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Taguchi design of experiments approach to the manufacture of one-step alumina microfilter/membrane supports by the centrifugal casting technique

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Abstract

The Taguchi design of experiments method was implemented for the optimization of the manufacture of sintered one-step alumina microfilter/membrane supports by the centrifugal casting technique for the first time. A 10 wt.% alumina aqueous slip containing Tiron (0.001 g/g alumina) as dispersant and PVA as binder were used. Acceleration (3 levels), slip volume (3 levels), binder content (3 levels) and pH (2 levels) were selected as controlling parameters (saturated L-9 array). The use of three different target functions has been discussed: (1) the product of top-layer surface porosity times the reciprocal of top-layer surface pore diameter; (2) the product of permeability times thickness; and (3) membrane curvature. It is deduced that the first target function is the most appropriate as far as the membrane characteristics of the sintered compact are concerned. Using this target function a distinct optimum configuration for the controlling parameter levels could be obtained.

Keywords: Shaping; Microstructure final; Al_2O_3; Membranes; Centrifugal casting

1. Introduction

Porous ceramic membranes generally have a composite structure, i.e. they consist of a porous support, intermediate porous layer(s) and the effective membrane layer. In many cases, it is possible to produce the (support/intermediate layers) body in one step. In other words, it is possible to produce a composite (support/intermediate layers) structure membrane with a continuous pore size profile (functionally graded) using a single ceramic processing step. This is of extreme industrial importance, as the production costs of ceramic membranes are generally high [1]. Production of functionally graded (FG) porous ceramic supports has gained emerging importance during the last decade. Different routes of particulate processing for the production of flat alumina FG microfilter/membrane supports (sedimentation, controlled slip-casting and centrifugal casting) have been compared by Falamaki and Veysizadeh [2]. The latter work showed that if a slip casting procedure were applied, the slip casting velocity would be of outmost importance. However, the same authors concluded that centrifugation may result in better FG-supports as far as surface characteristics (pore size and porosity) among other factors like total permeability are concerned. Darcovich and Cloutier [3] used a two-level factorial experimental design to optimize the slip-casting process parameters (pH, sintering temperature and polymer concentration) for different alumina powders. Till now no attempt to optimize the processing parameters for the centrifugal casting procedure for the production of FG-alumina supports has been reported in the open literature. Hong [4] modeled the microstructure of green compacts (and not the final sintered products) produced by centrifugation and investigated the effect of zeta-potential, slip solid concentration, acceleration, viscosity, initial particle size distribution, etc. Despite its industrial importance, optimization of the centrifugal procedure to obtain a proper final sintered product has not yet been reported.

The purpose of the present work is to realize such an optimization employing a Taguchi method of experimental design. To this end, a saturated array has been implemented. Saturated arrays suffer from the disadvantage of not handling factor interactions particularly well [5]. Actually, if factor interaction exists, the most accurate method to optimize the

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processing system is to perform experiments for all the influencing factors and their pertaining effective levels. Considering the complex process of sample preparation and characterization for the case of centrifugal casting method of membrane preparation, the use of saturated arrays outweighs the weakness of not using full factorial designs or unsaturated Taguchi arrays. This work considers three 3 level factors and one 2 level one. In arranging a Taguchi table, it is of extreme importance to know a priori 'the most influential factors (and their corresponding levels) in order to benefit the most out of the Taguchi design of experiments. The Taguchi method might result in appreciable results if the most influencing factors and their effective levels are known to some extent a priori. This selection was done based on our previous practical experiences in this area [2].

In the present paper we consider another subject of significant concern in analyzing the results obtained by implementing Taguchi tables. The definition of the target function for optimizing the centrifugal casting process of membrane fabrication is a rather complex item. Three different target functions have been defined and the results have been compared. To elucidate some important effects, a restricted number of additional one-factor-at-a-time experiments have been performed.

2. Experimental

The alumina used in this study was a (Zn-203, MARTOXID) powder with a purity of 99.3 wt.%. The particle size distribution is 95 wt.% < 3.5 μm, 85 wt.% < 2.5 μm, 45 wt.% < 1.0 μm, 30 wt.% < 0.5 μm and 20 wt.% < 0.3 μm.

Tiron ($C_6H_4O_8S_2Na_2{\cdot}H_2O$) from Acros was used as dispersant. The optimum amount of 0.001 g/g alumina was used according to our previous results [2]. Tiron was added to 10 wt.% alumina aqueous slurry and the whole was planetary milled for 20 min. Afterwards PVA as binder was added and the slurry was planetary milled again for 20 min.

