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Green Fabrication of Multi-functional Aerogel Composite from Fly Ash and Recycled Plastic Fibers for Heat and Sound Insulation

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Fly ash (FA), an abundant waste from thermal power plants in Vietnam, has been recycled 100 % into multi-functional aerogel composites for heat and sound insulation by the green fabrication developed in this study. The hollow structure of aerogel composites is created by sublimation of water in a mixture of FA and recycled polyethylene terephthalate (rPET) fibers bound by environmentally friendly adhesives including polyvinyl alcohol (PVA) and xanthan gum (XG). As result, the FA/rPET aerogel composites exhibit an exceedingly low density of 0.045 - 0.060 g/cm^3 and high porosity of 94.81 - 97.02 %. The aerogel composites have a low thermal conductivity of 0.035 - 0.040 W/(m·K), a comparably high noise reduction coefficient (NRC) of 0.49, and a high working temperature of 220 °C. The prepared FA/rPET aerogel composites display the ability to slow down the burning in the presence of FA at 3.0 wt% with a burning rate of 1.0 mm/s and extinguish the flame only 30 s. The effect of FA content on the morphology, physical and mechanical properties as well as thermal conductivity, acoustic insulation, and slow-burning of the aerogel composites is also investigated comprehensively. The features of slow-burning aerogel composites from FA and rPET fibers such as lightweight, flexibility, thermal and sound insulation give them great potential in civil engineering towards a green approach and sustainable development.

1. Introduction

In thermal power plants, after the combustion of coal to produce electricity, solid waste exists in two forms: the slag at the bottom and the lightweight ash consisting of fine particles obtained at the top of the furnace. Coal ash is generated approximately 600 – 800 Mt in the world (Jayaranjan et al., 2014). In Vietnam, the amount of ash and slag produced from domestic power plants is estimated at 19 x 10^6 t/y (Pham and Le, 2020). At present, fly ash (FA) is collected in dumps and large reservoirs surrounding the plants. The leaching and permeation of heavy metals and toxic chemicals from FA into groundwater have caused many serious environmental issues (Pham and Le, 2020). Studies on the utilization of FA as an alternative source to develop useful products should be carried out seriously. Recycling of FA, fabrication methods should be studied strictly. Not only solving the huge quantity of FA existed, but the conversion of FA into new materials should also limit the emission of toxic by-products into the environment. The procedure must be developed towards sustainable and eco-friendly criteria.

With the main composition of metal oxides (mostly silica and alumina) and poor flammability, FA could be considered as a flame retardant additive. Rejuvenated FA was utilized as fire resistance in polyvinyl alcohol (PVA)/Laponite composites aerogels (Kang et al., 2017). The raw FA was treated with 10 M NaOH solution before adding to PVA/Laponite mixture and freeze-drying to generate flame-retardant aerogels that could withstand smoke suppression. The addition of FA into the PVA/Laponite composite aerogels increases their

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thermal conductivity from 0.041 W/(m·K) (without FA) to 0.058 W/(m·K). Aerogels synthesized from fibers such as recycled polyethylene terephthalate (rPET) (Koh et al., 2018), pineapple leaves (Luu et al., 2019), sugarcane bagasse (Thai et al., 2019), to name a few, have extraordinary heat insulation but extremely poor fire resistance when exposed to flame. An addition of FA to the fiber-based aerogels may increase their flame retardancy but remain their low thermal conductivity. Morphologically, FA contains numerous hollow spherical particles that can capture and attenuate transmitted energies like sound and heat energy (Iordanidis et al., 2008). FA can improve the sound absorption of fiber-based aerogels and promote their acoustic insulation.

To the best of our knowledge, there has been no study on the utilization of raw FA without chemical pretreatment to fabricate flame-retardant FA/rPET aerogel composites for heat and sound insulation. In this study, the porous structure of rPET fibers is constructed via physical linking with PVA. FA is added to this base with an association of anti-settlement agent xanthan gum (XG). Finally, thanks to slow gelation of water at sub-zero temperature (-50 °C) and freeze-drying, the FA/rPET aerogel composites with high porosity above 90 % are obtained. Effect of FA content on their morphology, density, porosity, compressive durability, thermal conductivity, and acoustic performance as well as flame retardancy is also investigated to choose an appropriate synthesis condition.

