ALCHEMY Jurnal Penelitian Kimia

Official Website: https://jurnal.uns.ac.id/alchemy

ISSN 1412-4092
EISSN 2443-4183

Solvent and Heating Duration Effects on the Formation of Local Pyrophyllite PEG-4000 Hybrid Materials

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DOI: 10.20961/alchemy.21.2.104456.346-357

Received 19 June 2025, Revised 8 July 2025, Accepted 26 August 2025, Published 30 September 2025

Keywords:

clay-polymer hybrid;
FTIR;
polyethylene glycol 4000 local (Sumbermanjing) pyrophyllite;
sol-gel method.

ABSTRACT. Hybrid materials are new materials formed by combining two or more components to produce enhanced and distinct properties from the individual constituents. One example is the hybrid system of pyrophyllite and PEG 4000. This hybrid has not been reported and has potential for various applications. Local **(**Sumbermanjing) pyrophyllite-PEG 4000 hybrid was synthesized using casting and sol-gel methods with variations in solvent type (demineralized water, methanol, acetone) and heating time (1, 1.5, 2 hours) at a temperature of 100 – 105 °C. Acetone produced the highest yield (34.89%) due to superior solvent-material interactions, while two-hour heating achieved maximum yield (36.20%) through enhanced intermolecular bonding. Statistical analysis confirmed significant differences in solvent variations, while heating time showed no significant effect. FTIR characterization revealed specific functional groups O–H stretching (Al–OH linkage) at 3673.92 cm^{-1}, O–H stretching (3487.09 cm^{-1}), C–O stretching (1103.89 cm^{-1}), and Al–OH bending vibration at 841.46 cm^{-1}, thus confirming successful hybrid formation. Melting point analysis showed 128.13 °C (1 hour heating time) and 124.83 °C (2 hours heating time), indicating new material formation.

INTRODUCTION

A hybrid is a material formed by two or more different components with their unique attributes. During the hybridization process, the best characteristics of the components integrate to create a new material with superior traits. One of the most documented hybrid uses clay (filler) and polymer (matrix). Clay minerals are hydrous aluminum silicates with particular properties of large surface area, ion exchange capacity, thermal stability, and others (Balkanloo *et al*., 2022). A polymer is defined as a macromolecule made up of many repeating monomeric units connected by covalent bonds. Polymers can be either natural, semi-synthetic, or synthetic.

An example is polyethylene glycol (PEG). PEG is a synthetic polymer used for its hydrogel bioactive properties after being combined with clay (Gaballa *et al*., 2023). Adding clay to a polymer matrix like PEG allows for the formation of intercalated clay structures, which enhances the reinforcement provided by clay and the composite material's mechanical, thermal, and electrochemical properties (Hernández, 2016). Montmorillonit-PEG is a clay hybrid made of montmorillonite as a filler and PEG as a matrix with a melting temperature of 64.28 °C (Mahmood *et al*., 2017). In the montmorillonite group, pyrophyllite can be used as a substitute clay to produce a clay-PEG hybrid material.

Pyrophyllite is a clay commonly found in Indonesia, with the highest abundance in Sumbermanjing Wetan, Malang Regency, with 65,000 tons. Pyrophyllite is a neutral clay that forms a dioctahedral alumina sheet between two tetrahedral silica sheets (Martadiastuti *et al*., 2024). This clay has a chemical composition includes 57.7% silica, 16.7% alumina, 20.6% potassium, and other chemicals (Mutrofin *et al*., 2018). The pyrophyllite from

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Cite this as: Mutrofin, S., Shabiya, W. A., and Kamilah, H. (2025). Solvent and Heating Duration Effects on the Formation of Local Pyrophyllite PEG-4000 Hybrid Materials. *ALCHEMY Jurnal Penelitian Kimia*, 21(2), 346-357. http://dx.doi.org/10.20961/alchemy.21.2.104456.346-357.

Sumbermanjing Wetan consists of a high amount of silica of 85% (Winarno *et al.*, 2024). This high silica content contributes to high melting point values of pyrophyllite with 1580 °C (Ali *et al.*, 2021).

Hydrophilic polymers of polyethylene glycol (PEG) have a molecular weight between 200 and 35000 g/mol. Polyethylene glycol can be represented as $HO–(CH_2CH_2O)_n–H$, where ‘n’ indicates the number of repeating units of ethylene oxide, which determines the polymer molecular weight and physicochemical properties (Gaballa *et al.*, 2023). One example of a compound is polyethylene glycol 4000 (PEG-4000), a synthetic polymer with a molecular weight of 3500 – 4500 g/mol derived from ethylene glycol monomers (Money, 1989). This polymer has a melting point of 58 °C (Fadly *et al.*, 2017), commonly used as a humectant and skin conditioning agent in cosmetics, for concrete in construction, and as an emulsifier in the food industry (Gaballa *et al.*, 2023). PEG 4000 was chosen in this study due to its mid-range molecular weight, which affords a good blend of chain mobility and intercalation capability in forming a clay-PEG hybrid. The higher molecular weights of PEG can make a difficult intercalation process because they become mobile, flexible strands that coil into rigid and sluggish chains (Faradilla *et al.*, 2019).

Some methods are applied to synthesize a hybrid, including the casting and sol-gel methods. The casting method involves the formation of homogeneous composites through a solidification process, which results in improved mechanical properties and heat resistance (Adeleke *et al.*, 2022). Meanwhile, the sol-gel method is a method that transforms a liquid substance into a solid phase through hydrolysis or alcoholysis. This method can be carried out in low-temperature environments with particular particle size control (Danks *et al.*, 2016). Based on the exceptional properties of both materials, a composite of local pyrophyllite from Sumbermanjing and PEG 4000 can be produced by combining the sol-gel and casting methods with three solvent types and different heating durations. The composite produced was further characterized using Fourier Transform Infrared (FTIR) and melting point analysis.

