



Research Paper

Experimental study of the thermal conductivity of polyurethane foams

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HIGHLIGHTS

- Thermal conductivity of PU foam is measured under various environments by TPS method.
- Spectral extinction coefficient of PU foam is measured by FTIR.
- Thermal conductivity of PU foam increases non-monotonically with temperature.
- Thermal conductivity of PU foam increases as high as 10–18% in moist air.
- Radiative thermal conductivity of PU foam can be calculated by Rosseland model.

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ABSTRACT

Polyurethane foams are widely used in energy conservation field and thermal conductivity is one of the most important properties. To reveal and optimize the thermal insulation performance of PU foams, the thermal conductivity of five PU foam samples formed by blowing agents of CP, CP + IP, CP + 245fa and CP + 245fa + LBA are measured using transient plane source method under various environment. Influences of temperature, humidity, water uptake, alternate high and low temperature, long time storage at high temperature and gas pressure of the atmosphere on the thermal conductivity of PU forms are investigated comprehensively. The mechanism of temperature that affects the thermal conductivity of PU foams is discussed. Fourier transform infrared spectroscopy is adopted to measure the spectral extinction coefficients of these five samples. With the spectral extinction coefficient, the radiative thermal conductivity is calculated from Rosseland model. Then the contributions of radiative heat transfer to the effective thermal conductivity are decomposed. The thermal conductivity of five foams increases non-monotonically with temperature. When stored in moist air, thermal conductivity can increase as high as 10–18%. Radiative thermal conductivity contributes 3.6–4.1% at -40°C , 7.3–9.0% at 20°C and 9.1–11.8% at 70°C to the effective thermal conductivity.

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1. Introduction

Thermal insulation technique and thermal insulation materials have wide application scopes such as thermal insulation of building, food cold storage, refrigerator, petroleum pipe, transportation of liquid natural gas [1,2]. Refrigerators are one of the most energy consuming applications at home. In the last decades, the number of refrigerator has increased dramatically and the production of refrigerator of China in 2013 is more than 90 million [3–5]. Consequently, it desired to make the refrigerators more energy efficient by searching thermal insulation materials with higher performance. Polymeric foams are one of the most efficient thermal

insulation materials because blowing agent gas with extremely low thermal conductivity is trapped in the closed porous structures. Polyurethane (PU) foams are prepared by polymerization of an isocyanate reacting with a polyol and the mixture is foamed using one or more blowing agents. Thermal conductivity is one of the most important properties of PU foam. Typical thermal conductivity values for PU foams are between 0.02 and 0.03 W/m·K [2,6].

The thermal property of blowing agent is the key issue of reducing the thermal conductivity of PU foam. The ideal blowing agent should have low gaseous thermal conductivity and good chemical stability but be not inflammable and explosive, not hazard to environment and humans. Recently, Honeywell International Inc. developed solstice liquid blowing agent (LBA) because it has no ozone depletion and has very small GWP (less than 5) [1,6]. CP, isopentane (IP) and their mixtures are the most widely used

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flowing agents in refrigerator at present. Recently, R245fa, 365mfc, R134a, LBA and their multivariate mixtures are the most probable substitutes of current blowing agent and have been applied to some latest refrigerators.

A number of studies have been focused on predicting and optimizing the thermal insulation capacity of foaming materials in the last decades. The advantages and disadvantages of different insulation materials have been discussed [2,7,8]. Harikrishnan et al. [9] introduced a transient needle probe method to measure thermal conductivity of rigid PU foams. Pau et al. [10] measured the thermal conductivity and specific heat capacity of seven PU foams and their melts. Fan et al. [11] studied the physical properties of rigid PU foams with soy-phosphate polyol and water as the blowing agent. Song et al. [12] used hot wire method to study the influences of density and temperature on the thermal conductivity of rigid PU foam plastic and polystyrene foamed plastic and found that the thermal conductivity decreases first and then increases with the increase of density and increases with temperature. Caps et al. [13] studied the thermal conductivity of polyimide foams in the temperature range from $-100\text{ }^{\circ}\text{C}$ to $50\text{ }^{\circ}\text{C}$ for different gas types at different gas pressure and established a quantitative model to predict the thermal conductivity as a function of density, gas pressure and temperature. Ankang and Houde et al. [14] predicted the thermal conductivity of open cell PU foam based on fractal theory. Effective cell models can be used to predict the thermal conductivity of insulating foams [15]. Coquard and Baillis [16] modeled the coupling conductive and radiative heat transfer in low density expanded polystyrene. Wu and Chu [17] systematic investigated the coupled conductive and non-gray radiative heat transfer of open cell PU foam. Kaemmerlen et al. [18] revealed the radiative properties of extruded polystyrene foams using predictive model and experiment test. Tseng et al. [19] studied the thermal conductivity of PU foams from room temperature to $-253\text{ }^{\circ}\text{C}$ and found the gas conduction in the closed cells accounts for approximately 60–80% of the total heat transfer while the radiative transfer is not important in low temperature region but it accounts for 10–20% at room temperature. The thermal conductivity of open PU foams with cell sizes ranging from $150\text{ }\mu\text{m}$ to $350\text{ }\mu\text{m}$ was investigated theoretically and experimentally and found that the thermal conductivity increases with cell size and the gaseous conduction accounts for 70–80% of the total thermal conductivity from the measurement at different gas pressure [6]. The thermal conductivity of PU foam cell gases n-pentane, isopentane and nitrogen was measured at temperatures between $36\text{ }^{\circ}\text{C}$ and $141\text{ }^{\circ}\text{C}$ and at gas pressures from constant pressure to 1.0 MPa and the gases conduction account for 60–65% of the total heat transfer [1]. However, the gaseous conduction to the total thermal conductivity will vary with time because the blowing agent and other gases within the closed foams will diffuses out with time while the air will diffuses into the closed cells. Thus the Oak Ridge National Laboratory carried out years of studies of the aging of PU foam insulation in simulated refrigerator panels [20–22]. Arduini-Schuster et al. [23] investigated the infrared optical properties of closed micro-celled extruded polystyrene foam and PU foam via experimental test and theoretical modeling. Effective thermal conductivity calculated from theoretical model was also compared with measured results.