The centrifuge experiments were performed using a specially designed PTFE holder with inner glass tube (I.D. = 20 mm) and removable flat bottom section. Centrifuge experiments were performed using different rotational velocities (0, 1000, 2000 and 3900 rpm) with a duration of 20 min. To minimize any green compact deformation after supernatant slurry drainage, the holder was centrifuged again for 10 min and with the initial velocity.

To inhibit micro-crack formation, the following drying procedure was implemented: Drying for 1 week at RT and ambient humidity followed with 24 h drying at 50 °C in an oven.

Sintering was performed at 1525 °C for a period of 1 h with a heating rate from RT to the sintering temperature of 10 °C min^{-1}. This sintering temperature was selected according to our previous experience on the effect of the sintering temperature on the characteristics of disk-shaped microfilter/membrane supports using different routes of particulate processing [2]. Lower sintering temperatures did not result in adequate mechanical strength of that side of the graded membrane support which consisted of larger particles (due to centrifugal segregation). This is due to the fact that the raw material used was high purity α-alumina (>99.3 wt.%) without sintering aids. Sintering of high purity α-alumina is strongly size dependent. It was observed that for sintering temperatures lower than 1525 °C, the side consisting of larger particles could be easily scratched, i.e., they did not undergo proper sintering.

A SIGMA Laborzentrifugen apparatus was used for the centrifugation experiments. SEM pictures were taken using a Stereo Scan 360-Leica (Cambridge Instruments). The nitrogen gas permeability behavior of the sintered samples was investigated using a specially designed patented system [6]. The experimental setup of the latter is based on ASTM F316 with an extra compressed air sealing facility. The top-surface characteristics (surface porosity and average surface pore diameter) of the sintered samples were evaluated through analysis of the pertaining SEM pictures considering at least 100 pore per picture. This was performed using the image analyzer software Oracle 2.

3. Results and discussion

The following factors and corresponding levels were considered throughout this study:

- Factor A: acceleration

  Levels: 1 ≡ 10, 2 ≡ 7898 and 3 ≡ 30003 m s^{-2} (in terms of rotational speed: Levels: 1 ≡ 0, 2 ≡ 2000 and 3 ≡ 3900 rpm).

  Factor A was chosen in order to investigate the effect of centrifugal acceleration on the product characteristics. Level 1 of this factor resembles the conventional sedimentation method of preparation of FG porous structures.
- Factor B: slip volume

  Levels: 1 ≡ 10, 2 ≡ 15 and 3 ≡ 20 cm^3.

  Knowledge about the effect of system volume may be of great importance in up-scaling the results for developing semi industrial processes.
- Factor C: binder content

  Levels: 1 ≡ 2, 2 ≡ 4 and 3 ≡ 6 wt.% of used PVA solution concentration.

  Changing the binder content may induce significant effects on the packing behavior of the centrifuged powder even if the increment is rather small (2 wt.).
- Factor D: slip pH

  Levels: 1 ≡ 4 and 2 ≡ 7.

  Factor D has a significant effect on the pore microstructure of the shaped green compact. Although low pH's generally result in partial agglomeration which reduce to some extent the pore gradient along the body, they might increase the total permeability.

  An L-9 array was selected upon downgrading one of the columns through dummy treatment. This was indispensable as the standard L-9 array considers merely three level factors. Table 1 shows the modified L-9 array implemented.

  Generally speaking, applying the Taguchi approach using an L-9 array is justifiable only when the experimental

Table 1
Taguchi table of experiment design

Experiment no.	A factor level	B factor level	C factor level	D factor level
1	1	1	1	1
2	1	2	2	2
3	1	3	3	2
4	2	1	3	2
5	2	2	1	2
6	2	3	2	1
7	3	1	2	2
8	3	2	3	1
9	3	3	1	2

procedure is relatively time consuming and, moreover, the characterization procedures are lengthy and expensive. Considering our case, the compact green forming and drying had been considerably sensitive and time consuming. Actually taking out the centrifuged compact disks out of the holder assembly after appropriate partial drying in order to obtain a crack-free green compact has been very tricky. Most of the experiment configurations had been repeated up to four times and the average result of two successful experiments were reported for each configuration. It should be noted that L-9 arrays have been successfully applied in materials engineering cases so far (see ref. [7]).