RESEARCH METHODS

The materials used in this research are pyrophyllite obtained from Sumbermanjing District of South Malang, commercial PEG 4000, methanol p.a (99.5%), acetone p.a (99.5%), and demineralized water. The equipment and instruments utilized included a drop pipette (Onemed), glass stirrer (Pyrex), mortar stamper pestle (Onemed), 150 and 200 mesh sieve (Test Sieve), watch glass (Pyrex), digital balance, oven, glass funnel (Pyrex), desiccator (Duran), 50 mL and 250 mL beaker (Pyrex), 50 mL and 250 mL Erlenmeyer (Pyrex), 10 mL and 50 mL measuring cup (Pyrex), porcelain cup (Pyrex), bar magnetic stirrer (2 cm), magnetic stirrer (Thermo Scientific Cimarec), aluminum foil, capillary tube, FTIR spectrophotometer (Shimadzu IR Spiri-T) and melting point apparatus (Ivymen WRS-18).

Preparation of Sumbermanjing Pyrophyllite Solution

The pyrophyllite was ground into a powder and subsequently sifted with a 200-mesh screen. The powder that passed through the 200-mesh sieve and was held back by the 300-mesh sieve was collected. An amount of 100 g fraction was taken to serve as a sample with 200 – 300 mesh dimensions. Subsequently, the pyrophyllite was subjected to heating in a furnace at 300 °C for 5 hours. After heating, the pyrophyllite was allowed to cool in a desiccator. The pyrophyllite was then washed by mixing it with demineralized water at a ratio of 1:2 using a magnetic stirrer at 300 rpm for 45 minutes. The stirred pyrophyllite was filtered through Whatman 42 filter paper. Subsequently, the filtrate obtained was heated again in a furnace at 300 °C for 1 hour. The filtrate was then cooled in a desiccator, and the prepared pyrophyllite was collected. The 1.5 g of pre-processed pyrophyllite was placed into several 50 mL beaker glasses. Following this, 5 mL of each solvent (demineralized water, methanol, and acetone) was added to separate beakers containing the pyrophyllite. Each beaker was securely covered with plastic wrap. The mixtures were stirred using a magnetic stirrer for 1 hour at a speed of 900 – 1000 rpm to ensure thorough mixing and uniform distribution of the components. The procedure was repeated three times for each pyrophyllite solution in the various solvents.

Preparation of PEG 4000 solution

An amount of 2 g of PEG 4000 was weighed using an analytical balance. The PEG 4000 was then placed into several 50 mL beakers and dissolved using different solvents, namely 5 mL of demineralized water, methanol, and acetone, in separate beakers. Each beaker was securely covered with plastic wrap. The mixtures were stirred for 1 hour at a speed of 900 – 1000 rpm. The procedure was repeated three times for each PEG 4000 solution in various solvents.

Synthesis of Sumbermanjing Pyrophyllite-PEG 4000 Hybrid (Solvent Variation)

The prepared PEG 4000 solution from the previous stage was added to a beaker containing the pyrophyllite solutions in the same solvents. Then the beaker was covered with plastic wrap and was stirred using a magnetic stirrer for 3 hours at a speed of 900 – 1000 rpm. The lined empty petri dishes were prepared and weighed using an analytical balance. The suspensions that had been stirred were transferred into petri dishes. The suspensions were heated in an oven at 100 – 105 °C for 1.5 hours. The composites were placed in a desiccator for 7 days. After seven days of synthesis, the percentage yield of the composite in each variation was determined, and the solvent that produces the highest yield composite is selected as a solvent for the next stage. The percent yield of the hybrid was determined after being placed in a desiccator for 7 days. Equation 1 was used to calculate the percent yield of the hybrid.

$$\%\,Yield = \frac{A-C}{B-C} x\ 100 \tag{1}$$

Description :

A = dried composite + petri dish + baking paper (g)
B = wet composite + petri dish + baking paper (g)
C = petri dish + baking paper (g)

Synthesis of Sumbermanajing Pyrophyllite-PEG 4000 Hybrid (Heating Time Variation)

The composite in the heating time duration was prepared by combining the pyrophyllite solution and PEG 4000 solution using the selected solvent from the previous stage. Each beaker was covered with plastic wrap, and the solutions were stirred for 3 hours at 900 – 1000 rpm. The lined empty petri dishes were prepared and weighed using an analytical balance. After the stirring process, the resulting suspensions were transferred into petri dishes. The samples in the petri dishes were then heated in an oven at 100 – 105 °C for 1 hour and 2 hours. The composites were placed in a desiccator for 7 days. The percentage yield of the composite in each variation was calculated using Equation 1, and the highest percentage was determined after 7 days of synthesis**.**

Characterization of Sumbermanjing Pyrophyllite-PEG 4000 Hybrid

The hybrids with the highest percentage yield and the shortest heating duration were selected for further analysis. The higher yield percentage obtained indicates a good interaction between pyrophyllite and PEG 4000, while the shortest heating time was chosen due to the efficiency of the hybrid synthesis time. The selected hybrid underwent comprehensive characterization through several analytical techniques. Initially, FTIR spectroscopy was performed to identify the chemical bonds and functional groups in the synthesized hybrid samples by measuring the uptake of infrared radiation at specific frequencies. Consequently, melting point tests were conducted to evaluate the thermal properties of the hybrid, using a melting point apparatus (Ivymen WRS-18). Thermal testing using a melting point apparatus was carried out by placing approximately 1 mg of the hybrid into a capillary tube. The tube was positioned in the sample holder, with the initial temperature set at 110 °C and the heating rate maintained at 1 °C per minute. The test was initiated by pressing the *start* button and concluded when the initial and final melting temperatures of the hybrid were displayed on the screen.

Data Analysis

The data obtained in this study were analyzed using the IBM SPSS Statistics software. A one-way analysis of variance (ANOVA) was conducted to evaluate the effects of solvent variation and heating duration on the yield percentage in the synthesis of the Sumbermanjing pyrophyllite–PEG 4000 hybrid. Solvent type and heating time served as independent variables, with yield percentage as the dependent variable. Each treatment was performed in triplicate. Duncan's test was used for post-hoc comparison, and a T-test was conducted to evaluate statistically significant differences in melting point between sample groups. All statistical tests were conducted at a significance level of $\alpha = 0.05$.

