PSO-ANFIS MODELING FOR PREDICTION OF TITANIUM-PILLARED CLAY POROUS CHARACTERISTICS

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INTRODUCTION

Clay-based materials have attracted much attention due to their bio-compatibility, cost-effectiveness, and high potential in catalytic and adsorption processes. Pillared clays are particular modified clays that can be prepared by replacing the charge-balancing cations in the interlayers of clays with polymeric (or oligomeric) cationic species. It is followed by calcination to convert these intercalated precursors into rigid and stable columns. Pillarization leads to permanent porosity in the host clay and improves its specific surface area and thermal stability. These factors make pillared clays a better choice than the parent clay in catalytic and adsorption processes [1]. The type of the pillaring agent and the operating conditions play a key role in determining the physicochemical properties of the resulting pillared clay products [2]. Al_2O_3, Fe_2O_3, ZrO_2, SiO_2, and TiO_2 can be considered predominant and widely used pillaring agents in the pillaring process [3].

Compared to other pillared clays, Titanium Pillared Clay (Ti-PILC) has unique properties such as higher thermal stability and larger interlamellar spacing and pore size, leading to exceptional catalytic adsorption properties. The pillaring solution containing titanium polyoxycations could be prepared by acid hydrolysis of titanium tetrachloride or titanium alkoxides as titanium sources [4]. Due to the considerable importance of operating conditions in the synthesis of Ti-PILC, performing optimization studies is recommended. The synthesis was carried out in these studies with different hydrolyzing agents, titanium sources, hydrolyzing agent/Ti ratios, Ti/clay ratios, clay suspension concentrations, pH values, and calcination temperatures. The most influential parameters were identified based on the general properties of Ti-PILCs in terms of surface area, pore-volume, basal spacing, acidity, and thermal stability [5–7]. The literature review showed that titanium tetrachloride as a titanium source and dilute clay suspension have been further investigated. Therefore, in the present study, titanium isopropoxide and more concentrated clay suspensions were applied for synthesis. The present work also investigates the influence of multiple parameters on the synthesis of Ti-PILC, which were not previously studied

OBJECTIVES

In the present work, the optimization of three operating parameters, including clay suspension concentration, acid/Ti ratio, and calcination temperature, was carried out. Physicochemical analytical analyses were used to characterize the products. Moreover, a mathematical approach, adaptive neuro-fuzzy inference system (ANFIS) method integrated with particle swarm optimization (PSO), was used to determine the relationships among the parameters.

MATERIALS & METHODS

The pillarization in this study was carried out based on the method previously proposed by Bineesh et al. with some modifications [8]. At first, the purification of the raw clay was carried out in two steps: acid washing followed by saturation with NaCl solution. Na-saturated clay suspension in distilled water was prepared at four concentrations (0.5, 1, 3, and 10% w/v). The pillaring solution was then prepared by slowly adding titanium alkoxide to a 2 M HCl solution under vigorous stirring until various H^+/Ti ratio values (2, 4, 6, and 8) were obtained. After aging the pillaring solution for 15 h, the resulting solution was added dropwise to the clay suspension with a ratio of Ti/clay = 10 mmol/g and stirred for 24 h. The clay was then added to the pillaring solution with a ratio of Ti/clay = 10 mmol/g and stirred for 24 h. The intercalated clay was separated from the pillaring solution by centrifugation; then, it was washed with distilled water, dried at 90 °C, and then calcined at 300 to 600 °C for 3 h at a rate of 3 °C/min in a tube furnace.

The PSO-ANFIS model was used to estimate the specific surface area of Ti-PILC samples. The PSO-ANFIS architecture consists of if-then rules based on the first-order Sugeno fuzzy model formed by Gaussian membership functions. This system was performed and simulated by calculating the error signals by the propagation method from the output layer back to the input nodes and by the backpropagation method. All the data were divided into two groups, 68% for training, and the rest was in the test data group. In the PSO-ANFIS model, subtractive clustering was chosen for FIS production as it gives the best performance. The PSO-ANFIS architecture used in this study is shown in Figure 1; there are a total of five layers in these systems.

