Journal of Thermal Engineering Yildiz Technical University Press, Istanbul, Turkey Vol. 2, No. 6, Special Issue 5, pp. 978-982, December, 2016. http://eds.yildiz.edu.tr/journal-of-thermal-engineering/Articles Manuscript Received December 29, 2015; Accepted February 27, 2016

This paper was recommended for publication in revised form by Regional Editor Kwok-wing Chau

SEPIOLITE FOR POWDER-GLASS FIBER HYBRID CORE MATERIALS FOR VACUUM INSULATED PANELS: CRITICAL INNER PRESSURE AND THERMAL INSULATION PERFORMANCE FOR LONG SERVICE-TIME APPROACH

Zhao Feng Chen

Nanjing University of Aeronautics And Astronautics College of Material Science and Technology Nanjing, China

*Deniz Eren Erişen

Nanjing University of Aeronautics And Astronautics Super Insulation Composites Laboratory Nanjing, China

Keywords: Sepiolite, VIP, insulation, silicate, glass fiber, Knudsen, sustainable development * Corresponding author: D. E Erişen, Phone: +90551 154 5767 E-mail address: deerisen@anadolu.edu.tr

ABSTRACT

With increasing driving force on insulation panels with higher insulation performance with lower thicknesses, create a market need to develop new insulation materials. Especially, vacuum insulated panels as superinsulation materials, still have an opportunity to invent new core materials from locally occurring materials like natural fibers and different kind silicates with high porosity like zeolite, sepiolite, diatomite etc. That kind of materials can be used to produce powder-glass fiber hybrid core materials. They have the positive effect on the critical inner pressure of core material. Sepiolite is a Magnesium Hydroxyl Silicate clay with high surface area and fiber-like structure. So that makes the sepiolite is an interesting material for superinsulation research works. Investigation of glass fiber vacuum insulation panels is well justified. As seen from the work, the low sepiolite content and without sepiolite addition glass fiber core materials show better thermal insulation performance. The performance differences on thermal insulation. overall the objectives have been achieved in this study.

INTRODUCTION

After the driving force of economy and requirements of sustainable development or regulations of governments, tend to environmental protection; civilization gives more effort to invent new technologies or applications to increase energy efficiency than before. Especially after the Kyoto Protocol's targets, the developed countries start to force themselves to decrease total CO2 production per year. As a result of some research on developed countries; responsibility of modest quarter of national total energy consumption is addressed by building. On the other hand, developed countries already had got insulation

technologies at buildings and building appliances. If the force of total pressure on buildings which already have insulation on their covers; tend them to decrease their energy consumption what they have to get else besides of consuming less than before; could be increasing their insulation thicknesses or use more advanced materials or technologies can decrease energy efficiency of buildings and home appliances of buildings. As a result of mining new technologies, some materials which have modest ten times more heat resistivity at same thicknesses invited; they called as superinsulation materials. Vacuum insulated panels beside of gas-filled panels, aerogels, or some special foam-like materials; are good alternative superinsulation materials. Vacuum insulation panel is a composite material which is assembled of an envelope which have:

- High barrier properties for gas and humidity diffusion through core material,
- High reflectivity on thermal radiation,
- Low horizontal conduction to eliminate thermal bridging

and an evacuated core material which has:

- Easy to evacuate,
- Low solid conduction and high porosity,
- Opaque for thermal radiations.

So we can summarize as vacuum insulated panels are a composite of an evacuated core material which has mechanical and insulation properties and high barrier film which protect the vacuum below or behind of critical inner pressure of core material during the life cycle.

CRITICAL INNER PRESSURE OF CORE MATERIAL

Critical inner pressure is the point the pressure of vacuum which the thermal resistivity of evacuated core material changes radically. This point is seen as the point of inflection at the function of heat transfer of gas and gas pressure of media. That phenomenon can be explained by several works on the kinetic theory of Knudsen. As a simple statistic method of Knudsen; the relationship between the mean free path of gasses and mean wall size of porous media has an important role on the thermal resistivity of vacuum insulated panel. At very low Knudsen number than 1 (~10-3), the collision between gas molecules is dominant for gas conduction. It is called as continuum range at kinetic theory:

$$k_g = 1/3\rho_g C_V \lambda_g c \tag{1}$$

where the k_g is gas conduction λ_g is the mean free path of gas, C_V is the specific heat at constant volume and ρ_g is the density of gas, c is the speed of sound. Density of the gas depends to the pressure and as a reverse of it; the mean free path of gas decrease with the pressure. So the thermal conduction of the gas at a continuum range is an independent value than the gas pressure. But when the Knudsen number is close to the $(10^{-2} \sim 10^{0})$. We can use another equation to compute the gas conductivity of a gases at transition range. Because at this conditions the gas molecules tend to make a collision between pore walls rather than make a collision between them. We can say the gas molecules are trapped inside pore even the pores have connection between them. So at that situation thermal conduction of gases statistically more affected by pressure decreasing. Those condition is a balanced between molecule-molecule collision and molecule-wall collision. Conduction of gas molecules decreases sharply by decreasing pressure and convection between pore walls increases slowly. And the thermal conduction of gas is described with Knudsen number as many previous works.[2]