Table 2 summarizes the results of the Taguchi design of experiments on the different membrane characteristic parameters of the samples. Fig. 1a–d show the top surface of the sintered samples due to some of the experiments 1–8.

In order to obtain more information possible out of the Taguchi table avoiding complicated 'target functions', different ones were defined as follows: (1) the ratio (top surface porosity (ε_s)/top surface pore diameter $(d_{p,s})^{-1}$), (2) the product (permeability (P) x membrane thickness (t)), and (3) top surface curvature (as defined elsewhere [2]). It should be mentioned that for each Taguchi table, the pertaining target functions had been normalized.

- $\varepsilon_s/d_{p,s}$ as target function

As far as the membrane application is concerned, the top-surface should have a high surface porosity and, at the same time, small average surface-pore diameter. Higher surface porosity improves further surface coating characteristics for obtaining intermediate or ultrafilter membrane layers. Smaller top-surface pore diameters are needed for obtaining smaller microfilter pores or appropriate supports for depositing ultrafilter or intermediate layers for nanofilter manufacture.

A membrane support with a pore size gradient has the smallest pores in one of it faces. In producing a microfilter, we aim at producing these pores as small as possible (although a lower limit of 0.1 microns exist which was never reached in any of our experiments). Therefore we aim at increasing the value of $1/d_{p,s}$. At the same, the surface porosity (ε_s) should be as high as possible to reduce mass transfer resistance to flow. Hence, an optimum membrane support should have a maximal value of the product of $1/d_{p,s}$ and ε_s.

Fig. 2 shows the plot of the target function as a function of the corresponding experiment number in the Taguchi table. It should be noted that the top surface of samples due to experiment 1 were not at all acceptable due to a significantly high roughness. Therefore the target function for this experiment was deliberately assigned the value 0. A distinct maximum is observed for the experiment number 3. The percent influence of various parameters on the target function as a result of analysis of variance (ANOVA) is shown in Fig. 3. The binder content is observed to have a rather significant effect on the top-surface characteristics while the effect of the scale of process (volume, factor B) is minimal. The effect of the individual parameters at their corresponding levels is shown in Fig. 4a–d. According to these figures, the optimum configuration of factor levels should be $A_1B_3C_3D_2$. As a matter of chance, this configuration exists already in the selected Taguchi table (experiment 3). The predicted value of the target function for this composition considering a 90% confidence level is 4.06 ± 0.67. The actual experimental value is 4.29. As the optimum factor levels are already present in the Taguchi table, we might consider an insignificant interaction between factor levels near and at the optimum configuration and discuss the individual effect of factors shown in Fig. 4 (a–d).

Generally, increasing the acceleration has a negligible effect on the surface characteristics (Fig. 4a) up to a rotational speed of 2000 rpm. This is while a 3900 rpm rotational speed

Table 2
Measured experimental responses of the Taguchi table

Exp.#	Thickness (mm)	Surface porosity (%)	Surface average pore diameter (μm)	Knudsen permeability × 10^5 ($m^3\,s^{-1}\,Pa^{-2}\,m^{-2}$)	Total open porosity (%)	Curvature
1	1.20	–*	–**	2.820	50.66	0.00521
2	1.9	5.85	0.296	1.069	46.26	0.02895
3	2.62	10.79	0.175	0.745	46.46	0.02393
4	1.34	14.77	0.301	2.050	49.06	0.02630
5	1.97	12.67	0.544	1.057	48.44	0.02160
6	3.07	5.94	0.451	1.088	48.06	0.01281
7	1.24	3.54	0.341	2.280	49.15	0.03120
8	2.39	7.98	0.511	1.648	49.35	0.01690
9	2.56	6.52	1.00	2.367	51.20	0.01048

(*) and (**) due to a resultant very rough surface no value was assigned.