RESULTS AND DISCUSSION

Percent Yield Data and Visual Appearance of The Hybrid

In solvent variation, the highest yield (34.89%) was obtained from the pyrophyllite–PEG 4000 hybrid synthesized using acetone, while the lowest yield (27.09%) was from the hybrid using demineralized water (Table 1). The methanol-based hybrid showed a yield of 34.06%. Acetone has the largest molecular diameter (6.16 Å) compared to the other solvents. This larger molecular diameter can promote more efficient enlargement of the silicate layers in the pyrophyllite structure, resulting in a broader interlayer spacing. This, in turn, allows PEG 4000 molecules to integrate more optimally into the pyrophyllite structure (Qiao *et al*., 2013). Demineralized water has the highest polarity index (10.2), indicating greater polarity than methanol (5.1) and acetone (5.1). However, since pyrophyllite is relatively hydrophobic and less polar, it interacts poorly with highly polar demineralized water, thereby hindering hybrid formation. In contrast, methanol and acetone, which have moderate polarity, enable better compatibility with pyrophyllite, facilitating hybrid synthesis. The amphoteric PEG 4000 dissolves well in all three polar solvents due to interactions between hydroxyl groups and solvent molecules, promoting uniform dispersion and influencing hybrid structure and properties (Özdemir and Güner, 2007).

Table 1. Percent yield data of the hybrid (solvent variation).

Hybrid code	Percentage yield (Average)
DW (Demineralized water)	27.09%
M (Methanol)	34.06%
A (Acetone)	34.89%

The visual appearance data presented in Table 2 indicate that all three types of hybrids synthesized using different solvent variations exhibited the formation of new materials, as evidenced by a noticeable change in color. The resulting hybrids displayed a dark brown color, contrasting the original light brown color of pyrophyllite and the transparent nature of PEG 4000. A hybrid prepared with methanol and acetone exhibits pores on the surface after oven heating at 100 – 105 °C, while those with demineralized water didn't show any pores on the surface of the hybrid (Table 2). This is attributed to the lower boiling points of methanol (64.6 °C) and acetone (56.0 °C), which evaporate more rapidly than water (100 °C), leading to pore formation due to rapid solvent vapor release (Zhao *et al*., 2024). Based on the yield percentage results, acetone was chosen as the optimal solvent for pyrophyllite-PEG 4000 hybrid synthesis due to its highest yield. The use of a high-yield solvent is essential to ensure effective interaction and dispersion between PEG 4000 and the pyrophyllite matrix, promoting stable hybrid formation and reducing material loss during synthesis. This selection is crucial for subsequent analyses, including heating time variation.

Table 2. Visual appearance of the hybrid (solvent variation).

Hybrid code	Hybrid before heating	Hybrid after heating
DW (Demineralized water)		
M (Methanol)		
A (Acetone)		

Furthermore, in heating time variation, based on percent yield data represented in Table 3, the highest percent yield (36.20%) was achieved at a heating duration of 2 hours at 100 – 105 °C. In contrast, the lowest yield was observed at 1.5 hours at 34.89%. Moreover, the hybrid that used an 11-hour heating time resulted in a 35.64% percent yield. This variation may be attributed to the influence of heating time on solvent evaporation, reaction efficiency, and potential thermal degradation during synthesis (Riesco-Avila *et al*., 2022). At shorter heating durations, such as 1 hour, the yield was higher than at 1.5 hours, possibly due to incomplete solvent evaporation.

However, after 2 hours, the yield increased again, likely due to improved solvent removal and the formation of a more stable hybrid structure.

Table 3. Percent yield data of the hybrid (heating time variation).

Hybrid code	Percentage yield (Average)
T1 (1 hour heating time)	35.64%
T1.5 (1.5 hours heating time)	34.89%
T2 (2 hours heating time)	36.20%

The visual appearance data presented in Table 4 indicate that all three types of hybrids synthesized using different heating time variations exhibited the formation of new materials, as evidenced by a noticeable change in color. The resulting hybrids displayed a dark brown color, contrasting with the original light brown of pyrophyllite and the transparent nature of PEG 4000. Additionally, the longer the heating time, the smaller the pore size in the resulting hybrid (Table 4). This happens because longer heating causes changes in the material structure, where the particles in the hybrid begin to fuse (sintering) and some pores are narrowed or even closed. At high temperatures, pore restructuring occurs due to increased internal pressure, making the pore size smaller. In addition, longer heating can cause pore collapse due to phase changes and material displacement within the hybrid structure (Li *et al*., 2021).

Table 4. Visual appearance of the hybrid (heating time variation).

Hybrid code	Hybrid before heating	Hybrid after heating
T1 (1 hour heating time)		
T1.5 (1.5 hours heating time)		
T2 (2 hours heating time)		

Percent Yield Statistical Analysis

The percent yield data from each variation were further analyzed using one-way ANOVA analysis and Duncan test to determine the treatment differences. In solvent variation, the One-way ANOVA results (Table 5) showed a significant difference among treatments ($F(2,6) = 56.65$, $p < 0.001$). Levene's test result (Table 6) shows each treatment's confidence interval and indicates that the assumption of equal variances is met ($p = 0.80$). The result of the normality test in Table 7 shows that all data groups are normally distributed and the normality assumption is satisfied ($p > 0.05$). Additionally, the post hoc analysis was subsequently performed to determine specific treatment differences**.** Duncan's Multiple Range Test (DMRT) showed groups of means within the same subset had no significant differences, while treatment groups containing means from different subsets differed significantly. The demineralized water treatment (mean = 27.09) was assigned to subset 1 (different from methanol and acetone treatments, means = 34.06 and 34.89). Meanwhile, methanol and acetone were treated as a combined treatment for purposes of analysis because each had a similar effect compared to demineralized water (Table 8).

Table 5. ANOVA for solvent variation.