2. Experimental

2.1 Materials

FA is collected from a local Vietnamese Thermal Power Plant, and sieved through a 50 μm sieve to obtain FA with uniform particle size. Its chemical composition is determined by X-ray Fluorescence spectrometry (XRF, Jasco FP-8500) and shown in Table 1. Its density (ρ_{FA}) is also determined at about 2.26 g/cm^3. rPET fibers are obtained from Nam Vang Ha Nam Company with a length of 64 mm and a density (ρ_{rPET}) of about 1.38 g/cm^3. XG with a density (ρ_{XG}) of 1.5 g/cm^3 is bought from England. PVA with a density (ρ_{PVA}) of 1.19 g/cm^3 and sodium hydroxide (NaOH) are purchased from China. All the solutions are made in distilled water (DW).

Table 1: Chemical composition of fly ash used for preparing FA/rPET aerogel composites

Chemical composition	Al_2O_3	SiO_2	Fe_2O_3	K_2O	CaO	TiO_2	Others
Content (wt%)	20.19	57.40	7.70	3.10	0.94	0.91	9.76

2.2 Fabrication of FA/rPET aerogel composites

rPET fibers are activated by immersed in aqueous NaOH 4 wt% with a solid-to-liquid ratio of 1:100 g/mL in 80 °C for 2 h to generate functional groups including carboxyl and hydroxyl groups (Koh et al., 2018). The fibers are then rinsed thoroughly with DW before drying at 60 °C for 12 h. The activated rPET fibers are blended in a blender to reduce their length to below 100 μm. PVA flakes are dispersed in DW and stirred continuously at 80 °C for 3 h to prepare a PVA solution of 0.50 wt%. FA and XG are added into the PVA solution with the desired ratio and stirred until a homogenous mixture is obtained at room temperature. The pre-treated fibers are cast on a mold, followed by slow addition of the FA/XG/PVA mixture. To distribute PVA universally into the matrix of fibers and FA and enhance the physical interactions among them, a sonication in 10 min is carried out, followed by incubation at 80 °C for 3 h. Finally, the mixture is frozen at -5 °C before freeze-dried in a pilot-scale Toption TPV-50F dryer at Institute for Tropical Technology and Environmental Protection to obtain the FA/rPET aerogel composites. The aerogel composites are fabricated with different FA concentrations of 1.0, 2.0, and 3.0 wt%. The control sample is also synthesized without FA.

2.3 Characterization

The densities of aerogel composites are calculated by measuring their weight and volume in the form of a cylinder. Their porosities (φ) are determined by Eq(1) based on their density (ρ_a) and average densities of components (ρ_b) calculated by Eq(2).

$$\varphi = \left(1 - \frac{\rho_a}{\rho_b}\right) \times 100\% \quad (1)$$

$$\rho_b = \frac{C_{rPET} + C_{FA} + C_{PVA} + C_{XG}}{\frac{C_{rPET}}{\rho_{rPET}} + \frac{C_{FA}}{\rho_{FA}} + \frac{C_{PVA}}{\rho_{PVA}} + \frac{C_{XG}}{\rho_{XG}}} \quad (2)$$

where C_{rPET}, C_{FA}, C_{PVA}, C_{XG} are the contents of rPET fibers, FA, PVA, and XG, before drying.

Morphologies and structure of the FA/rPET aerogel composites are observed based on their images captured by Scanning Electron Microscopy (SEM). Their thermal conductivities at ambient conditions (25 °C, 1 atm) are

recorded by TCi Thermal Conductivity Analyzer (C-Therm Technologies) with high accuracy. Thermal stability of the aerogel composites is analysed by Thermogravimetric Analysis (TGA) and Differential Thermal Analysis (DTA) from 25 ºC to 800 ºC in the air with a heating rate of 10 ºC/min. To investigate their sound insulation, SW422 and SW477 Impedance Tubes are used to measure their sound absorption coefficients following ASTM E1050-08 standard. To evaluate their ability to slow the burning down, pieces of FA/rPET aerogel composites with the same dimension of 125.0 x 13.0 x 3.2 mm are prepared. Each piece is exposed horizontally to the flame whose height is about 3.5 cm. The sample is burnt for 10 s and the combustion duration denoted time t_1 from the removal of sample out of the flame until it is completely extinguished is recorded. The burning rate is calculated by the length of the burnt sample divided by the time t_1.