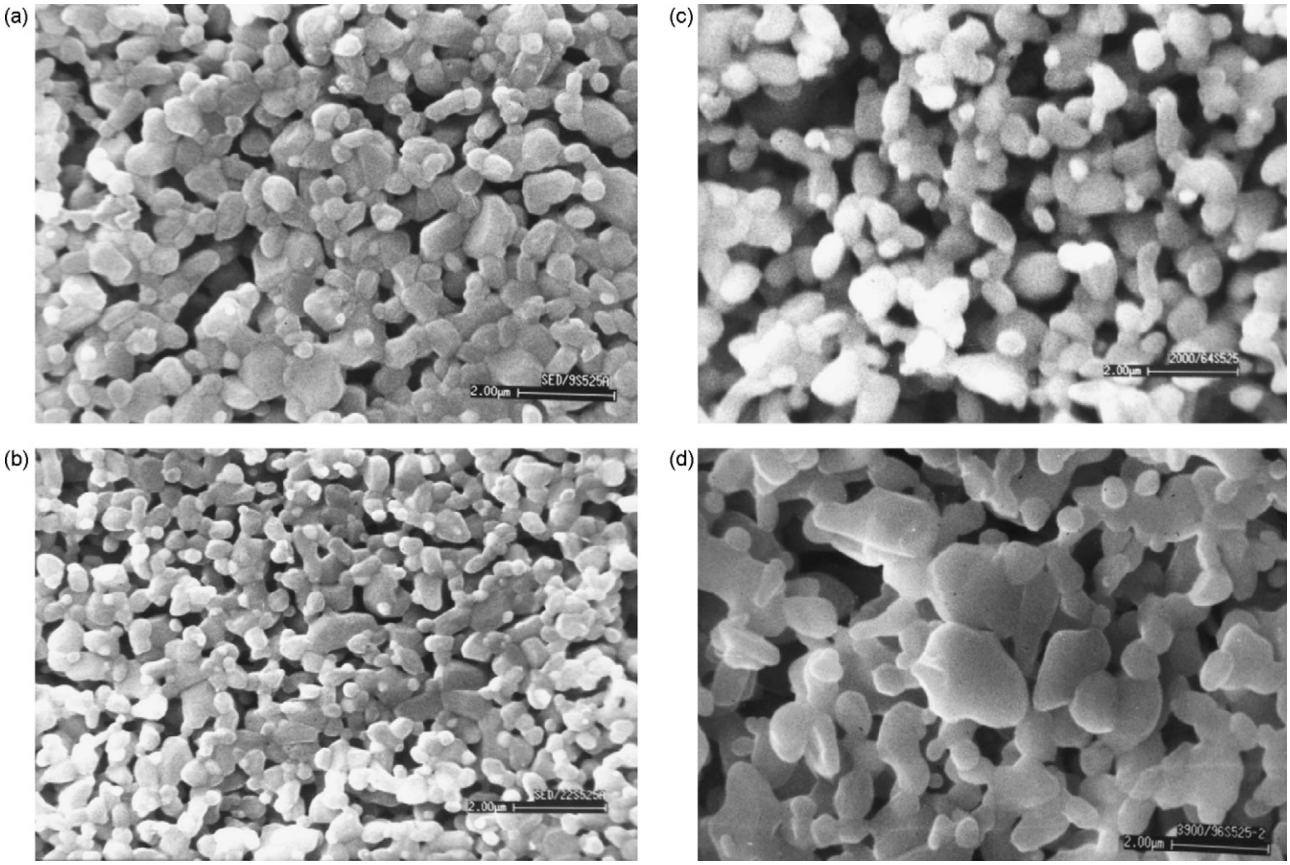

Fig. 1. SEM picture of the top surface of some of the samples of the Taguchi table (a) exp. #2 (b) exp. #3 (c) exp. #4 (d) exp. #8.

has a clear detrimental effect on the top surface characteristics. Two additional experiments with level configurations $A_1B_2C_1D_2$ and $A_3B_2C_1D_2$ were performed to explain more the effect of acceleration on the target function. The Taguchi table already contains the $A_2B_2C_1D_2$ configuration. The predicted and experimental results are shown in Fig. 5. The predicted target function for a configuration $A_wB_xC_yD_z$ is evaluated according to the following relation [8]:

$$X = \bar{Y} + (\bar{A}_w - \bar{Y}) + (\bar{B}_x - \bar{Y}) + (\bar{C}_y - \bar{Y}) + (\bar{D}_z - \bar{Y}) \qquad (1)$$

where X is the predicted target function, $\bar{Y}$ is the sum of the experimental target functions, $\bar{A}_w$ is the average effect of factor A at level w, $\bar{B}_x$ is the average effect of factor B at level x, $\bar{C}_y$ is the average effect of factor C at level y and $\bar{D}_z$ is the average effect of factor D at level z.