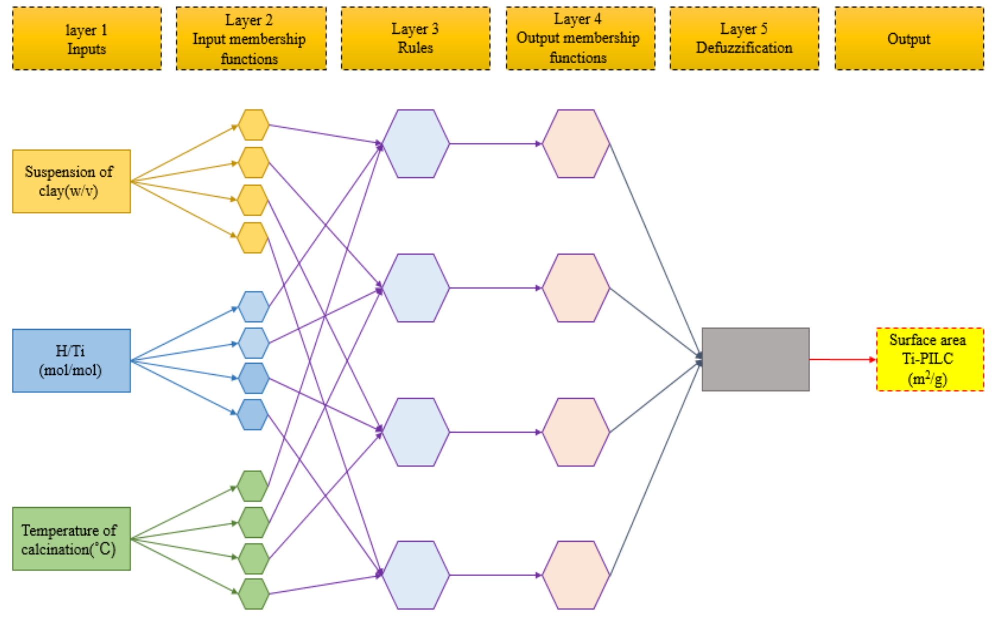

Figure 1: The structure of the PSO-ANFIS model in the present work.

RESULTS

As mentioned in the previous section, 64 experiments were performed. The variation of specific surface area at different concentrations of clay suspension, molar ratios of H^+/Ti and calcination temperatures are shown in Figure 2. According to Figure 2, it was found that the calcination temperature was not very effective in the specific surface area of pillared clays when the H^+/Ti ratio and the concentration of clay suspension were large enough. Moreover, a higher specific surface area was obtained in dilute clay suspensions when the calcination temperature was decreased. On the other hand, the results showed that the H^+/Ti ratio played a crucial role in dilute suspensions. The best specific surface area, 164 m^2/g, which was 310% greater than raw clay, was obtained at clay/water=0.5% (w/v), H^+/Ti=6, and a calcination temperature of 300°C.

