



Original article

Evaluation of benzenesulfonyl hydrazide concentration on mechanical properties, swelling and thermal conductivity of thermal insulation from natural rubber



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ARTICLE INFO

Article history:

Received 19 June 2015

Accepted 4 January 2016

Available online 25 June 2016

Keywords:

Benzenesulfonyl hydrazide

Mechanical properties

Natural rubber

Thermal conductivity

Thermal insulation

ABSTRACT

An experiment was carried out to obtain the optimum natural rubber thermal insulation for a refrigeration and air conditioning system. Thermal insulation not only adds value to the rubber, but also develops an improved new product from natural rubber. Concentrations of benzenesulfonyl hydrazide (BSH) blowing agent at 2, 4, 6, 8 and 10 parts per hundred of rubber (phr) were used to study the effect of the BSH concentration on the mechanical properties, swelling and thermal conductivity of thermal insulation which were evaluated using the American Society for Testing and Materials (ASTM) standards (ASTM D412, ASTM D471 and ASTM C518, respectively). The rubber compound was prepared using a two-roll mill and expanding in a hot mould at 150 °C. The results showed that both the average values of the tensile modulus and tensile strength were reduced by 25%. The average value of elongation at break not only increased by 33% before thermal aging, but also reduced by 33% after thermal aging. The maximum value of swelling was 324%. Moreover, the lowest value of thermal conductivity was 0.040 W/m/K. Therefore, the optimum concentration of the BSH was 6 phr which had optimum properties for thermal insulation.

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Introduction

In general, the thermal insulation materials used in a refrigeration system or an air conditioning system can be made from metals, polymers and composites such as aluminum foil, polyethylene foam and fiberglass according to Al-Homoud (2005). He discussed other desirable features and noted that materials should possess the ability to limit heat gain or loss from surfaces operating at temperatures above or below ambient temperature. He defined thermal conductivity as a measure of the effectiveness of a material in conducting heat. Therefore, knowledge of the thermal conductivity values allows a quantitative comparison to be made between the effectiveness of different thermal insulation materials. For example, the thermal conductivity values of rock wool, fiberglass and polyethylene are 0.037 W/m/K, 0.040 W/m/K and 0.041 W/m/K, respectively (Al-Homoud, 2005). In particular, an important role of thermal insulation is to maintain the temperature and to increase the energy efficiency of refrigeration systems or air

conditioning systems (Leon and Ellann, 2012). At present, developing and improving new products from natural rubber is an important alternative not only for adding value to natural rubber but also for increasing the efficiency of new products, such as a natural rubber nanocomposite of multi-walled carbon nanotubes (Takeuchi et al., 2015), the application of natural and synthetic rubber/waste in the footwear industry (Diana et al., 2015), natural rubber Ca/P blends for biomedical uses (Nascimento et al., 2014), and especially natural rubber for thermal insulation (Abdel-Kader et al., 2012). In polymer-based thermal insulation, chemical blowing agents are required to produce rubber foam with low thermal conductivity and the decomposition rate of the blowing agent is the critical parameter for selecting the optimum blowing agent and furthermore, the amount of blowing agent, the heating time and the processing temperature play important roles in determining the cell morphology (Guobin et al., 1998). The chemical blowing agent influenced the structure and mechanical properties of ethylene propylene diene monomer (EPDM) foam by increasing the number of cell structures, increasing the porosity, lowering the thermal conductivity and increasing the concentration of the blowing agent for optimum interfacial adhesion (Yamsaengsung and Sombatsompop, 2009). An investigation of the

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mechanical response and stability of closed-cell foams found that the presence of internal pore pressure significantly stiffens and stabilizes the response of elastomeric foams (Oscar et al., 2012). Nabil et al. (2014) reported the effect of the accelerators and vulcanizing system on the thermal stability of natural rubber/recycled EPDM blends with four types of accelerators: *n*-tert-butyl-2-benzothiazylsulphonamide (TBBS), *n*-cyclohexylbenzothiazylsulphenamide (CBS), disulphide tetramethylthiuram (TMTD) and mercaptobenzothiazol (MBT). They found that CBS-vulcanized blends exhibited satisfactory overall mechanical and thermal stability compared to the other accelerators used. Moreover, the optimal vulcanizing system involved semi-efficient vulcanization which had the highest tensile strength compared with the other vulcanizing systems.

This article considers part of the development of a thermal insulation system using natural rubber air-dried sheet (ADS) by focusing on improvement of the thermal insulation properties of the natural rubber foam. The natural rubber compounds were cured using a conventional vulcanization system. Natural rubber foam was processed into thermal insulation using the BSH. The effects were investigated of the BSH blowing agent and its content on the tensile modulus, tensile strength, elongation at break, compression set and swelling of the natural rubber foam. Moreover, the thermal conductivity of the natural rubber foam was also studied.