The trend of the curves is very similar and the error between the experimental and predicted target functions is smaller than the confidence interval calculated for the optimum. These results show insignificant interaction of the four factors as the Taguchi table results could reasonably predict the experimental ones.

The initial increase of the target function from A_1 to A_2 is mainly due to the increase in the magnitude of the top surface porosity (3.59–11.49%). This is attributed to the forced egress of the liquid through the tortuous green compact

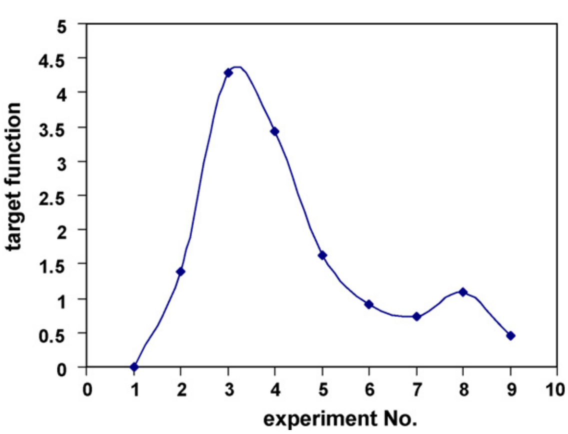

Fig. 2. Taguchi table experimental values for $\varepsilon_s/d_{p,s}$ as the target function.

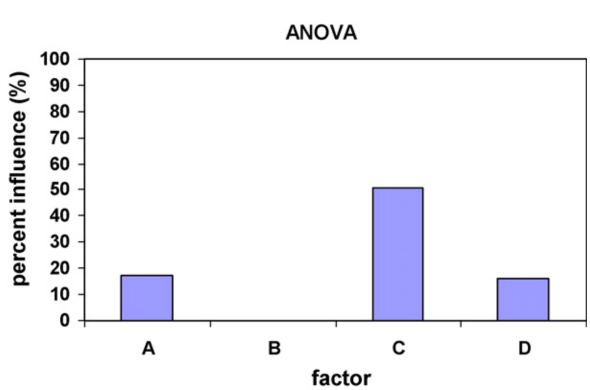

Fig. 3. Factor influence for $\varepsilon_s/d_{p,s}$ as the target function.

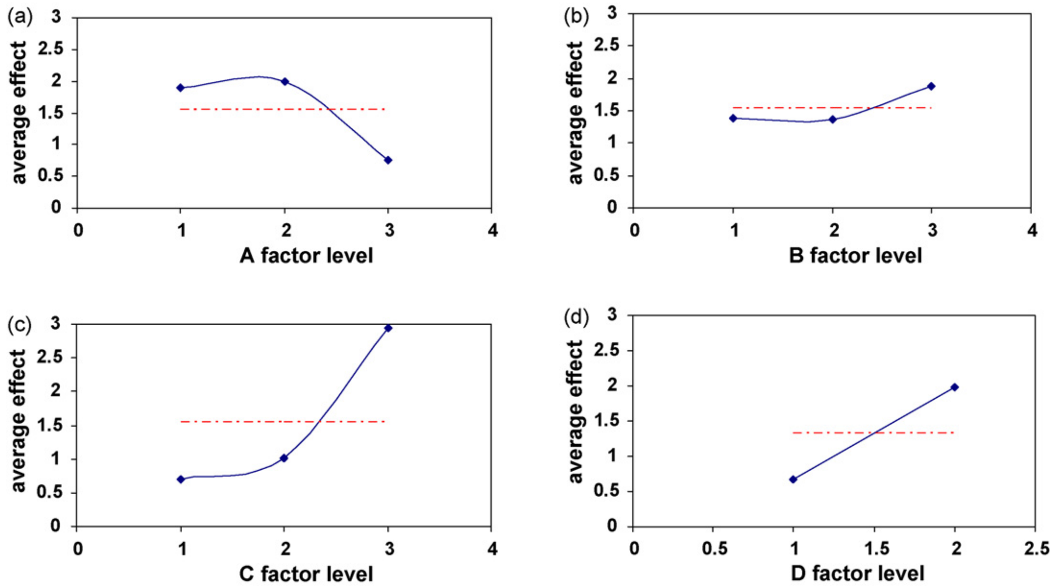

Fig. 4. Individual factor effect for $\varepsilon_s/d_{p,s}$ as the target function.

during centrifugation. As higher rotational speeds are used (A_3), the former phenomenon is outweighed by the enhanced segregation of the small and large particles due to a higher acceleration. This explanation is confirmed by the experimental values of the curvature for these samples (0.0216 and 0.028 for A_2 and A_3, respectively). The more segregation occurs, the higher the particle size gradient develops which results in a more concave shape (higher curvature) of the sintered compact [2].