3. Results and discussion

3.1 Morphology and structure of FA/rPET aerogel composites

The effect of FA content on the physical properties and morphology of FA/rPET aerogel composites is studied and shown in Table 2. As can be seen, the addition of FA into the fiber matrix causes the composite to become heavier and denser. With increasing FA content from 1.0 to 3.0 wt%, the density of aerogel composite rises from 0.045 to 0.060 g/cm^{3,} and its porosity declines from 97.50 to 94.81%. In comparison to other hybrid PVA aerogels such as Fe_3O_4/cellulose/PVA (0.10 - 0.12 g/cm^3), PVA/cellulose nanofiber (0.11 - 0.13 g/cm^3), the FA/rPET aerogel composites are much lighter (Koh et al., 2018).

Table 2: Physical properties and thermal conductivity of FA/rPET aerogel composites.

FA content (wt%)	rPET content (wt%)	PVA content (wt%)	Density (g/cm^3)	Porosity (%)	Thermal conductivity (W/(m·K))
0.0	2.0	0.5	0.036 ± 0.003	97.02 ± 0.08	0.040 ± 0.001
1.0	2.0	0.5	0.045 ± 0.004	97.50 ± 2.05	0.035 ± 0.001
2.0	2.0	0.5	0.054 ± 0.004	95.61 ± 0.38	0.037 ± 0.001
3.0	2.0	0.5	0.060 ± 0.004	94.81 ± 0.07	0.040 ± 0.001

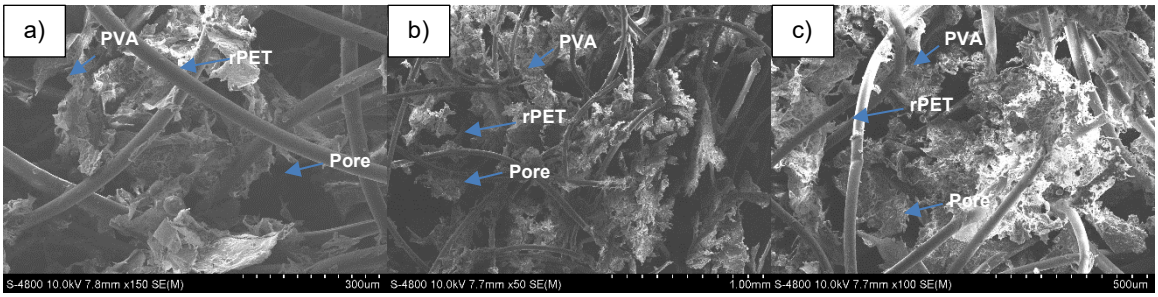

Figure 1: Porous structure of the FA/rPET aerogel composites with various FA contents: (a) 1.0 wt%, (b) 2.0 wt%, and (c) 3.0 wt%.

To study the morphology of FA/rPET aerogel composites, they are captured images by FE-SEM as shown in Figure 1. The matrix of rPET fibers interwoven with PVA/XG arrays is observed in Figure 1b. Following the synthesis procedure, FA particles are mixed with the homogenous PVA/XG solution before adding rPET fibers. The binders cover FA to form bundles and help FA to penetrate the fiber framework without a hitch. The liquid (water) in the mixture is gelled at extremely low temperature (-50 °C) and sublimated leaving voids between the rPET fibers and between the binder bundles. The increase in FA content causes the structure of FA/rPET aerogel composites to become denser meaning the decrease in porosity but an increase in their density. The results of morphological FE-SEM imaging have demonstrated the effectiveness of the developed synthesis. It is the addition of FA that maintains the hollow structure of rPET aerogels. The FA/rPET aerogel composites are expected to have better mechanical strength compared to the original rPET aerogels.

3.2 Thermal properties of FA/rPET aerogel composites

Due to the porous structure as depicted in Figure 1, FA/rPET aerogel composites are predicted to have good thermal insulation. Their thermal conductivity is measured by a highly accurate technique and tabulated in Table 2. It is interesting that although FA is mainly composed of oxides, the effective thermal conductivity of FA/rPET aerogel composite (0.035 W/(m·K)) is much lower than that of the control sample (0.040 W/(m·K)) at ambient

