing the values of the answers envisaged and those observed. This will make it possible to predict the answers in the field defined for the study.

- **Linear regression coefficient (R^2)**

The analysis of the variance makes it possible to calculate very useful statistics: R^2 or R square. It indicates how well the model explains the variability of the response variable. A higher R^2 value suggests a better fit, meaning the model can effectively capture the relationship in the data. These statistics are the ratio of the sum of squares of the calculated answers (corrected average) to the sum of squares of the measured answers (corrected average):

$$R^2 = \frac{\text{corrected average sum of squares of the calculated answers}}{\text{corrected average sum of squares of the measured answers}} \tag{3}$$

For the model to be valid, the R^2 value has to be >95%. In addition to the linear regression coefficient (R^2), other mathematical procedures and tools were used. The Absolute Average Deviation (*AAD*) and the Bias factor (Bf).

- **Absolute Average Deviation (*AAD*)**

The absolute average deviation quantifies the average magnitude or errors in the model's predictions, providing insights into the precision of the model. By analysing the *AAD*, we can determine how closely the model's predictions, which is essential for validating the model's usefulness in real-world scenarios. The absolute average deviation was determined using the formula below. For the model to be valid, the *AAD* has to be comprised between $0 \leq AAD \leq 0.2$.

$$AAD = \frac{\sum_{i=1}^{p}\left(\frac{\left|Y_{iexp} - Y_{ical}\right|}{Y_{iexp}}\right)}{p} \tag{4}$$

With: Y_{iexp} = experimental answer, Y_{ical} = calculated response starting from the model for an experiment I, and p = total number of experiments.

- **Bias factor (*Bf*)**

The Bias factor measures the systematic error of the model's prediction. A bias factor close to 1 indicates that the model's predictions are unbiased and generally accurate. Assessing the bias helps identify whether the model consistently overestimates or underestimates the response, which is crucial for practical applications. The Bias factor was determined using the formula below. For the model to be valid, the Bias factor has to be ≤1.2.

$$Bf = 10^{B} \tag{5}$$

The Bias (B) is given by the relation:

$$B = \frac{1}{n}\sum\log\left(\frac{Y_{theo}}{Y_{obs}}\right) \tag{6}$$

MINITAB version 2017 and Statgraphics plus software were used to analyse the data obtained.

2.3. Preparation of the Slurry

From the component design mixture given in Table 2 above we have 13 experiments (mixtures) with 3 components. In general, rheological behaviour of clay suspensions or slurry depends on many characteristics such as proportions and nature of clay, binder and pH of solution. For this reason and in order to understand the effect of proportion of clay, binder and water on the rheological behaviour of the slurry we have worked in constant conditions (pH, nature and experiment conditions). The protocol for the preparation of the slurry is as Figure 3:

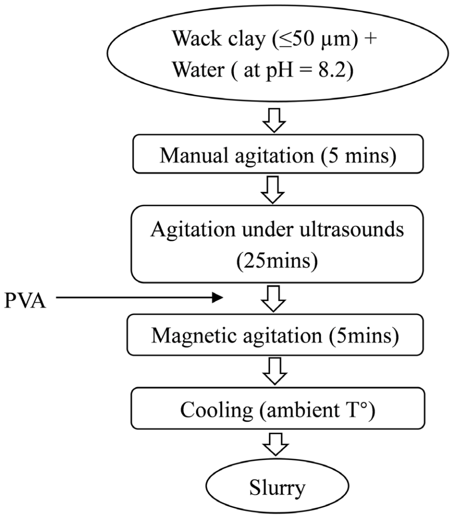

Figure 3. Process diagram for the preparation of Wack clay slurry [5].

- The 50 μm sieved wack clay powder is mixed first with an aqueous solution of the water at a constant pH of 8.2 and manually agitated for 5 minutes.

- The desagglomeration of the clay powder is done through ultrasounds agitation for 25 minutes at a power of 150 W.
- The binder (PVA) is then added and homogenized by magnetic agitation for 5 mins.
- It is then cooled under tap water and the viscosity of the slurry is determined.

Measure of the Viscosity of the Clay Slurry

The viscosity of the slurry should be sufficient such that the capillary adsorption of the slurry on the support should allow a uniform flow and coating without any infiltration into the depths of the support. Indeed, the viscosity of the suspension must be sufficient to prevent the fast formation of a thick and inhomogeneous layer related to a very fast absorption of solvent by the support. This viscosity must also make it possible for the solution to be run inside the support without difficulties. Generally, the viscosity and contact time slurry/support determines the thickness and homogeneity of the deposited layer [8].

The dynamic viscosity was measured under the same conditions for all samples using the same cycle with a DV III Ultra Rheometer. The rheological test consists of measuring the shear stress, shear rate and viscosity of the clay slurry at the same turn per minutes (tpm) of 10, 30, 60, 100, 150, 200 and 250 and at ambient temperature (25˚ ± 1˚) and viscosity values, shear rate and shear stress are retained.