Materials and methods

Raw materials additives for the natural rubber

The materials used for the preparation of the compounds were natural rubber and chemical agents. Natural rubber air dried sheets (ADS) were supplied from plantations. The characteristics of the natural rubber ADS were investigated by the Rubber Research Institute (Bangkok, Thailand). The physical properties of the ADS (dirt content, volatile matter, ash content, nitrogen content, initial plasticity, plasticity retention index and Mooney viscosity) were determined using *Standard Malaysian Rubber (1992)* as shown in Table 1.

The additive essential for the natural rubber foam was the benzenesulfonyl hydrazide (BSH) blowing agent. The molecular formula of the BSH blowing agent is $C_6H_8N_2O_2S$. It was used to change the cell structure of natural rubber in this work. The decomposition temperature of the BSH blowing agent was 105 °C at 120 cm³/g gas evolutions. The loading of BSH was varied from 2 parts per hundred of rubber (phr) to 10 phr as shown in Table 2. The BSH blowing agent and flame retardant (antimony trioxide) were supplied by Kij Paiboon Chemical Limited Partnership (Bangkok, Thailand). The other additives consisted of stearic acid, zinc oxide, TMTD, TBBS, sulfur, polydicyclopentadiene-co-p-cresol (wingstay L) and paraffin oil and were supplied by the Rubber Research Institute (Bangkok, Thailand). The chemical structure of the natural rubber and the BSH blowing agent are shown in Fig. 1.

Table 1
Characteristics of air dry sheet natural rubber.

Parameter	Value
Dirt content (%weight basis)	0.027
Volatile matter (%weight basis)	0.830
Ash content (%weight basis)	0.160
Nitrogen content (%weight basis)	0.350
Initial plasticity	46.200
Plasticity retention index	66.500
Mooney viscosity	78.000

Table 2
Compound formulation of natural rubber used in this study.

Ingredient	Contents (parts per hundred of rubber)				
	100	100	100	100	100
Natural rubber	100	100	100	100	100
Zinc oxide	5	5	5	5	5
Stearic acid	2	2	2	2	2
Wingstay L	4	4	4	4	4
Paraffin oil	10	10	10	10	10
Antimony trioxide	20	20	20	20	20
BSH	2	4	6	8	10
TMTD	0.2	0.2	0.2	0.2	0.2
TBBS	1	1	1	1	1
Sulfur	2	2	2	2	2

Compounding and vulcanization process

The mechanical mixture was prepared to attain a homogeneous mixture of the rubber compound in three stages. In the first stage, the natural rubber ADS and plasticizer were masticated in a two-roll-mill mixer (Model R11-3FF; Kodaira Seisakusho; Bunkyo-Ku, Japan) at a temperature of 30 °C for 30 min (Somjate et al., 2014). The Mooney viscosity of the rubber compounds was determined using a Mooney viscometer in compliance with Spanish Association for Standardization and Certification (AENOR, 2007). In the second stage, the rubber compound obtained in the first stage was mixed with various chemicals at 70 °C for about 15 min to attain a homogeneous mixture of the rubber compound, according to the formulations for mixing shown in Table 2. In the final stage, the rubber compounds were then left 24 hr prior to testing for curing characteristics. Each batch of rubber compound was rolled to produce a sheet of approximately 5 mm thickness. Cure characteristics were analyzed using a moving die rheometer for 30 min at 150 °C according to ASTM International (2007). In the vulcanization process, all rubber compounds were expanded in a hot mould at 150 °C according to their respective cure time (t_{90}) which was the time for the completion of cure determined with a moving die rheometer (Arayaprane and Rempel, 2007).

Measurement of mechanical properties

In this work, the mechanical properties, consisting of the tensile modulus, tensile strength and elongation at break, were measured following the standard test methods for vulcanized rubber and thermoplastic elastomers—tension (ASTM International, 2013). Dumbbell samples were prepared with dimensions 115 mm × 25 mm × 2 mm. The testing speed used was 500 mm/min at room temperature. The average value of five tests for each sample was reported. Therefore, in total, 50 specimens were tested for their mechanical properties (tensile modulus, tensile strength and elongation at break). The testing of compression under a constant deflection in air followed ASTM International (2014). The sample was in the form of a cylindrical disk with a diameter of 30 mm and thickness of 13 mm. The thickness of the space bar was 9.5 mm. The temperature and testing time were conditioned in an oven at 100 °C for 22 hr and then removed from the fixture and

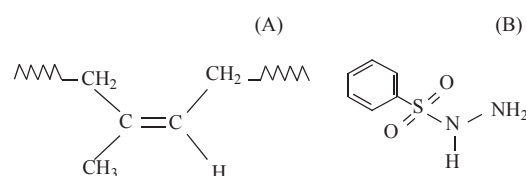


Fig. 1. Chemical structure: (A) natural rubber; (B) benzenesulfonyl hydrazide.

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