It should be mentioned that although the system behaves quasi-linear for some factor level configurations far from the optimum, generally factor level interaction exists. This has been evaluated as the severity index (see ref. [8] for a proper definition of the severity index) and is summarized in Table 3. The severity index is expressed in percentage and a value of zero for it corresponds to no interaction between the factors considered.

The effect of binder concentration has been evaluated as the most determining one. According to Fig. 4c, increasing binder concentration from 2 to 4 wt.% results in a slight increase of the target function. Instead, a 6 wt.% binder concentration increases the target function remarkably. This can be explained resorting to the phenomenon of non-adsorbed binder migration to the upper surface during the drying stage. As the binder content exceeds the amount needed for adsorption on the available surface of the alumina particles, the extra dissolved PVA migrates to the upper layers through the upward flow of the solvent (water). As the binder cannot evaporate at the relatively low temperature of the drying step, it accumulates in solid form in these layers. Such a phenomenon is well known in ceramic processing and several researchers have reported it [9–10]. During the heating stage to reach the sintering temperature, the polymeric binder eventually evaporates/decomposes producing surface porosity.

The effect of increasing the pH from 4 to near 7 is significantly positive (Fig. 4d). This may be attributed to the fact that the initial slurry is highly dispersed at its native pH due to the effect of the Tiron surfactant. The optimum concentration of Tiron has been chosen 0.001 g (g alumina)$^{-1}$ according to our previous study [2]. Lowering the pH may enhance flocculation which furthermore results primarily in surface pore enlargement.

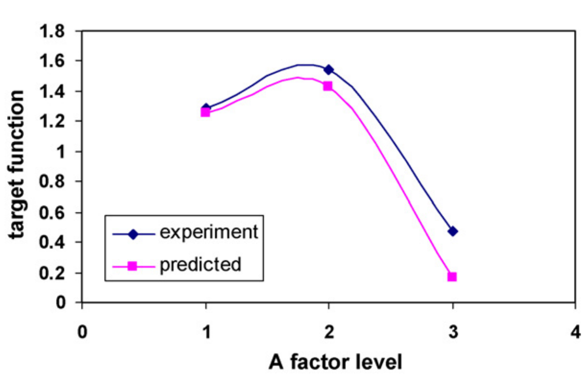

Fig. 5. Target function variation as a function of A level for a constant $B_2C_1D_2$ configuration.

- ($P \times t$) as target function

A proper membrane support should have a high permeability. As the permeability is also a function of membrane thickness, it is imperative to use the product $P \times t$ for comparing the permeance of membranes with different thicknesses.

Table 3
Severity index

Two-factor interaction	AB	AC	AD	BC	BD	CD
$\varepsilon_s/d_{p,s}$ as target function	35.43	21.19	16.21	6.66	31.47	6.58
Curvature as target function	54.65	62.55	23.79	35.87	32.18	12.92

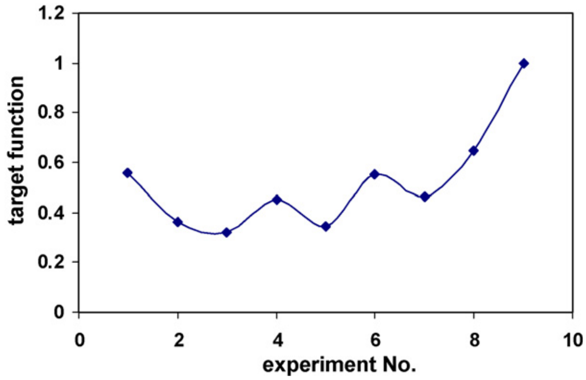

Fig. 6. Taguchi table experimental values for $P \times t$ as the Target function. The target function is evaluated as $(P \times t)/6.0595$. This has been done to make the maximum target function be equal to 1.