The model of Herschel-Bulkley is the model that is mostly used to describe the behaviour of clay slurry. The viscosity of the prepared slurry has to follow the dynamics of the Herschel-Bulkley model with $n < 1$, for it to be adapted for coating on the support [16]. The reduced viscosity at higher shear ensures that the slurry can penetrate into fine details of the support structure, leading to a more consistent and even coating.

2.4. Coating Technique: Slip Casting of Clay Slurry over the Tubular Support Layer

The technique used for coating of the clay is called slip casting. It consists is putting the support in a vertical position held by a clamp and stand while blocking one end of the tube in order to fill the support with the optimal slurry (suspension) obtained from the mixture design with constraint for a contact of time 2 minutes, 4 minutes and 6 minutes. At the end of each contact time, the support is emptied and then dried at ambient temperature for 24 h. It should be noted that when we carry out slip casting, the pores of the macro-porous membrane exert a capillary suction force which enables the powder particles to be deposited on the support. At the same time, water infiltrates itself into the pores of the membrane. When these pores are filled with water, there is no longer a force to hold the solid particles deposited on the porous support forming a layer with cracks which is not homogeneous. The thickness of the layer is controlled by the contact time slurry-support, the characteristics of the support (pore diameter and porosity) and that of the deposited slurry (concentration and viscosity).

2.4.1. Drying and Thermal Treatment

After draining, the membrane is dried at ambient temperature vertically in order to allow the total evacuation of the extra slip remaining in the support. The qualities of the layers are modified by a premature rise in the temperature. In our case, drying is done at ambient temperature during 24 hours. The thermal treatment which is an operation that allows the elimination of organic products and the sintering of the membrane ensures the consolidation of the layer, its adherence with the support and gives its final characteristics. It is the most important stage in the elaboration of the membranes that should be well mastered. The temperature platform and the speed of heating must allow the evolution of organic matter, then the diffusion of the matter between the grains thus supporting the consolidation. By taking into account the presence of water and organic materials in the slurry, we established an adequate program for sintering allowing us to obtain a membrane without defect. The platform at 250˚C corresponds to the departure of PVA. The program used is as follows:

The final temperature of sintering (T_f) and the final time of sintering (t_f) are very important parameters when sintering the membrane for microfiltration. Indeed, it is general judicious to choose temperatures approximately 50˚C at 100˚C lower than the sintering temperature of the support layer, in order not to densifier the layer [5]. In our case T_f was at 900˚C for a support layer membrane sintered at 950˚C as shown below.

$$25^{\circ}\mathrm{C}\xrightarrow{1^{\circ}\mathrm{C/min}}25^{\circ}\mathrm{C}/1\mathrm{h}\xrightarrow{2^{\circ}\mathrm{C/min}}\mathrm{T_f}/2\mathrm{h}$$

Figure 4 below shows the stages of the casting process of the clay slurry over the tubular support by slip casting and **Figure 5** shows the imagine of the slurry in the tubular support layer and when emptied and drying.

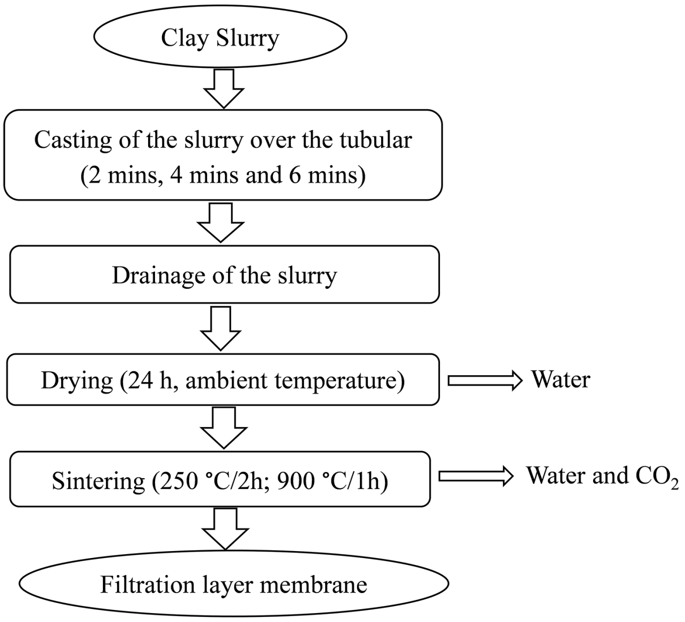

Figure 4. Classic technique for depositing a suspension by slip casting.

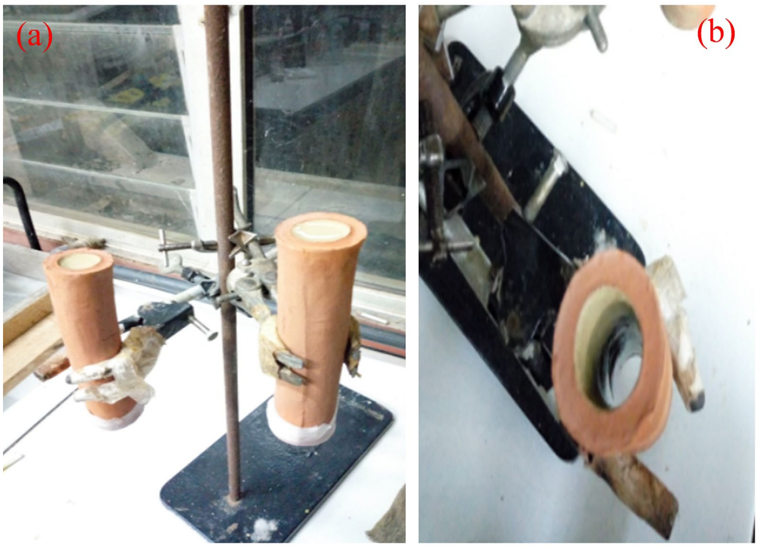

Figure 5. Casting of the slurry on a membrane for microfiltration: (a) Support with slurry and (b) Support coated with slurry.