$$k_g = \frac{k_{g,0}}{1+2\beta K_n}$$

$$K_n = \frac{\lambda_g}{\ell_s}$$

$$\lambda_g = \frac{c_B}{\sqrt{2}d_g^2 P_a}$$

$$(2)$$

$$(3)$$

$$K_n = \frac{\lambda_g}{\ell_s} \tag{3}$$

$$\lambda_g = \frac{c_B}{\sqrt{2}d_g^2 P_a} \tag{4}$$

where the β is constant for the material and the gas; d is the diameter of gas molecule; cB is the Boltzmann constant; Pa is the total gas pressure; Is is the average wall size of the pores.

Theoretically; Kn can be zero or very close to the zero.

At that conditions which Kn is higher than 1 ($\sim 100^{0}$ - 10^{∞}) we can not talk about gas conduction between gas molecules. On the other hand, other thermal transfer phenomena can be dominant at that point like convection of gas between walls of porosities of the core material and radiation in gas. The gas can not move freely at that point but some molecules with high density at gas phases can make a capillary movement at walls of porosities. Those have not significance effect on total conduction. Probably that point will be an important parameter for the aging assessment of vacuum insulation panels. Because diffusion of humidity through barrier film is the important phenomenon of the aging of vacuum insulated panel. The other parameter is devacuuminng of panels via gas diffusion through high barrier film. That equation can be describing as a function of properties which is useful for vacuum insulation topics. So if we calculate the mean free path of air from equation (5):

$$\lambda_a = \frac{6.65 \cdot 10^{-3}}{P_a} \tag{5}$$

And we calculate the beta. As Kwon et all. calculate from the thermal flux. [3] So we can describe the thermal conduction of air at room temperature as:

$$k_g = \frac{k_{g,0}}{1 + \frac{0.032}{P_g \ell_s}} \qquad (6)$$

Devacuuming of panel can change inner pressure of the panel. Some material can pass the point Knudsen number is very near to 1. As we look common exercise about thermal conduction of various core materials under vacuum. We can estimate a critical pressure which thermal conduction is dramatically decrease. If we describe that point which the thermal conduction of air in porous media at vacuum pressure is half of free air: as the critical inner pressure of vacuum insulation panel $(P_{\frac{1}{2}})$:

$$k_g = \frac{k_{g,0}}{1 + \frac{0.032}{P_{1/2}^{\ell_S}}}$$

$$1 = \frac{2}{1 + \frac{0.032}{P_{1/2}^{\ell_S}}}$$

then
$$P_{1/2} = \frac{0.032}{\ell_{\rm S}}$$
 (7)

So we can see the critical inner pressure of vacuum insulation panels is related to mean pore size of core material at room temperature. So we can say that for vacuum insulation panel; if the lifespan of vacuum insulation panel described as lower pressures than critical pressure. The lifespan of vacuum insulation panel as a function of core material properties dominantly depends on pore size of core material. When we see the differences thermal conduction measurements at different core materials. The reason of that is solid conduction, radiation or convection properties of material. As a described before if you go lower value than the K_n=1 convection between the gas and a solid walls slightly increases. Actually, the constant β describe this interaction of the gas with and a solid walls. So it is also slightly depended on the material properties. [4]

The general engineering applications can have an equation which assumed from equations 2.6 and 2.5:

$$k_g = \frac{k_{g,0}}{1 + \frac{P_{1/2}}{P_g}} \tag{8}$$

So if we measure a critical pressure from experimental methods; we can use this data to compute all gas conduction values of the core materials at severe pressures where the mean free path of the gas is not much away from Kn=1.

Powder-Glass Fiber Hybrid Core Material

Even a glass fiber core material has good conduction values at the fresh days of the panel which the internal pressure of the panel is low. Glass fiber based core materials have good insulation performance (~5 mWm⁻¹K⁻¹ at ~10Nm⁻²) as seen as at the experimental data. [5] Despite its low critical inner pressure where the insulation performance sharply decreases, with using some desiccator which can absorb water and some organic gasses and getter which can absorb oxygen and nitrogen the lifespan of the panel can be increased to fifteen years. [6] Moreover, type of laminated barrier film and panel size also affect the service life of panels. [7] There are many recent works about increasing barrier properties of polymer films via increasing tortuosity with organo-clay addition to polymer matrix such as montmorillonite and also sepiolite. [8-11] On the other side of development long life-time glass fiber vacuum insulation panels, some core material improvements can be investigated. Filling larger porosities with smaller minerals which they have also good insulation performances because of their properties like inner porosity, fiber-like structure etc. Moreover, using locally occur silicate based minerals can help to invent new core materials with low cost. [12,13,14,15]

MATERIALS AND METHOD

Sepiolite sample from Turkey Eskişehir region is used. This sample has high content on sepiolite about to %85 and other purities are silica and dolomite. Secondary electron images at Figure-1 of this sample show us its fiber like structure at nano size and those fibers are agglomerated like nano size rods and bundles.