condition. Like other aerogels synthesized in a similar way that is the use of PVA as a binder, an addition of FA from 1.0 to 3.0 wt% causes a gradual increase in the effective thermal conductivity of the FA/rPET aerogel composites, from 0.035 to 0.040 W/(m·K). The developed aerogel composites have better thermal insulation than previous FA aerogels (0.042 – 0.050 W/(m·K)) prepared by using a PVA cross-linker as well (Duong et al., 2021). Compared to aerogels from different sources such as recycled cellulose (0.037 – 0.041 W/(m·K)) (Nguyen et al., 2014), silica/ceramic fibers composite (0.042 W/(m·K)) (Zhang et al., 2017) and commercial products like polyurethane (0.020 – 0.030 W/(m·K)), polystyrene (0.030 – 0.040 W/(m·K)), fiberglass (0.033 – 0.044 W/(m·K)), and mineral wool (0.030 – 0.040 W/(m·K)) (Jelle, 2011), the FA/rPET aerogel composites have equivalent thermal conductivity and prove to be a promising new alternative material in the near future.

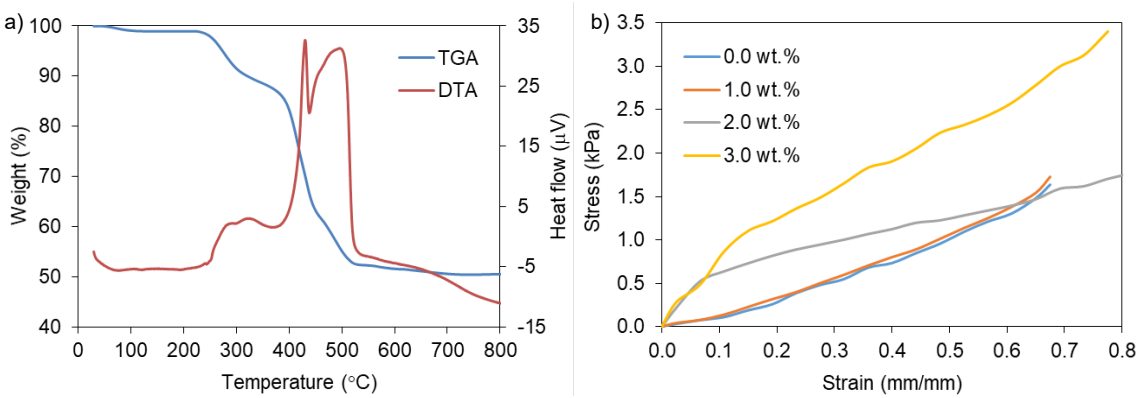

Figure 2: (a) TGA and DTA of the FA/rPET aerogel composite synthesized from 2.0 wt% FA and (b) stress-strain curves of the aerogel composites with various FA contents.

The thermal stability of FA/rPET aerogel composite is studied based on the TGA result illustrated in Figure 2a. The sample prepared from 2.0 wt% FA has two main thermal degradable stages: (i) 220 – 400 °C and (ii) 400 – 540 °C. In the first stage, nearly 15 % of weight loss is witnessed in Figure 2 due to the decomposition of binders including XG (235 °C) and PVA (280 °C). The sample is degraded about 35 % of its weight in the next stage because of rPET fibers decomposition at 430 °C (Koh et al., 2018). The remaining weight of FA/rPET aerogel composite is constant up to 800 °C and is known as FA. The thermal degradation of FA/rPET aerogel composite is primarily exothermic because the peaks on the DTA graph are all upward, especially in the stage above 400 °C. The maximum heat flow is recorded at about 35 μV and the minimum one is approximately 5 μV.

3.3 Compressive strength of FA/rPET aerogel composites

The results of the compressive test on FA/rPET aerogel composites are displayed in stress-strain curves as depicted in Figure 2b. The curves of all surveyed aerogel composites are above that of the control sample indicating that FA has a great effect on improving the durability of aerogels containing only rPET fibers. Although the aerogel composites are composed of FA and rPET fibers, they still exhibit excellent flexibility as demonstrated by their low Young's modulus of 1.21 kPa compared to that of the control sample (1.01 kPa). More and more FA in the aerogel composites makes them more durable as evidenced by a proportional increase in Young's modulus from 1.21 to 7.57 kPa with increasing FA content from 1.0 to 3.0 wt%. The aerogel composite containing 3.0 wt% FA has the highest compressive strength among the investigated samples and equal thermal conductivity compared to the control sample, so it is expected to have the highest sound absorption coefficient as well to become versatile insulation.