2.4.2. Determination of the Best Contact Time Relating to the Thickness and Homogeneity of the Deposited Filtration Layer

The optimal proportion of the slurry obtained from the mixture design with constraint was cast into the tubular support for contact times of 2 minutes, 4 minutes and 6 minutes. The support was dried, sintered and then analysed using a scanning electron microscope (SEM Hitachi S-4500) in order to study the influence of contact time slurry/support and to determine the best optimal contact time with respect to the homogeneity and thickness of the deposited filtration layer. The contact time that gives a homogeneous layer with no cracks and a thickness of layer in the range 5 - 20 μm will be considered as the best contact time.

2.4.3. Determination of the Level of Influence of the Proportion of Clay

Proportions of clay of 10%, 11% and 12% was mixed with the optimal proportion of binder obtained from the mixture design with constraint and the corresponding proportion of water. The three mixtures were then cast into three tubular supports using the best contact time obtained from above. After drying and sintering of the tubular membrane, it was then analysed using a scanning electron microscope (SEM Hitachi S-4500) in order to study the level of influence of the proportion of clay on the thickness of the deposited filtration layer.

2.4.4. Determination of the Level of Influence of the Proportion of PVA

Proportions of binder of 11%, 16.5% and 22% was mixed with the optimal proportion of clay obtained from the mixture design with constraint and the corresponding proportion of water. The three mixtures were then cast into three tubular supports using the optimal contact time obtained from above. After drying and sintering of the tubular membrane, it was then analysed using a scanning electron microscope (SEM Hitachi S-4500) in order to study the level of influence of the proportion of PVA on the thickness of the deposited filtration layer.

2.5. Characterisation of the Tubular Multilayer Membrane Properties

The deposited layer was then characterized by determining some intrinsic properties of the tubular multilayer membrane. These properties are given below:

2.5.1. Morphology and Thickness

Scanning Electronic microscope (SEM) gives information of the surface and morphology of a massive sample. The scanning electronic microscope used in this work is a microscope at variable pressure (Hitachi S-4500). The analysed sampled was segment of the tubular multilayer membrane (surface and depth) which was snapped by the scanning electronic microscope which is equipped with a computer that has a software that treats the data in terms of sharpness and enlargement of the image that appears on the screen.

2.5.2. Permeability

The water permeability (L_h) of the membrane was determined by distilled water flux at different applied pressures. It was evaluated from the slope of the pure water flux (J_W) versus applied pressure across the membrane (ΔP).

$$Jw = L_h \Delta P \tag{7}$$

2.5.3. Porosity

The porosity of the tubular multilayer membrane is measured using the below expression [1] as described in the previous chapter:

$$Porosity(\%) = \frac{(W_1 - W_0)}{W_1} \times 100 \tag{8}$$

It was done using the Archimedes method whereby the dry tubular multilayer filter is weighed (W_0) and put into water for about 4 h and then reweighed (W_1).

2.5.4. Average Pore Size

Hagen-Poiseuille equation was employed to estimate the average pore size of the tubular multilayer membrane by assuming pores are cylindrical in shape [1] [17] as described in the previous chapter.

$$r = \sqrt{\left[\frac{L_h 8\mu\tau l}{\varepsilon}\right]} \tag{9}$$

where, ε is the porosity of the membranes, r is the pore radius of the membrane (m), l is the pore length which is generally taken as thickness of the membrane, τ is the tortuosity factor (generally used as 1), and μ is the viscosity of water (0.00089 Pa·s).

3. Results and Discussion

3.1. Optimization of the Viscosity of the Clay Slurry

Thirteen mixtures of clay slurry were prepared according to the experimental conditions. The measured and calculated values for the viscosity of the slurry are given in Table 3 below. Table 3 below gives the values of the viscosities of the slurry in the experimental matrix which varies between 32.2 - 88.6 mPa·s.

Table 3. Experimental design for the viscosity of the clay slurry.