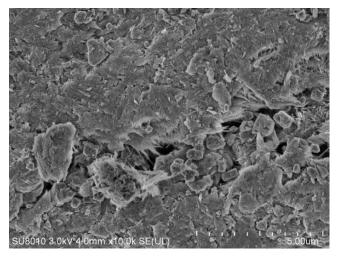


Figure 1. Secondary electron image of sepiolite at 5 μm

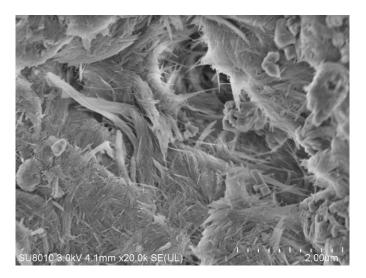


Figure 2. Secondary electron image of sepiolite at 5 μm

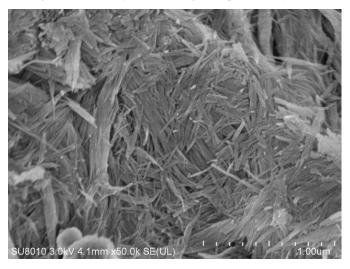


Figure 3. Secondary electron image of sepiolite at 1 μm

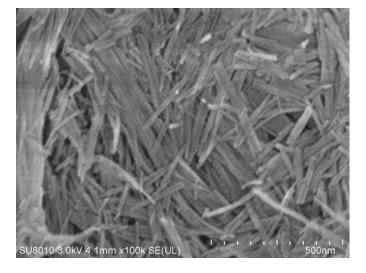


Figure 4. Secondary electron image of sepiolite at 500 nm

And particle size distribution of the raw sepiolite powder as given at Figure-5. Sepiolite sample is used without any treatment. Before application on core material; they are dried at 120-100° C temperature range along several days. [16] Glass fiber nonwovens from Suzhou VIP New Materials Co are used to prepare core material. Powder and glass fiber nonwovens are layered [12] Then they vacuumed inside the CVM-201 Super Bee test kit and thermal conduction tests held with Netzsch HFM 436 with various inner pressures between 1.4 Nm⁻² and 40 Nm⁻² air pressure. This method doesn't give exact value for critical inner pressure but we can estimate from Thermal Conductivity versus inner pressure graphics.

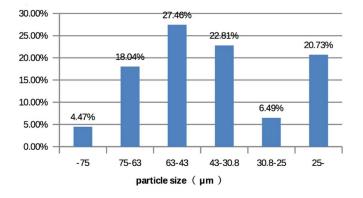


Figure 5. Particle size distribution of Sepiolite sample

RESULTS AND DISCUSSION

As seen at Figure 6 low sepiolite content and without sepiolite addition glass fiber core materials show better thermal insulation performance at high vacuum levels between 1.4 -5 Nm⁻² inner pressure; then its thermal conduction increase sharply with inner pressure. Moreover; when those samples come to about 15 Nm⁻² they start to show more thermal conduction than samples with high sepiolite content. On the other hand, samples with higher sepiolite content just show sharp thermal conduction increase after 20 Nm² inner pressure.

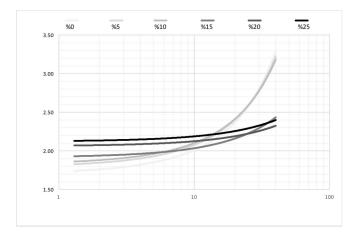


Figure 6. Inner Pressure and thermal conductivity graph of core materials which have various amount of sepiolite

CONCLUSION

Investigation of glass fiber vacuum insulation panels is well justified. As seen from the work, the low sepiolite content and without sepiolite addition glass fiber core materials show better thermal insulation performance. The performance differences on thermal insulation. overall the objectives have been achieved in this study.

If we accept service lifetime of vacuum insulation panel is where the inner pressure between 5-20 Nm² there are no much mean performance differences on initial thermal insulation. Added sepiolite on glass fiber core material create worse initial insulation performance because of increased density of panels but it can be change during the lifespan on panel. The reason of some fails can be abstracted as lack of pre-treatment of sepiolite; doubtfulness on the process of panels in the view of drying and absorbing conditions of sepiolite; using less fibre like sepiolite samples; using the basic processes to create hybrid core material.