3.4 Acoustic insulation of FA/rPET aerogel composites

The aerogel composites are expected to be sound insulation based on the determined physical properties in Table 2. The acoustic performance of studied aerogel composites with the same thickness of 30 mm illustrates in Figure 4. In the low frequency from 50 to 3,000 Hz, the sound absorption coefficient denoted α of all samples increases sharply and approaches 100 % at 3,000 Hz. In the higher frequency range, α fluctuates in the range of 90 – 100 % due to the coincidence dip phenomenon (Soltani and Zerrebini, 2012). The noise reduction coefficients (NRC) of the control sample and aerogel composites prepared from 1.0, 2.0, 3.0 wt% with the same thickness of 30 mm are determined 0.36, 0.38, 0.44, and 0.49. The results demonstrate that the presence of FA in the fibers network increases the acoustic insulation of FA/rPET aerogel composites significantly. Based on

SEM images, the structure of FA/rPET aerogel composites have large packages of FA and PVA attached to the rPET fiber framework. The energy of sound passing through these packages is spread out and subsequently, reduced because of friction and vibration of the fibers (Nandanwar et al., 2017). As the content of FA increases, the aerogel composites become denser and their sound absorption coefficient increases as seen in Figure 4. Our FA/rPET aerogel composites also exhibit better sound absorption than commercial soundproof Basmel whose NRC is 0.40 in the same thickness (Do et al., 2020).

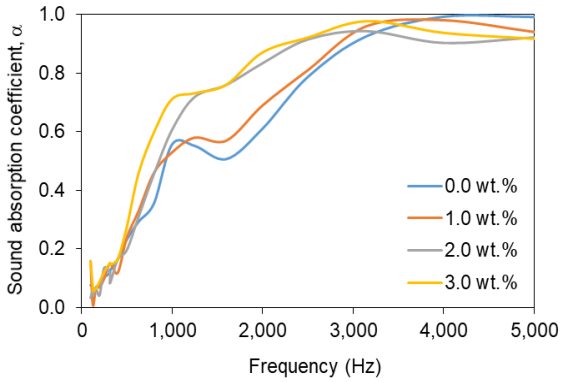

Figure 4: Sound performance of FA/rPET aerogel composites with increasing FA content.

3.5 Preliminary test on flame retardancy of FA/rPET aerogel composites

The results of the preliminary combustion test on investigated FA/rPET aerogel composites are shown via two factors of burning rate and combustion duration as displayed in Figure 5. The test shows that the control sample seems to be burnt completely when exposed to the flame with a rapid burning rate of 2.5 mm/s. This is because its composition consists of only rPET fibers and binders which have a low thermal degradation temperature of below 500 °C. When FA is combined with rPET fibers and binders, the burning rate is reduced to 1.8 mm/s with an increase in combustion duration from 50 to 68 s in the sample containing 1.0 wt% FA. The increase in FA content prolongs the combustion time and causes the burning rate to decrease. It is observed that samples containing 1.0 wt% and 2.0 wt% FA are all burnt out, while the aerogel composite having 3.0 wt% FA has the best flame retardancy to extinguish the flame in only 30 s and the lowest burning rate of 1.0 mm/s. This can be explained due to the metal oxides and inorganic oxides in the FA adhere to the PVA surface and act as a barrier to trap the flame into its spherical particles, slow down the heat diffusion through the aerogel composite. FA in the aerogel composites generates a high char barrier to delay heat transfer and spread of flame largely due to the presence of silica in it as well (Chen and Schiraldi, 2018).

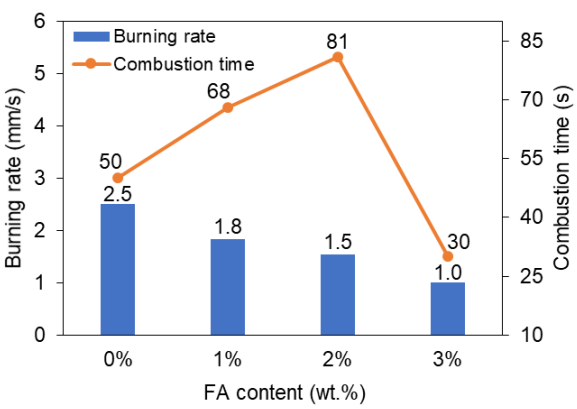

Figure 5: Flame retardancy of investigated FA/rPET aerogel composites with increasing FA content.