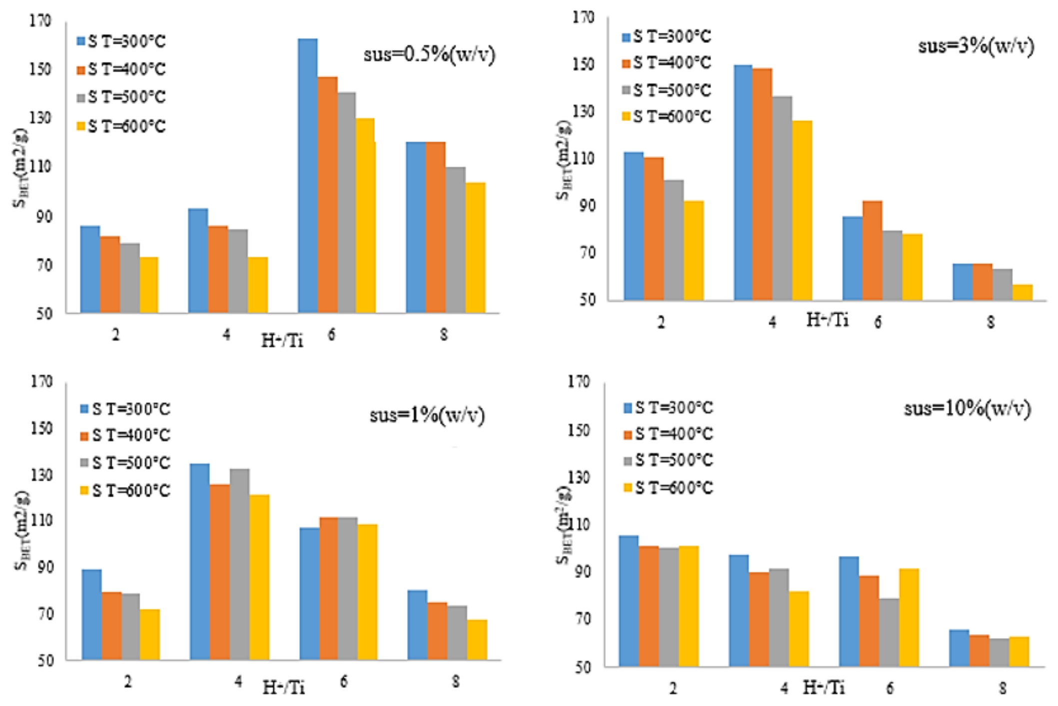

Figure 2: Effects of clay suspension concentration on the specific surface area of Ti-PILC at different ratios of H^+/Ti and calcination temperatures.

N_2 adsorption-desorption, XRD, FTIR, and FESEM analyses for the raw clay and Ti-PILC (optimum conditions) were carried out, and the results are shown in Figure 3. The montmorillonite, quartz, clinoptilolite, calcite, and cristobalite were detected in the XRD spectra of the raw clay in Figure 3-a. The XRD analysis of Ti-PILC confirmed the formation of rutile and anatase phases of TiO_2 in the interlayer region of the host clay. The vibrations of the functional groups -OH, Si-O-Si, Si-O, and Al-O were identified in the FTIR spectra in Figure 3-b. Several changes in the vibrations of the hydroxyl functional groups compared to the raw clay were observed as a result of the pillarization. The layered texture of clay and the heterogeneous surface texture of Ti-PILC can be seen in Figure 3-c. Finally, Figure 3-d showed the more developed porosity of Ti-PILC than clay and the improvement in the amount of adsorbed N_2.