S/N	Real Values			Viscosity exp (Vep) mPa·s	Viscosity cal (Vcal) mPa·s
	Clay (%)	PVA (%)	Water (%)		
1	11.0	22.0	67.0	66.5	70.04
2	11.5	13.75	74.75	43.4	41.12
3	12.0	11.0	77.0	32.3	34.79
4	10.0	11.0	79.00	38.2	40.34
5	12.0	16.50	71.5	56.0	55.76
6	11.5	19.25	69.25	62.0	62.85
7	10.5	13.75	75.75	50.8	42.27
8	12.0	22.0	66.0	88.6	86.49
9	11.0	11.00	78.00	36.8	34.98
10	10.0	13.75	75.75	40.7	42.65
11	10.0	22.00	68.0	61.3	58.14
12	11.0	16.50	72.5	45.2	46.78
13	10.5	13.75	75.75	43.5	39.02

3.2. ANOVA and Validation of the Model

The good quality of the fitted model was attested with analysis of the variance (ANOVA) as shown in Table 4. Indeed, this table shows that the sum of squares related to the regression was statistically significant when using the F-test at a 99.9% probability level, which suggests that the variation accounted by the model was significantly greater than the residual variation.

Table 4. ANOVA for the clay slurry.

Source	Sum of squares	Degree of freedom	Mean square	Rapport F	Proba.
Special-cubic	2799.07	6	466.51	37.04	0.0002**
Residual (total error)	75.5712	6	12.59		
Total (corr.)	2874.64	12			

**Significant at the level 99.9%.

- **Validation of the model**

Table 5 below gives the criterions of evaluation and validation of the model. The analyses of variance give an R^2 value of 97.37% which is greater than 95% meaning the model explains 97.37% the variability of the viscosity of the clay slurry. The Bias factor is less than 1 implying that the model error (uncertainty of prediction) is less than a unit. The AAD equal to 0.003 implying that there is no great variability in the data (the data is not spread out) and since the AAD is small it implies that the average value is indicative of the other values within the data set thus the data in the set are close together. With all these criterions respecting the reference values, it can be concluded that the proposed model is valid (accepted) and will explain the variation in the viscosity of the clay slurry at a confident level 95%.

Table 5. Criterion of validation of the model.

Validation indicator	Viscosity slurry	Reference value
R^2	97.37%	>95%
R^2 adjusted	94.74%	>90%
AAD	0.003	0 ≤ AAD ≤ 0.2
Bias factor	0.99989	≤1.2

3.2.1. Experimental and Statistical Modeling

It consists of a group of techniques used in establishing empirical study of the relationship between a response and several input variables. Interpretation starts with the calculation of the coefficients of the model. The coefficient of the model is represented in the Table 6 below. The equation of the special cubic model for the viscosity of the slurry is given below:

$$Y = 206.39X_1 + 23.27X_2 + 6.89X_3 + 3.13X_1X_2 - 2.90X_1X_3 - 0.50X_2X_3 + 0.03X_1X_2X_3$$

where Y = viscosity of clay slurry; X_1 = proportion of clay; X_2 = proportion of binder; X_3 = proportion of water.

Table 6. Coefficient of the model.

Terms	Coefficient
X_1	+206.39
X_2	+23.27
X_3	+6.89
X_1X_2	+3.13
X_1X_3	−2.90
X_2X_3	−0.50
$X_1X_2X_3$	+0.03

From the table above, all of the coefficients of the principal parameters that is proportions of clay, binder and water have a positive effect on the viscosity of the slurry (increases the viscosity of the slurry). But the proportion of clay has the highest positive effect on the viscosity. This can be explained by the fact that the clay particles in the medium hinder the flow of liquid limiting their freedom of movement hence resulting in an increase in the viscosity of slurry. For the interactions, only the interaction proportion of clay-proportion of binder has a positive effect on the viscosity of the slurry. The presence of binder makes the concentrated clay particles in the medium to stick together due to its gluing and gelling effect which forms miscelles with the particles through the formation of hydrogen bonds, increasing in the viscosity of the clay slurry. The third order interaction between the proportions of clay, binder and water influences positive the viscosity of the slurry but it is significantly low. Since the proportion of the clay has the highest individual effect and its interactions are low, it implies that the proportion of the clay is the component to follow as it influences significantly the viscosity. This is better seen and explained using the cox's diagram below.

3.2.2. Cox's Diagram (Contribution of the Various Factors)

Figure 6 shows the response trace of the viscosity of the clay slurry using a reference mixture S the centroïd of the simplex domain ($X_1 = 14.33$, $X_2 = 15.33$, $X_3 = 70.33$). In this figure, the vertical axis is the predicted response and the horizontal axis is the incremental change in each component. The reference mixture is shown as the point 0.00 on the horizontal axis. Each component is represented by one curve.

From the diagram below it is observed that the blue curve for the effect of the proportion of clay changes significantly, this means it has a significant effect (increases) along its axial. This is explained by the fact that the clay particles are closely packed and their interaction hinders the freedom of movement of the fluid in the medium hence increasing the viscosity of clay slurry [15]. Meanwhile the red curve for the effect of the proportion of binder is almost flat; this means when the proportion of binder changes and other components kept fixed, the viscosity does not change very much. The pink curve for the effect of proportion of water is between component clay and binder and decreases along the axis. This is due to the dilution effect of water which breaks the links between the clay layers enhancing repulsion between them thus decreasing the viscosity of slurry [18].