Source of Variances	Sum of Squares	Df	Mean Square	F	Sig.
Between Groups	110.10	2	55.05	56.65	<0.001
Within Groups	5.83	6	0.97		
Total	115.93	8			

Table 6. Levene's test for homogeneity of variances and confidence interval.

Groups	**SD**	**95 % Confidence Interval for Mean**		**Levene Statistic**	**df1**	**df2**	**Sig.**
		Lower	Upper				
Demineralized Water	1.02	24.55	29.63	0.22	2	6	0.80
Methanol	1.09	31.34	36.79				

Table 7. Test of normality for solvent variation.

Groups	**Shapiro-Wilk**		
	Statistics	**Df**	**Sig.**
Demineralized water	0.83	3	0.21
Methanol	0.92	3	0.47
Acetone	0.77	3	0.06

Table 8. Duncan Multiple Range Test (DMRT) result (solvent variation).

Treatment	**N**	**Subset of alpha = 0.05**	
		1	**2**
Demineralized water	3	27.09	
Methanol	3		34.06
Acetone	3		34.89
Sig.		1	0.34

Moreover, the ANOVA test result (Table 9) shows there is no significant difference in heating time variation (F(2,6) = 1.23, p = 0.35). This result did not require further post hoc analysis, and it can be stated that there were no significant differences in heating time variation. The Levene's test result (Table 10) revealed the confidence interval and confirmed the assumption of equal variances within the data (p = 0.13). The normality test result, Table 11, exhibits that all data groups followed a normal distribution, and the normality assumption is fulfilled (p > 0.05).

Table 9. ANOVA for heating time variation.

Source of Variances	**Sum of Squares**	**df**	**Mean Square**	**F**	**Sig.**
Between Groups	2.62	2	1.31	1.23	0.35
Within Groups	6.38	6	1.06		
Total	9.00	8			

Table 10. Levene's test for homogeneity of variances and confidence interval.

Groups	**SD**	**95 % Confidence Interval for Mean**		**Levene Statistic**	**df1**	**df2**	**Sig.**
		Lower	Upper				
1 hour	0.34	34.78	36.51	2.88	2	6	0.13
1.5 hours	0.81	32.86	36.92				
2 hours	1.55	32.35	40.06				
Total	1.06	34.76	36.39				

Table 11. Test of normality for heating time variation.

Groups	**Shapiro-Wilk**		
	Statistics	Df	Sig.
1 hour	0.99	3	0.81
1.5 hours	0.77	3	0.06
2 hours	0.96	3	0.65

The percentage yield data acquired from the heating time versions and the usage of the chosen solvent from the solvent variation procedure, supported by statistical analysis, indicate that there is no significant difference among the heating time versions. Therefore, the pyrophyllite-PEG 4000 hybrid, organized with one-hour and two-hour heating durations, was selected for additional characterization using infrared and melting point analysis.

Further testing was carried out on the pyrophyllite-PEG 4000 hybrid synthesized with a two-hour heating duration, as it exhibited the best yield percent compared to the other variations. Additionally, the choice of the pyrophyllite-PEG 4000 hybrid with a one-hour heating period was based on time efficiency concerns, as its yield became not significantly different from that of the two-hour variation.

Fourier Transform Infrared Characterization

Fourier Transform Infrared Spectroscopy (FTIR) spectra of pyrophyllite–PEG 4000 hybrid made with heating times of 1 and 2 hours are presented in Figure 1. The spectra were acquired inside the interval of 4000 – 400 cm^{-1}. The intensity of each specific functional group from the raw material is recorded to identify the changes or shifts in the new spectra of the hybrid. The interactions between the components can be determined by comparing the intensities of the characteristic peaks. In the hybrid with one-hour heating time (Table 12), a peak at 3673.92 cm^{-1} was observed, indicating O–H stretching groups from Al–OH linkage in pyrophyllite, with an increase in vibration intensity from 82.21% to 87.95%. A blue shift was also observed in the O–H stretching region of PEG 4000, from 3428.61 cm^{-1} to 3501.35 cm^{-1}, accompanied by a slight decrease in intensity from 92.43% to 91.98%. Aliphatic C–H stretching vibrations exhibited a blue shift from 2882.37 cm^{-1} to 2883.80 cm^{-1} with increased intensity from 49.73% to 50.62%. The Si-O stretching group absorption from pyrophyllite showed a blue shift from 1121.00 cm^{-1} to 1146.67 cm^{-1} with a decrease in intensity from 61.94% to 57.36%. The C–O stretching groups of PEG 4000 confirmed a slight blue shift from 1098.18 cm^{-1} to 1101.04 cm^{-1}, with intensity increasing from 10% to 11.05%. A blue shift at 834.33 cm^{-1} from 841.46 cm^{-1} with a decrease in intensity from 67.5% to 19.30% occurs, indicating the Al–OH bending group from pyrophyllite. The red shift was also observed at 529.12 cm^{-1} compared to 531.98 cm^{-1}, indicating Si–O–Al bending from pyrophyllite with intensity increasing from 21.46% to 23.57%.