The thermal insulation capacity of foam materials in refrigerator will be not only influenced by constitues but also depends on the environmental factors such as temperature and humidity. In addition, the gas diffusion through the foam struts will result in the thermal insulation performance degradation with time and the aging will be affected by the operating environment. Radiation within PU foam relies on its extinction coefficient. According to the best of our knowledge, there are few studies focus on the systematically influences of humidity, temperature and blowing agent

and radiative property to the thermal insulation of foams. In this work, the thermal conductivity of five PU foams foamed with different blowing agents is measured using transient plane source (TPS) method at different temperature and humidity. The influences of storage environment with alternate change of temperature and long time high temperature on the thermal conductivity of PU foams are also investigated. Extinction coefficients of five PUs foams are measured by Fourier transform infrared spectroscopy (FTIR) to reveal their radiative thermal conductivity.

2. Experimental measurement

2.1. Thermal conductivity measurement

Steady state method will be questionable for the thermal conductivity measurement of moist materials because more than $10\text{ }^{\circ}\text{C}$ temperature difference across the tested samples will affect the moisture distribution in the porous materials. In addition, the large temperature difference across the samples makes it difficult to distinguish the thermal conductivity at certain temperature when the thermal conductivity is very sensitive with temperature. To study the thermal conductivity of PU foams at different temperature and humidity, the measurement is conducted by the TPS 2500S thermal constant analyzer (Hot Disk AB, Sweden) which is based on the TPS method. Compared with steady state method, this method not only could overcome the above problems but also requires very short test time and small sample size as well. TPS method has been widely used to measure the thermal conductivity of high thermal insulation materials [10,24–28]. TPS method is developed by Gustafsson et al. [29] and later the mathematical derivation was explained in detail by He [30]. When performing the test, a double spiral sensor is placed between two pieces of plain samples like a sandwiched structure [31]. In the test, the sensor is used both as a heater and as a temperature sensor. The test theory of TPS method is presented in detail in literatures [29–31].

The TPS apparatus is validated using NIST1453 (an expanded polystyrene board) with thermal conductivity of $0.032\text{ W/m}\cdot\text{K}$ at room temperature and the deviation is within 3%. Thus the nominal accuracy of thermal conductivity measured by TPS method is 3%. The uncertainty brought by the theoretical assumptions is estimated less than 3% for materials with thermal conductivity $\sim 0.03\text{ W/m}\cdot\text{K}$ [31]. The uncertainty brought by regarding the thermal radiation within the porous materials with extinction coefficient higher than 2000 ($1/\text{m}$) as a diffusion process in the TPS theory is less than 0.3% [32]. The uncertainty introduced by input parameters determination is evaluated at 3.6% for extended TPS technique [33]. In addition, the thermal conductivity of PU foams measured by TPS method is about 10% higher than that measured by 1D state steady method. Therefore, the uncertainty of thermal conductivity of PU foams measured by TPS method is estimated as much as 10%.

2.2. Radiative property measurement

2.2.1. Transmittance and extinction coefficient

FTIR spectroscopy is adopted to measure the normal transmittance of PU foam samples to obtain their spectral extinction coefficient through Lambert Beer's law [23,34,35]:

$$\tau_{\lambda} = \frac{I_{\lambda}(L)}{I_{\lambda}(0)} = \exp(-k_{\lambda}L) \quad (1)$$

Here τ_{λ} is transmittance and identical to the ratio of the collimated radiation intensity ($I_{\lambda}(L)$, W/m^2) transmitted through test material with thickness of L (m) to the intensity of radiation ($I_{\lambda}(0)$) that initially enters the material at wavelength of λ (μm). k_{λ} is spectral

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