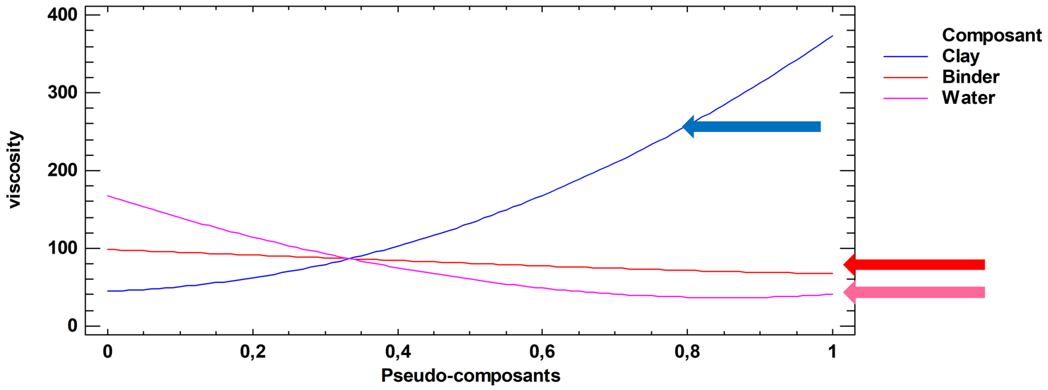

Figure 6. Cox's Diagram using a reference mixture S domain ($X_1 = 14.33$, $X_2 = 15.33$, $X_3 = 70.33$).

3.2.3. Iso-Response Curve

Following the validation of the model, it was used to generate response surface as a contour plot or a three dimensional surface plot over the explored domain as shown in Figure 7. The shaded area in red shows the optimal zone in the domain of study for the viscosity of the clay slurry. In order to evaluate the contribution of each of the three components and to determine the optimal values of each, the response trace technique is used [19]. In practice, the response trace is a plot of the estimated response value as we move away from a reference mixture and along the component axes.

Recall that in ceramic industry, clay slurry used for coating support must have a low viscosity in the range 25 - 100 mPa·s in order to facilitate the adhesion of the slurry on the support by capillary suction [5] [8] [10]. The contour plot (iso-response diagram) in **Figure 8** indicates that formulations which correspond to this requirement (viscosity < 100 mPa·s) are mixtures rich in water and binder with a small amount of clay. The optimal zone from the domain of study corresponds to the area with viscosity between 32.2 - 88.6 mPa·s (shaded portion in red). The optimal zone falls within the experimental domain midpoint between the proportion of binder and water and a decrease in the proportion of clay.

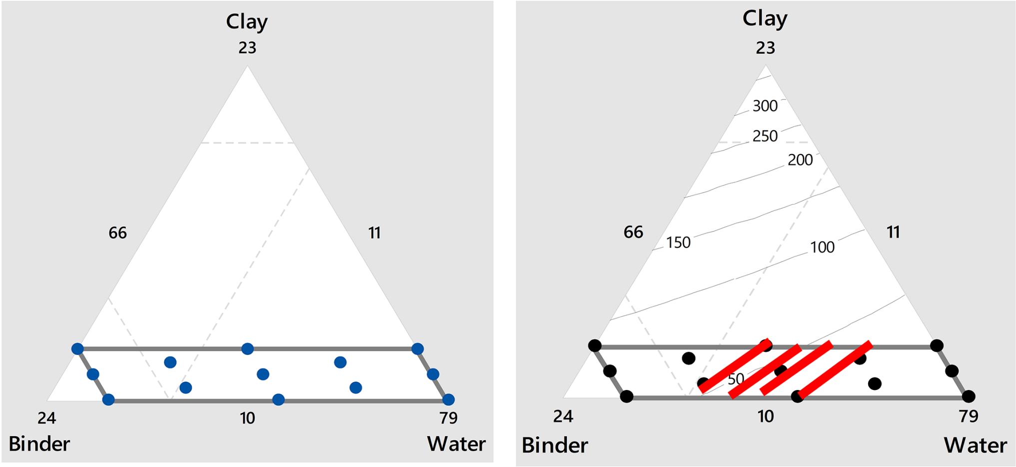

Figure 7. Experimental domain and the Iso-response diagram.

Table 7 gives the optimal values of the proportions of clay, binder and water for the clay slurry. It is a formulation with low values of clay and binder but higher values of water. The optimal viscosity of 40.4 mPa·s is obtained with lower proportions of clay and binder and a higher proportion of water. This is because high proportions of clay and binder lead to an increase in the viscosity of slurry which is not needed for the coating of the slurry over the support leading to thick and fragile layers. The optimal viscosity of the slurry is found within this viscosity range making it good for adhesion over the support by capillary suction.

Table 7. Optimal proportion of raw materials for the clay slurry.

Proportions (%)	Optimum
Clay	10
PVA	11
Water	79
Viscosity (mPa·s)	40.4

3.2.4. Flow Index of the Optimal Clay Slurry

The Herschel-Bulkley model was used to describe the rheological behaviour of the optimal clay slurry. The model is given by the equation below where n is the flow index.

$$\tau = \tau_o + k\acute{y}^n \tag{10}$$

$$n = \log\frac{\tau - \tau_o}{k} - \log y \tag{11}$$

where k is the consistence of the fluid, τ is shear stress and $\acute{y}$ is the shear stress, n the flow index ($n < 1$ the fluid is shear thinning; $n > 1$ the fluid is shear thickening).