For further study can be used some treatment on sepiolite to distribute sepiolite fibres and increase the density of the powder. Therefore, sepiolite can be used in wet-laid processes of nonwovens. Moreover; variation of sepiolite clay should be examined be repeated by same methods

NOMENCLATURE

Cv Specific heat of constant volume, j/kgK

c Speed of the sound, m/sn

c_B Boltzamn constant, m² kg s⁻² K⁻¹

d Diameter of the gas molecule, m

P Pressure, N/ m²

K_g Thermal conductivity of the gas, W/mK

Kn Knuddsen Number

l_s the average wall size of pores, m

T Air temperature, ⁰K

Greek letters

 λ_g The mean free path of gas, m

 β the coefficient of thermal expansion, K⁻¹

 ρ_g Density, kg/m³

REFERENCES

- [1] J.M. Lafferty, Foundations of Vacuum Science and Technology, John Wiley and Sons, New York, 1998.
- [2] G. Reichenauer, U. Heinemann, H.-P. Ebert, Relationship between pore size and the gas pressure dependence of the gaseous thermal conductivity, Colloids Surfaces A Physicochem. Eng. Asp. 300 (2007) 204–210. doi:10.1016/j.colsurfa.2007.01.020.
- [3] J.-S. Kwon, C.H. Jang, H. Jung, T.-H. Song, Effective thermal conductivity of various filling materials for vacuum insulation panels, Int. J. Heat Mass Transf. 52 (2009) 5525–5532. doi:10.1016/j.ijheatmasstransfer.2009.06.029.
- [4] R. Baetens, B.P. Jelle, A. Gustavsen, Aerogel insulation for building applications: A state-of-the-art review, Energy Build. 43 (2011) 761–769. doi:10.1016/j.enbuild.2010.12.012.
- [5] X.B. Di, Y.M. Gao, C.G. Bao, Y.N. Hu, Z.G. Xie, Optimization of glass fiber based core materials for vacuum

- insulation panels with laminated aluminum foils as envelopes, Vacuum. 97 (2013) 55–59. doi:10.1016/j.vacuum.2013.04.005.
- [6] Standard Specification for Vacuum Insulation Panels ASTM C 1484 01, 2014.
- [7] X. Di, Y. Gao, C. Bao, S. Ma, Thermal insulation property and service life of vacuum insulation panels with glass fiber chopped strand as core materials, Energy Build. 73 (2014) 176–183. doi:10.1016/j.enbuild.2014.01.010.
- [8] M.C. Carrera, E. Erdmann, H.A. Destéfanis, Barrier Properties and Structural Study of Nanocomposite of HDPE / Montmorillonite Modified with Polyvinylalcohol, 2013 (2013).
- [9] E. Picard, E. Espuche, R. Fulchiron, Effect of an organo-modified montmorillonite on PLA crystallization and gas barrier properties, Appl. Clay Sci. 53 (2011) 58–65. doi:10.1016/j.clay.2011.04.023.
- [10] Z. Ke, B. Yongping, Improve the gas barrier property of PET film with montmorillonite by in situ interlayer polymerization, Mater. Lett. 59 (2005) 3348–3351. doi:10.1016/j.matlet.2005.05.070.
- [11] E. Ruiz-Hitzky, P. Aranda, A. Álvarez, J. Santarén, A. Esteban-Cubillo, Advanced Materials and New Applications of Sepiolite and Palygorskite, 2011. doi:10.1016/B978-0-444-53607-5.00017-7.
- [12] P. Mukhopadhyaya, K. Kumaran, N. Normandin, Fibre-powder composite as core material for vacuum insulation panel, in: 9th Int. Vac. Insul. Symp., London, 2009.
- [13] P. Mukhopadhyaya, K. Kumaran, N. Normandin, High-Performance Vacuum Insulation Panel: Development of Alternative Core Materials, 22 (2009) 103–123

- [14] Li, C. et al., Thermo-physical properties of polyester fiber reinforced fumed silica hollow glass microsphere composite core and resulted vacuum insulation panel. Energy and Buildings, 2016(125): p. 298-309
- [15] Li, C. et al., Fabrication and characterization of low-cost and green vacuum insulation panles with fumed silica/rice husk ash hybrid core material, Materials & Design, 2016. 107: p. 440-449
- [16] Li, C. et al., The effect of drying condition of glassfibre core material on the thermal conductivity of vacuum insulation panel. Materials & Design, 2013(50): p. 1030-1037
- [16] Li, C. et al., Efect of pressure holding time of extaction process on thermal conductivity of glassfiber VIPs. Journal of Materials Processing Technology, 2014. 214(3): p. 539-543