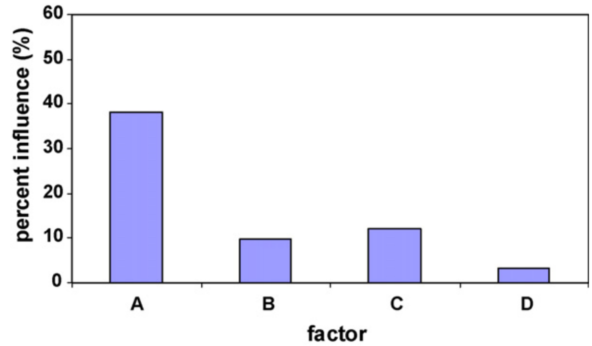

Fig. 7. Factor influence for $P \times t$ as the target function.

As far as the microfilter characteristics of the one-forming-step membrane are concerned, the higher the value of target function the lower resistance to flow is expected.

For membrane support applications usually $(P \times t)$ should not be lower than a minimum limit. This limit may be still lower if the sintered porous compact is used as support for ultra and nanofiltration applications as the controlling resistances to flow are usually the upper ultra or nanofilter layers.

Fig. 6 shows the experimental values of the target function for the different runs of the Taguchi table. The multimodal curve shows a significant maximum for run 9 ($A_3B_3C_1D_2$). The influence of factors on the target function as evaluated by ANOVA is shown in Fig. 7. The individual effects of factors are shown in Fig. 7a–d. The predicted optimal value belonging to the configuration $A_3B_3C_1D_1$ is 0.95 ± 0.13. According to Fig. 8a–d, the experiment number 9 ($A_3B_3C_1D_2$) is near to the predicted optimum configuration while the corresponding value of the target function is 0.89. Considering the calculated confidence interval of 0.13 for the target function at the predicted optimum, the latter result shows the absence of significant interaction of the factor levels near the optimum. Accordingly the effect of the individual factors near the optimum configuration will be explained as follows.

Referring to Fig. 8a, increasing acceleration increases the target function. Higher porosities increase the thickness and, theoretically, the permeability. One possible explanation for higher porosity of the centrifuged samples is the existence of fast flow streamlines of the liquid in the opposite direction of the depositing particles which enforce the creation of higher porosity and more connectivity of the pores during the forming process. Referring to Table 2, the sample due to experiment 9 has the maximum porosity (51.20%) among all the experiments. Also, the average pore diameter is substantially high (0.63 μm). To elucidate further the effect of acceleration on the pore microstructure evolution as cited above, the results of the additional experiment $A_1B_2C_1D_2$ mentioned earlier will be used. Considering $A_1B_2C_1D_2$ and $A_2B_2C_1D_2$ configurations, only the acceleration factor has been changed (from 0 to 2000 rpm). The predicted and experimental values of the target functions are (0.34, 0.42) and (0.35, 0.46) for the $A_1B_2C_1D_2$, and $A_2B_2C_1D_2$ experiments, respectively. This is while the porosity increases

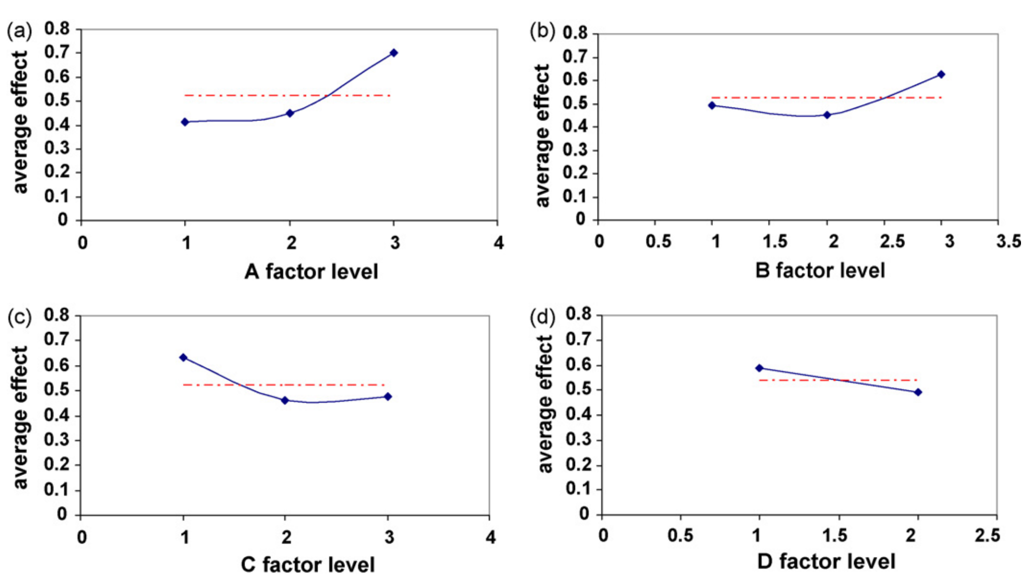

Fig. 8. Individual factor effect for $P \times t$ as the target function.