Flow index of the optimal slurry (n) was 0.79 ($n < 1$) which is characteristic of a pseudoplastic fluids thus stable during the casting period. The flow curve of the optimal clay slurry is shown in **Figure 8** below. From the curve it is observe that as the shear rate increases, the viscosity decreases. This can be explained by the fact that the particle-particle interactions in the slurry are broken down causing a random movement of particles, reducing fluid flow and thus the viscosity of the slurry. This brings about a non-Newtonian, shear thinning behaviour such that the viscosity decreases when shear is increased. The shear-thinning behaviour of slurries enhances their flow characteristics, improves the coating process, reduces defects and allows for more efficient production [16].

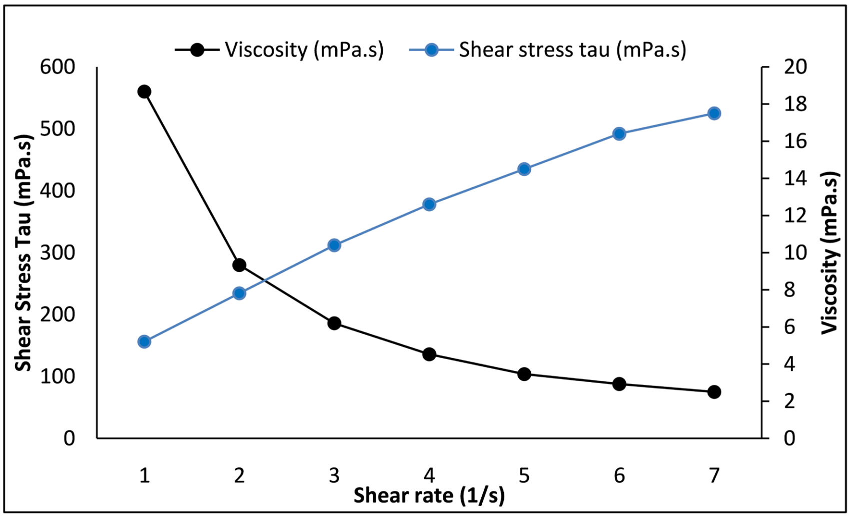

Figure 8. Rheogram of the optimal clay slurry.

3.3. Influence of Contact Time on the Properties of the Filtration Layer (Homogeneity and Thickness of Layer)

Figure 9 below shows the variation of the homogeneity and thickness of the deposited layer with respect to contact time. Looking at the surface view for the evolution of the homogeneity of the layer with respect with time, it is observed from the **Figure 10** below that as the contact time increases from 2 minutes to 6 minutes there is the gradual appearance of cracks. Meanwhile, looking at the cross-section view for the evolution of the thickness of the layers, it is observed that as the contact time increases from 2 minutes to 6 minutes, the thickness of the deposited layer also increases from 8.8 µm to 23 µm. The gradual appearance of cracks and the increase in the thickness of the deposited layer with time can be explained by the fact that as the contact time increases from 2 to 6 minutes, there is an increases in capillary suction of the slurry onto the support which leads to the formation of a heterogeneous compressible deposit which becomes thicker but fragile with time (appearance of cracks) due to saturation of the pores. With extended contact time, particles in the slurry have more time to rearrange and pack more efficiently. This leads to a reduction in the overall porosity and permeability as the particles settle more tightly together [3] [6] [10]. With the specifications set on the properties of the deposited layer for it to be homogenous (no cracks) and have a thickness comprised between 5 - 15 µm, the contact time of 2 minutes was chosen as the best as it gave homogenous layer with thickness of 8.8 µm.