4. Conclusion

The FA/rPET aerogel composites are successfully synthesized from a combination of FA and rPET fibers by using PVA/XG cross-linkers and the sublimation method. The results also demonstrate the initial hypothesis that using FA as an additive to the rPET fiber network cross-linked by PVA and XG binders improves the

mechanical strength, soundproofing performance, fire retardancy of the aerogel composite but maintains its low thermal conductivity which is equivalent to the control sample. The developed aerogel composites have proven their versatility thanks to their outstanding results in terms of low density, high porosity, poor thermal conductivity, and good sound absorption. The preliminary assessment shows that the FA/rPET aerogel composites can delay burning effectively, so further studies should be performed in triplicate samples to evaluate comprehensively the criteria regarding their flame retardancy and to strengthen their feasibility as an insulating material.

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References

Chen H.B., Schiraldi D.A., 2018, Flammability of polymer/clay aerogel composites: An overview, Polymer Reviews, 59(1), 1-24.

Do N.H.N., Luu T.P., Thai Q.B., Le D.K., Chau N.D.Q., Nguyen S.T., Le P.K., Phan-Thien N., Duong H.M., 2020, Heat and sound insulation applications of pineapple aerogels from pineapple waste, Materials Chemistry and Physics, 242, 122267.

Duong H.M., Ling N.R.B., Thai Q.B., Le D.K., Nguyen P.T.T., Goh X.Y., Phan-Thien N., 2021, A novel aerogel from thermal power plant waste for thermal and acoustic insulation applications, Waste Management, 124, 1-7.

Iordanidis A., Buckman J., Triantafyllou A.G., Asvesta A., 2008, ESEM–EDX characterisation of airborne particles from an industrialised area of northern Greece, Environmental Geochemistry and Health, 30, 391–405.

Jayaranjan M.L.D., van Hullebusch E.D., Annachhatre A.P., 2014, Reuse options for coal fired power plant bottom ash and fly ash, Reviews in Environmental Science and Bio/Technology, 13, 467–486.

Jelle B.P., 2011, Traditional, state-of-the-art and future thermal building insulation materials and solutions—Properties, requirements and possibilities, Energy and Buildings, 43(10), 2549-2563.

Kang A.H., Shang K., Ye D.D., Wang Y.T., Wang H., Zhu Z.M., Liao W., Xu S.M., Wang Y.Z., Schiraldi D.A., 2017, Rejuvenated fly ash in poly(vinyl alcohol)-based composite aerogels with high fire safety and smoke suppression, Chemical Engineering Journal, 327, 992-999.

Koh H.W., Le D.K., Ng G.N., Zhang X., Phan-Thien N., Kureemun U., Duong H.M., 2018, Advanced recycled polyethylene terephthalate aerogels from plastic waste for acoustic and thermal insulation applications, Gels, 4(2), 43-55.

Luu T.P., Do N.H.N., Chau N.D.Q., Lai D.Q., Nguyen S.T., Le D.K., Thai Q.B., Le P.T.K., Duong H.M., 2019, Morphology control and advanced properties of bio-aerogels from pineapple leaf waste, Chemical Engineering Transactions, 78, 433-438.

Nandanwar A., Kiran M.C., Varadarajulu K.C., 2017, The development of sound absorbing materials using natural bamboo fibers, High Performance Structures and Composites, 7, 1-9.

Nguyen S.T., Feng J., Ng S.K., Wong J.P.W., Tan V.B.C., Duong H.M., 2014, Advanced thermal insulation and absorption properties of recycled cellulose aerogels, Colloids and Surfaces A: Physicochemical and Engineering Aspects, 445, 128-134.

Pham N.Q., Le A.K., 2020, Coal fly ash in Vietnam and its application as a lightweight material, Chemical Engineering Transactions, 83, 31-36.

Soltani P., Zerrebini M., 2012, The analysis of acoustical characteristics and sound absorption coefficient of woven fabrics, Textile Research Journal, 82, 875–882.

Thai Q.B., Nguyen S.T., Ho D.K., Tran T.D., Huynh D.M., Do N.H.N., Luu T.P., Le P.K., Le D.K., Phan-Thien N., Duong H.M., 2019, Cellulose-based aerogels from sugarcane bagasse for oil spill-cleaning and heat insulation applications, Carbohydrate Polymers, 228, 115365.

Zhang H., Fang W.Z., Wang X., Li Y.M., Tao W.Q., 2017, Thermal conductivity of fiber and opacifier loaded silica aerogel composite, International Journal of Heat and Mass Transfer, 115, 21-31.