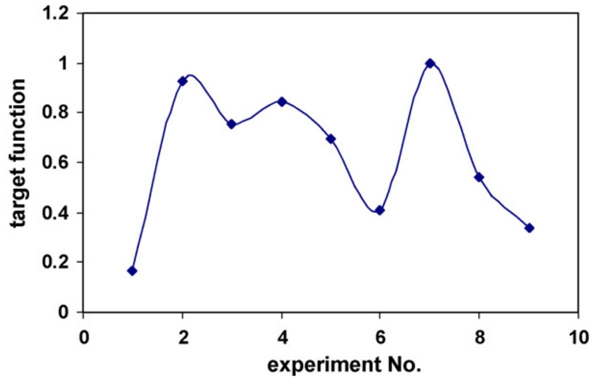

Fig. 9. Taguchi table experimental values for curvature as the target function. The target function is evaluated as (curvature)/0.03120. This has been done to make the maximum target function be equal to 1.

from 45.22 to 48.44% for the latter experiments, respectively. Higher porosity for centrifuged samples with respect to sediment samples have been reported by Falamaki and Veysizadeh [2] for a wide range of sintering temperatures.

The effect of binder content increase is detrimental, as far as $P \times t$ as target function is considered. Higher amount of binder (4 wt.%) results in a more compact green body due to a more appropriate slip rheology. More compaction results in enhanced sintering due a better packing and increased interface area between particles. This ultimately decreases the permeability of the sintered body. Fig. 8c also predicts that further increase of the binder content increases slightly the permeability. Actually if the binder content is larger than a minimum, the binder will act as a porosifier during the sintering stage.

Partial flocculation due to applying a pH of 4 increases $P \times t$ due to reduced segregation and production of particle agglomerates in the green compact (Fig. 8d).

- Curvature as target function

Curvature may be considered an important target function a priori considering the task of obtaining highly graded porous materials. To a large extent, the amount of curvature of the sintered sample is an indicator of the degree of the pore gradient in the green compact [2].

Referring to Fig. 9, it is observed that in contradistinction with $\varepsilon_s/d_{p,s}$ and $P \times t$ as target functions, curvature shows multiple local maxima but with minor difference in magnitude. In other words, the experimental observation shows that there are several factor-level combinations that yield highly graded green compacts. The predicted optimum combination of factor levels is $A_3B_2C_2D_2$ with the corresponding calculated value of target function equal to 0.90 ± 0.10. For this definition of the target function no justification exists to trying explaining the factors effects based on the individual factor effect plots. This is due to the high two-factor interactions present (see Table 3).

4. Conclusion

At this point it can be deduced that, as far as proper top-surface characteristics are needed, a one-forming-step microfilter or membrane support may be obtained by using the gravitational acceleration, together with proper volume, binder concentration and pH. Actually using this method requires long processing times. Our results show that using a 2000 rotation speed along proper other factor levels ($A_2B_1C_3D_2$) results in reasonable top-surface characteristics (see Fig. 2, experiment 4). This configuration has a distinct advantage over the sediment type one ($A_1B_3C_3D_2$): the (permeability $\times$ thickness) value is more than 40%.

Using ($P \times t$) as target function the optimum configuration deviates significantly from the one predicted using the $\varepsilon_s/d_{p,s}$ as target function. As far as the surface pore diameter should be minimum as a controlling parameter in the use of the microfilter/membrane supports, it is obvious that ($P \times t$) cannot be considered as a proper target function. Nevertheless, using the latter target function could elucidate to some extent the effect of each factor on the characteristics of the final product.

Despite being an index of particle segregation during the forming step, curvature has also been shown to be an inadequate target function.

The present work had two main outcomes. First, it was shown that the Taguchi approach could be successfully applied to improving the centrifugal processing route for membrane support manufacture. The second outcome is that the product of surface porosity x (1/surface average pore diameter) is a major membrane support characteristic and can be used effectively in evaluating the experimental results of statistical design of experiments.

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