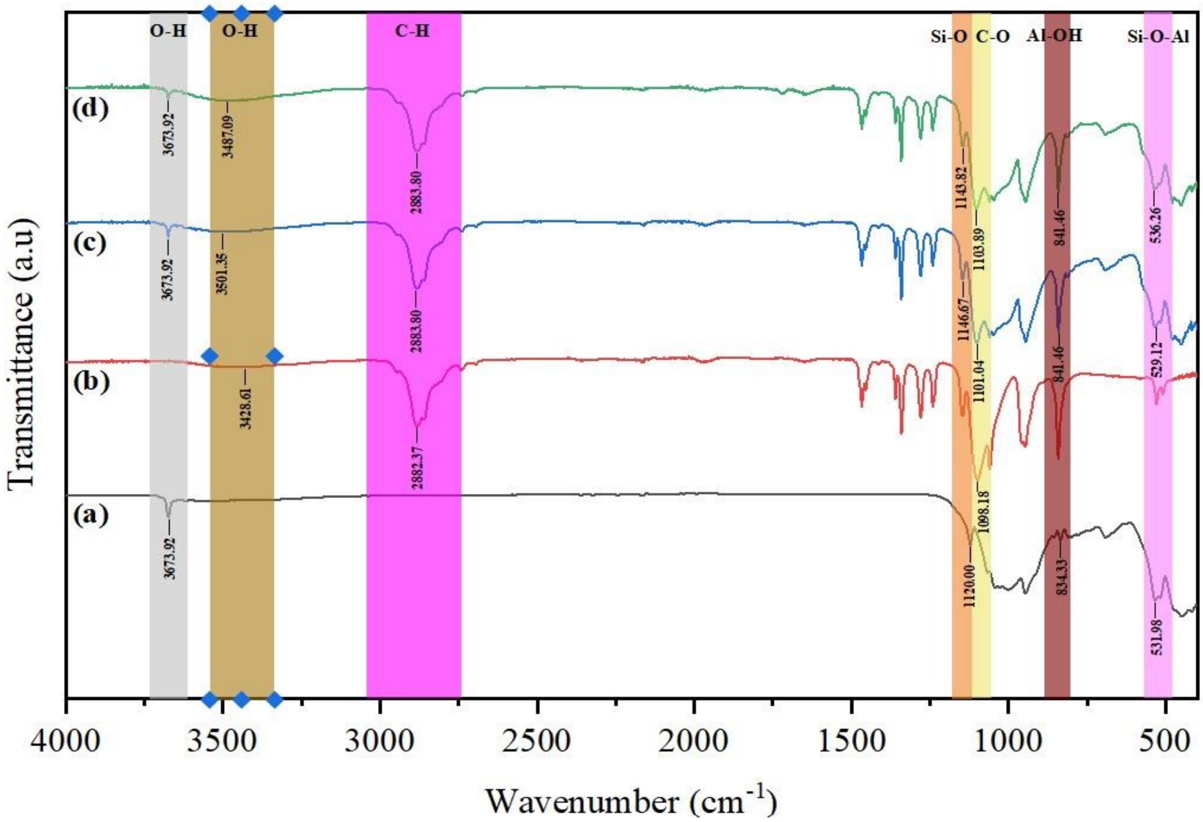

Figure 1. FTIR spectra (a) pyrophyllite, (b) polyethylene glycol 4000, pyrophyllite PEG hybrid prepared in (c) one-hour heating time, and (d) two-hour heating time.

Table 12. FTIR data analysis of pyrophyllite PEG hybrid prepared in a one-hour heating time.

Functional group	Sample	Wavenumber (cm^{-1})	Intensity (%)
O–H stretching	Pyrophyllite	3673.92	82.21
(Al–OH linkage)	Hybrid	3673.92	87.95
O–H stretching	PEG 4000	3428.61	92.43
	Hybrid	3501.35	91.98
Aliphatic C–H stretching	PEG 4000	2882.37	49.73
	Hybrid	2883.80	50.62
Si–O stretching	Pyrophyllite	1121.00	61.94
	Hybrid	1146.67	57.36
C–O stretching	PEG 4000	1098.18	10.00
	Hybrid	1101.04	11.05
Al–OH bending	Pyrophyllite	834.33	65.26
	Hybrid	841.46	19.30
Si–O–Al bending	Pyrophyllite	531.98	21.46
	Hybrid	529.12	23.57

Moreover, in the IR spectra of the hybrid prepared in a two-hour heating time (Table 13), a prominent absorption peak at 3673.92 cm^{-1} of O–H stretching vibrations in Al–OH linkage of pyrophyllite is observed with an increase in the intensity from 82.21% to 89.39%. There was a blue shift which was seen from O–H stretching vibrations of the PEG 4000 at 3428.61 cm^{-1} to the range of 3487.09 cm^{-1}, though the intensity decreased from 92.43% to 88.66%. A minor blue shift indicated an aliphatic C–H stretching group at 2882.37 cm^{-1} to the range of 2883.80 cm^{-1} and increased intensity from 49.73% to 51.78%. The Si–O stretching group absorption from pyrophyllite showed a blue shift from 1121.00 cm^{-1} to 1143.82 cm^{-1}, with a decrease in intensity from 61.94% to 55.13%. The C–O stretching groups of PEG 4000 confirmed a slight blue shift from 1098.18 cm^{-1} to 1103.89 cm^{-1}, with the stable intensity in 10%. Furthermore, a blue shift at 834.33 cm^{-1} from 841.46 cm^{-1} indicated the Al-OH bending group from pyrophyllite, with a decrease in intensity from 67.5% to 22.16%. The blue shift was also observed at 536.26 cm^{-1} compared to 531.98 cm^{-1}, indicating Si–O–Al bending from pyrophyllite with intensity increasing from 21.46% to 23.90%.

Table 13. FTIR data analysis of pyrophyllite PEG hybrid prepared in a two-hour heating time.

Functional group	Sample	Wavenumber (cm^{-1})	Intensity (%)
O–H stretching	Pyrophyllite	3673.92	82.21
(Al–OH linkage)	Hybrid	3673.92	89.39
O–H stretching	PEG 4000	3428.61	92.43
	Hybrid	3487,09	88.66
Aliphatic C–H stretching	PEG 4000	2882.37	49.73
	Hybrid	2883.80	51.78
Si–O stretching	Pyrophyllite	1121.00	61.94
	Hybrid	1143.82	55.13
C–O stretching	PEG 4000	1098.18	10.00
	Hybrid	1103.89	10.00
Al–OH bending	Pyrophyllite	834.33	65.26
	Hybrid	841.46	22.16
Si–O–Al bending	Pyrophyllite	531.98	21.46
	Hybrid	536.26	23.90

In the pyrophyllite-PEG 4000 hybrid synthesis process, hydrogen bond formation occurs between the oxygen atom of the hydroxyl group of polyethylene glycol (PEG) 4000 and the hydrogen atom on the Al–OH group of pyrophyllite in the octahedral layer on the basal side (Figure 2). This interaction causes an increase in the intensity of O–H vibrations bound to Al, indicated by an increase in the O–H stretching (Al–OH linkage) absorption peak at a wave number of 3673.92 cm-^{1} in the hybrid compared to pure pyrophyllite. In addition, the formation of hydrogen bonds also causes a reduction in the flexibility of the bending vibration angle of the Al–OH group in the pyrophyllite structure. This limits the deformation movement, so the intensity of Al–OH bending vibrations in the hybrid decreases, which is seen at a wave number of 841.46 cm-¹ compared to pure pyrophyllite.