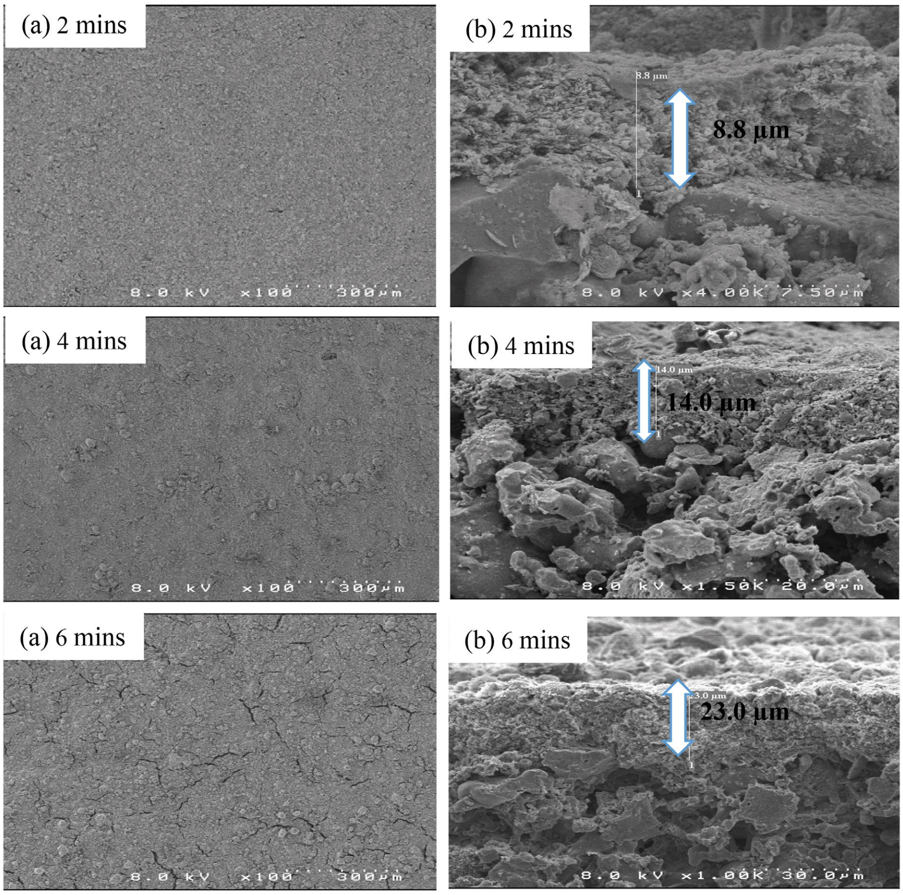

Figure 9. (a) Surface and (b) cross-section view for the evolution of the homogeneity and thickness of deposited layer with respect to contact time (clay 10%; binder 11%; water/dispersant 79%).

3.3.1. Determination of the Level of Influence of the Proportion of Clay on the Thickness of the Deposited Layer

Figure 10 below shows the variation of the thickness of the deposited layer with respect to the proportion of clay. The mineral content in the slurry (clay in our case), influences the thickness of the deposited layer at fixed contact time (best). It was noted during this work that at a fixed contact time of 2 minutes and using the optimal proportion of binder and water of 11% and 79% respectively, the thickness of the deposited layer support increases from 8.8 µm to 29.7 µm with an increase in the proportion of clay from 10% to 12%. This can be explained by the fact that as the proportion of clay increases, there is an increase in sedimentation and accumulation of the clay on the surface of the support by capillary suction of the clay particles in the slurry.

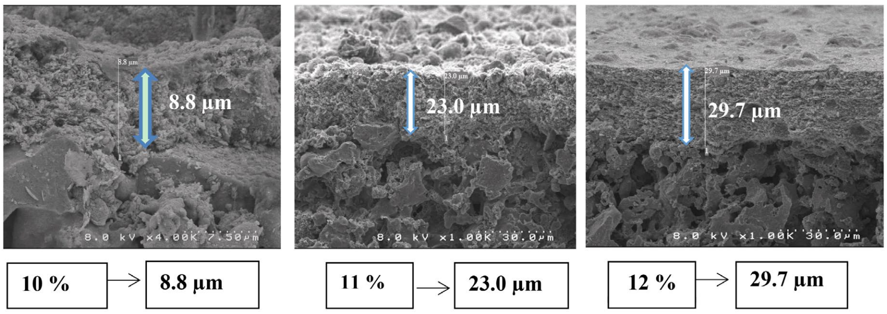

Figure 10. Evolution of the thickness of the filtration layer with respect to the proportion of clay (Clay 10%; 11%; 12%, binder 11%, water 79% at contact time 2 minutes).

3.3.2. Determine of the Level of Influence of the Proportion of Binder on the Thickness of the Deposited Layer

Figure 11 below shows the variation of the thickness of the deposited layer with respect to the proportion of binder. The proportion of binder also influences the thickness of the deposited layer for a fixed contact time of 2 minutes, optimal proportion of clay of 10% and proportion of water of 79%. It was observed here that the thickness of the deposited layer increases from 8.8 µm to 35.0 µm as the proportion of the binder increases from 11% to 22%. This is explained by the fact that as the proportion of binder increases, it leads to an increase in the formation of the clay aggregate due to the gelling and gluing effect of the binder. This causes the clay particles to settle on the surface of the support thus increasing the thickness of the layer.

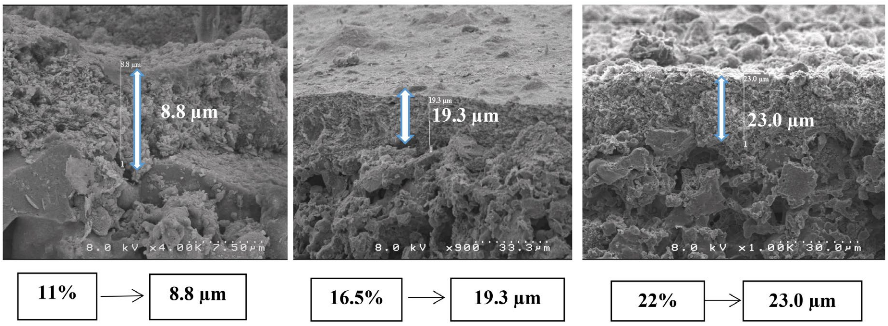

Figure 11. Evolution of the thickness of the deposited layer with respect to the proportion of Binder (PVA 11%; 16.5%; 22%. Contact time 2 minutes, Clay 10% and water 79%).