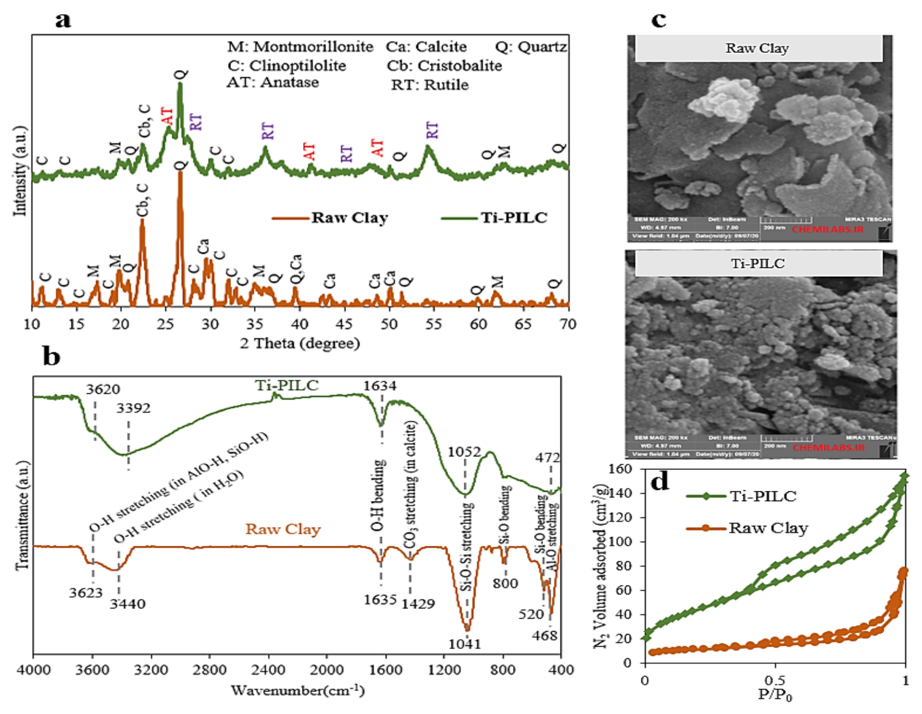

Figure 3: XRD patterns (a), FTIR spectra (b), FESEM micrographs (c) and N_2 adsorption-desorption isotherms (d) of clay and Ti-PILC.

Due to the complexity of the interaction between the factors, it is a real challenge to determine the effects of the different operating parameters. The present study showed the relationship between the specific surface area (output parameter) and the three input parameters. The parameters required to fit the PSO-ANFIS model and the results of its performance are presented by statistical indexes in Table 1. The three-dimensional (3D) prediction of PSO-ANFIS for the specific surface area as a function of the two input parameters is shown in Figure 4 based on the normalized data. According to Figure 4, the negligible effect on the specific surface area is related to the calcination temperature

Table 1: PSO-ANFIS model parameters and results.

Model details		Model evaluation criteria		
PSO-ANFIS parameters	value		Training	Testing
Number of nodes	38	R	0.9800	0.9700
Number of linear parameters	16	R^2	0.9600	0.9400
Number of nonlinear parameters	24	SSE	0.0460	0.0511
Total number of parameters	40	SAE	0.9063	0.7995
Number of training data pairs	44	MSE	0.0010	0.0026
Number of test data pairs	20	RMSE	0.0316	0.0510
Number of fuzzy rules	4	VFA	97.4000	92.9300

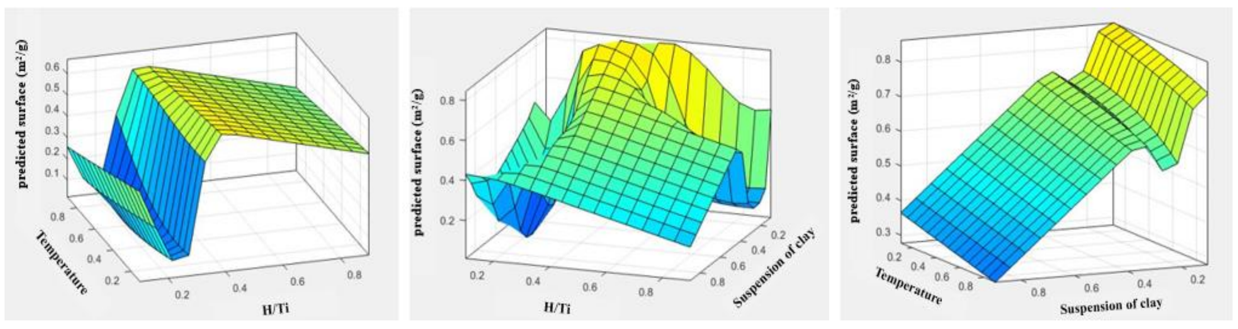

Figure 4: The three-dimensional prediction of PSO-ANFIS for the specific surface area of Ti-PILC.

CONCLUSIONS

The Ti-PILC samples were prepared, characterized, and the process was modeled using PSO-ANFIS. In the optimal conditions (clay/water=0.5% (w/v), H^+/Ti=6, and the calcination temperature of 300°C), a 4-fold increase in the specific surface area of Ti-PILC compared to raw clay was observed. The changes observed in the XRD patterns and FTIR spectra indicated the success of the pillaring process. The observations showed that the clay/water and H^+/Ti ratios had a noticeable effect on the change in the specific surface area compared to the calcination temperature. The results showed that the PSO-ANFIS model agreed with the experimental data with a correlation coefficient of 0.98, indicating the model's accuracy.

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