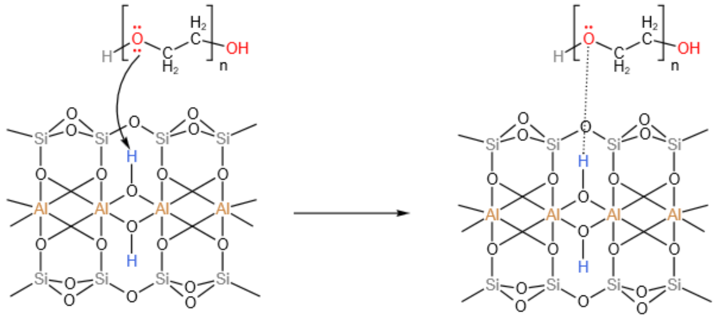

Figure 2. Hydrogen bond interaction between the oxygen atom in PEG 4000 and the hydrogen atom in pyrophyllite.

In addition, during the hybrid synthesis process, a Lewis acid-base interaction is suspected to occur between the oxygen atoms of the C–O group in PEG 4000 as a Lewis base and the aluminium atoms coordinated in the Si–O–Al structure of the pyrophyllite octahedral layer as a Lewis acid (Figure 3). In this interaction, the free electron pair of the PEG oxygen atom can bind to the Al atom, forming the possibility of a coordination bond. The formation of this interaction causes the C–O bond to become more polarized, so that the stretching vibrations of the C–O group become more active towards infrared absorption. This can be seen from the increase in the intensity of C–O stretching vibrations at a wavenumber of 1103.89 cm-¹ in the hybrid spectrum when compared to pure PEG 4000. Additionally, this interaction can also affect the environment around the Si–O–Al group in pyrophyllite, making Si–O–Al bending vibrations more active towards IR, which is seen from the increase in absorption intensity at wavenumber 531.98 cm-¹ in the hybrid compared to pure pyrophyllite.

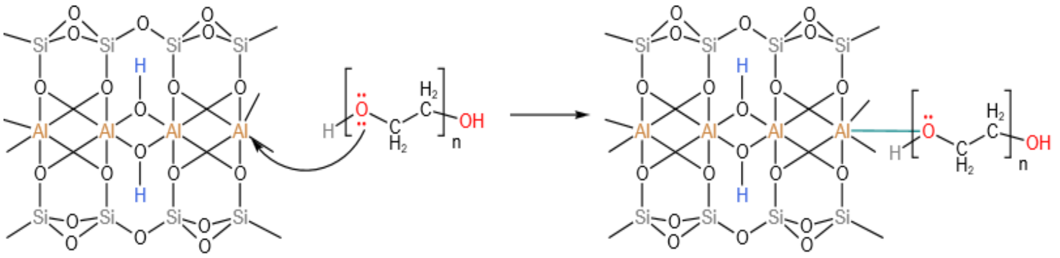

Figure 3. Covalent coordination bonding between oxygen atoms in PEG 4000 and aluminum atoms in pyrophyllite.

Moreover, during the hybrid synthesis process, it is suspected that hydrogen bond formation occurs between the oxygen atoms of the Si–O–Al group in the pyrophyllite octahedral layer (lateral side) and the hydrogen atoms of the hydroxyl group (–OH) in PEG 4000 (Figure 4). The formation of this hydrogen bond causes the dipole moment in O–H stretching vibrations to decrease, because some of the H atoms from OH interact with the O atoms from pyrophyllite. As a result, the intensity of O–H stretching vibrations in the hybrid decreased, which can be seen in the absorption band around 3487.09 cm-¹, compared to pure PEG 4000.

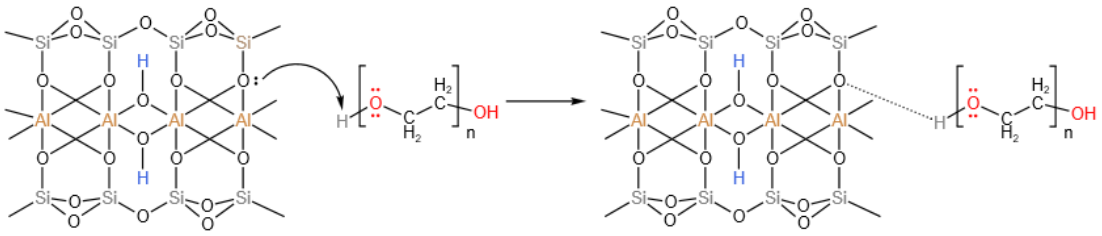

Figure 4. Hydrogen bonding between oxygen atoms in pyrophyllite and hydrogen atoms in PEG 4000.

Melting Point Characterization

The melting point analysis of pyrophyllite–PEG 4000 hybrid, synthesized at heating durations of 1 and 2 hours, revealed changes in thermal properties compared to the raw materials. The initial temperature used in the test is 110 °C, and the heating speed is 1 °C per minute. The melting point apparatus screen displays an initial and a final temperature after the melting point test finishes, indicating a temperature range at which the sample transforms from the solid to liquid phase (melt). During this characterization, the heat applied to the sample is absorbed, causing a phase change in the sample. The temperature at which this occurs wass recorded as the melting point.