3.4. Characteristics of the Tubular Multilayer Membrane

3.4.1. Porosity

The porosity of the fired deposited filtration layer membrane was found to be 32.6%, which is in accordance with values reported in the literature for microfiltration membranes. The porosity of the tubular multilayer ceramic membrane is greater than 30% and this is due to disappearance of the binder as from 250˚C.

3.4.2. Permeability

Figure 12 illustrates the water flux of the fired deposited filtration layer membrane as a function of the different applied pressures. It can be noticed that the water flux increases linearly with an increase of applied pressures (0.5 - 3 bar). This stipulates that the variation in pressure is the barely driving force for permeation. For transportation operation exclusively by convection, the flow rate is proportionate to the pressure, and is in accordance with Darcy's law. The water permeability (L_h) of the tubular multilayer membrane was determined from the graph of water flux against different applied pressures. The gradient of this plot gives the permeability of the membrane. The water permeability (L_h) of the tubular multilayer membrane was found to be 305.67 L/h·m^{2}bar which is in the domain for microfiltration (greater than 300 L/h·m^2·bar).

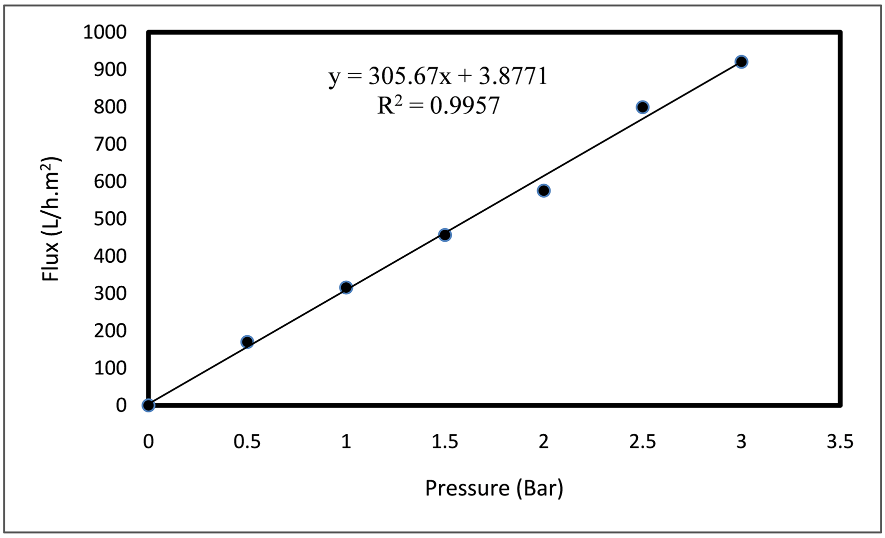

Figure 12. Determination of water permeability of the tubular multilayer membrane.

3.4.3. Average Pore Size

The average pore size of the fired tubular multilayer membrane was calculated to be 0.48 μm (0.48 × 10^{-6} m). The multilayer membrane is macroporous and adapted for microfiltration. Similar results were obtained by [20].

3.4.4. Chemical Resistance

The corrosion resistance test was performed with respect to weight decrement of the membrane after keeping in harsh environments (acid and alkali). The weight decrement of the membrane is calculated to be 0.38% in acidic condition and

0.17% in alkali condition. In acid and alkali conditions, the membrane displays an excellent result in resisting corrosion. The loss of weight is <3% indicating the support is stable chemically. The obtained results are more commensurable in comparison with the cordierite membrane fabricated by [21].

4. Conclusion

The objective of this work was to elaborate and coat a filtration layer (tubular multilayer) from wack clay by the slip casting technique applied in water treatment. In order to do these, two objectives were set. The first objective was to determine the optimal formulation of raw materials (clay, binder and water) on the viscosity of the slurry using a mixture design with constraint. It resulted from this objective that the optimum formulation of the slurry is obtained at lower values of clay and binder (10% and 11%) but at higher values of water (79%) because high proportions of clay and binder lead to an increase in the viscosity of slurry which is not needed for the coating of the slurry over the support leading to thick and fragile layers. The interaction proportion of clay-proportion of binder has a positive effect on the viscosity of the slurry as the binder causes the concentrated clay particles within the medium to stick together through hydrogen bond links (its gelling and gluing effect) thus increasing the resistance of flow of the slurry. The optimal proportion of clay, binder and water was 10%, 11% and 79% respectively with optimal viscosity of the clay slurry was found to be 40.4 mPa·s. The second objective was to determine the best contact time to have the required homogeneity and thickness of the filtration layer. It resulted from this objective that contact times of 2 minutes give the best compromise relating to the homogeneity and the thickness of the filtration layer giving a homogenous layer with thickness of 8.8 µm. This is so because higher contact times lead to thick and fragile (appearance of cracks) layers. The tubular multilayer membrane filter sintered at 900˚C, gave a membrane of porosity of 32.6%, permeability of 305.67 L/h·m^{2}bar, average pore diameter of 0.48 µm which is chemically stable and is adapted for microfiltration.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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