The hybrid heated for 1 hour showed an average melting point of 128.13 °C, while the 2-hour variation had an average of 124.83 °C. These values, which fall between the melting points of pure pyrophyllite (~1580 °C) and PEG 4000 (53 – 58 °C), indicate successful interaction between the two materials, confirming the formation of a new hybrid structure (Ali *et al*., 2021; Fadly *et al*., 2017). Consequently, the melting point changes within the hybrid indicate the interaction between pyrophyllite and PEG 4000 during the synthesis technique. The pyrophyllite–PEG 4000 hybrid exhibited different melting points depending on heating duration. The one-hour heating time produced a higher average melting point than the two-hour variation. This difference may be attributed to reduced crystallinity at longer heating times.

Heating significantly affects the crystallinity of polymer-based hybrids by rearranging their molecular structures. However, prolonged heating seemed to reduce the number of crystalline regions because more chains achieved sufficient thermal energy to become amorphous well before melting (Denry *et al*., 2012). This loss of crystallinity as a result of thermal chain mobility occurs because the structured lattice is disrupted, leading to a lower degree of crystallinity and melting point. The irregularly ordered, “amorphous” regions are easier to flow past forces between molecules and require less energy, resulting in a lower melting point (Abdul *et al*., 2020). On the other hand, crystalline solids have their atoms arranged in a regular, three-dimensional pattern, leading to higher bond-breaking energy and melting point (Wieczerzak *et al*., 2023). In addition, longer heating times might have caused hybrid degradation, which could cause structural changes. Such degradation may weaken the PEG and pyrophyllite interaction with intercalated clay and thus lower the thermal stability of the hybrid and reduce its melting point (Leszczyńska *et al*., 2007).

In addition, the difference in surface charge of the two clays helps explain why the pyrophyllite–PEG 4000 hybrid has a higher melting point than the montmorillonite–PEG hybrid. In montmorillonite, increased layer charge decreases thermal stability because of structural defects and layer collapse during heating. This supports the concept that negative layer charge in montmorillonite contributes to its hybrids having lower melting points (Qin *et al*., 2021). However, pyrophyllite has a weaker surface charge, which means stronger interactions with PEG, leading to less intercalation. Consequently, PEG chains are more ordered, increasing the hybrid’s crystallinity and melting point.

Melting Point Statistical Analysis

The melting point of hybrids prepared in one- and two-hour heating time were analysed further with the T-test analysis. The mean value for the one-hour heating treatment was 128.13 with a standard deviation (SD) of 1.10, while the mean value for the 2-hour heating treatment was 124.83 with an SD of 0.58. Levene’s test for equality of variance indicated no significant difference in variances; thus, equal variances were assumed (F = 0.91, p = 0.39). Furthermore, the independent T-Test showed a considerable difference between one- and two-hour heating treatment (t(4) = 4.56, p = 0.01, 95% CI [1.29, 5.31]) (Table 14).

Table 14. T-test analysis of the hybrid melting point.

Heating Treatments	**N**	**Mean**	**SD**	**Levene’s Test**		**t**	**df**	**Sig. (two sided-p)**	**95% Confidence Interval of the Difference**	
				F	**Sig.**				**Lower**	**Upper**
One-hour	3	128.13	1.10	0.91	0.39	4.56	4	0.01	1.29	5.31
Two-hours	3	124.83	0.58							
Total	6	126.48	1.97							

CONCLUSION

This study successfully synthesized Sumbermanjing pyrophyllite-PEG 4000 hybrid using combined casting and sol-gel methods. Acetone proved to be the optimal solvent with the highest yield (34.89%), while 2-hour heating produced the most stable hybrid structure (36.20% yield). Statistical analysis confirmed significant differences among solvent treatments but no significant effect of heating time variations. Infrared confirmed the formation of new hybrids through the appearance of characteristic functional groups, including O–H stretching (Al–OH linkage) at 3673.92 cm^{-1}, O–H stretching (3487.09 cm^{-1}), C–O stretching (1103.89 cm^{-1}), and Al–OH bending vibration at 841.46 cm^{-1}. These spectra demonstrate successful chemical interaction between components. Melting point analysis showed new values of 128.13 °C and 124.83 °C for one- and two-hour heating durations, respectively, confirming the formation of a new material.

CONFLICT OF INTEREST

There is no conflict of interest in this article.

AUTHOR CONTRIBUTION

SM: Conceptualization, Methodology, Formulation, Funding, Resources, Supervision, Data Validation, Manuscript Review, Manuscript Revision; WHS: Synthesis, Characterization, Data Analysis, Manuscript Drafting; HK: Manuscript Review.

ACKNOWLEDGEMENT

The authors would like to express their sincere gratitude to the Chemistry Department and Inorganic Chemistry Laboratory of Brawijaya University for providing the facilities and support that made this research possible.

REFERENCES

Abdul, M., Najeeb, M., Mahdi, Shatha Hashim, Sattar, M.A., Mahdi, Shatha H, and Fadil, N.Q., 2020. The Effect of Heat Treatment on the Degree of Crystallinity and Grain Size for (N6, PS) Polymers. *Journal of Xi'an University of Architecture & Technology*.

Adeleke, A.A., Oki, M., Anyim, I.K., Ikubanni, P.P., Adediran, A.A., Balogun, A.A., Orhadahwe, T.A., Omoniyi, P.O., Olabisi, A.S., and Akinlabi, E.T., 2022. Recent Development in Casting Technology: A Pragmatic Review. *Revue des composites et des matériaux avancés*, 32, 91–102. https://doi.org/10.18280/rcma.320206.

Ali, M.A., Ahmed, H.A.M., Ahmed, H.M., and Hefni, M., 2021. Pyrophyllite: An Economic Mineral for Different Industrial Applications. *Applied Sciences*, 11, 11357. https://doi.org/10.3390/app112311357.

Danks, A.E., Hall, S.R., and Schnepp, Z., 2016. The Evolution of 'Sol–Gel' Chemistry as a Technique for Materials Synthesis. *Materials Horizons*, 3, 91–112. https://doi.org/10.1039/C5MH00260E.

Denry, I., Holloway, J.A., and Gupta, P.K., 2012. Effect of Crystallization Heat Treatment on the Microstructure of Niobium-doped Fluorapatite Glass-ceramics. *Journal of Biomedical Materials Research Part B: Applied Biomaterials*, 100B, 1198–1205. https://doi.org/10.1002/jbm.b.32684.

Fadly, T.A., Fauziyah, N.A., Rosyidy, A., Mashuri, and Pratapa, S., 2017. Degradation Activation Energy Determination of PEG 4000-Quartz Composites Using Dynamic Mechanical Analyzer (DMA) Measurements. p. 030033. https://doi.org/10.1063/1.4968286.

Faradilla, R.F., Lee, G., Sivakumar, P., Stenzel, M., and Arcot, J., 2019. Effect of Polyethylene Glycol (PEG) Molecular Weight and Nanofillers on the Properties of Banana Pseudostem Nanocellulose Films. *Carbohydrate Polymers*, 205, 330–339. https://doi.org/10.1016/j.carbpol.2018.10.049.

Gaballa, sherif, Naguib, Y., Mady, F., and Khaled, K., 2023. Polyethylene Glycol: Properties, Applications, and Challenges. *Journal of advanced Biomedical and Pharmaceutical Sciences*, 7, 26–36. https://doi.org/10.21608/jabps.2023.241685.1205.

Balkanloo, P.G., Marjani, A.P., Zanbili, F., and Mahmoudian, M., 2022. Clay Mineral/Polymer Composite: Characteristics, Synthesis, and Application in Li-Ion Batteries: A Review. *Applied Clay Science*, 228, 106632. https://doi.org/10.1016/j.clay.2022.106632.

Hernández, K.A.H., 2016. Polymer-Clay Nanocomposites and Composites: Structures, Characteristics, and Their Applications in the Removal of Organic Compounds of Environmental Interest. *Medicinal Chemistry*, 6, 201–210. https://doi.org/10.4172/2161-0444.1000347.

Leszczyńska, A., Njuguna, J., Pielichowski, K., and Banerjee, J.R., 2007. Polymer/Montmorillonite Nanocomposites with Improved Thermal Properties. *Thermochimica Acta*, 453, 75–96. https://doi.org/10.1016/j.tca.2006.11.002.

Li, T., Wu, J., Wang, X., and Huang, H., 2021. Particle Size Effect and Temperature Effect on the Pore Structure of Low-Rank Coal. *ACS Omega*, 6, 5865–5877. https://doi.org/10.1021/acsomega.0c06280.

Mahmood, W.A.K., Azarian, M.H., Wan Fathilah, W.F. bt, and Kwok, E., 2017. Nanoencapsulation of Montmorillonite Clay within Poly(Ethylene Glycol) Nanobeads by Electrospraying. *Journal of Applied Polymer Science*, 134, 45048. https://doi.org/10.1002/app.45048.

Martadiastuti, V., Winarno, T., Ali, R.K., and Purba, E.C., 2024. Characteristics of Pyrophyllite and Hydrothermal Alteration at Argotirto Area, Malang Regency, East Java, Indonesia. *IOP Conference Series: Earth and Environmental Science*, 1378, 012027. https://doi.org/10.1088/1755-1315/1378/1/012027.

Money, N.P., 1989. Osmotic Pressure of Aqueous Polyethylene Glycols. *Plant Physiology*, 91, 766–769. https://doi.org/10.1104/pp.91.2.766.

Mutrofin, S., Setyaningsih, T., Wati, F.N., and Purwonugroho, D., 2018. Physical-Chemistry of Nawangan's Phyropyllite and Its Prospective as Environmental Friendly Geopolymer Materials. *IOP Conference Series: Materials Science and Engineering*, 285, 012026. https://doi.org/10.1088/1757-899X/285/1/012026.

Özdemir, C., and Güner, A., 2007. Solubility Profiles of Poly(Ethylene Glycol)/Solvent Systems, I: Qualitative Comparison of Solubility Parameter Approaches. *European Polymer Journal*, 43, 3068–3093. https://doi.org/10.1016/j.eurpolymj.2007.02.022.

Qiao, Z., Wang, Z., Zhang, C., Yuan, S., Zhu, Y., Wang, J., and Wang, S., 2013. PVAm-PIP/PS Composite Membrane with High Performance for CO2/N2 Separation. *AIChE Journal*, 59, 215–228. https://doi.org/10.1002/aic.13781.

Qin, Y., Peng, T., Sun, H., Zeng, L., Li, Y., and Zhou, C., 2021. Effect of Montmorillonite Layer Charge on the Thermal Stability of Bentonite. *Clays and Clay Minerals*, 69, 328–338. https://doi.org/10.1007/s42860-021-00117-w.

Riesco-Avila, J.M., Vera-Rozo, J.R., Rodríguez-Valderrama, D.A., Pardo-Cely, D.M., and Ramón-Valencia, B., 2022. Effects of Heating Rate and Temperature on the Yield of Thermal Pyrolysis of a Random Waste Plastic Mixture. *Sustainability*, 14, 9026. https://doi.org/10.3390/su14159026.

Wieczerzak, K., Sharma, A., Hain, C., and Michler, J., 2023. Crystalline or Amorphous? A Critical Evaluation of Phenomenological Phase Selection Rules. *Materials & Design*, 230, 111994. https://doi.org/10.1016/j.matdes.2023.111994.

Winarno, T., Martadiastuti, V., and Puspitasari, E.I., 2024. Analysis of Pyrophyllite Quality as a Potential Industrial Raw Material in Argotirto Area, Sumbermanjing Wetan District, Malang Regency, East Java, Indonesia. *Journal of Geoscience, Engineering, Environment, and Technology*, 9, 259–265. https://doi.org/10.25299/jgeet.2024.9.3.10153.

Zhao, Q., Morawietz, T., Gazdzicki, P., and Friedrich, K.A., 2024. Effect of High-Boiling Point Solvents on Inkjet Printing of Catalyst Layers for Proton Exchange Membrane Fuel Cells. *Electrochimica Acta*, 508, 145273. https://doi.org/10.1016/j.electacta.2024.145273.