Effect of Type and Content of Blowing Agent on Properties of NR/EPDM/EVA Foam

B Kim¹,⁴, P Nun-anan², K Hancharoen³, K Seiichi⁵, K Boonkerd¹,⁴*  
¹ Department of Materials Science, Chulalongkorn University, Bangkok, Thailand 10330  
² Center of Excellence on Petrochemical and Materials Technology, Faculty of Science, Chulalongkorn University, Bangkok, Thailand 10330  
³ Center of Building Innovation Technology, Department of Building Innovation, Faculty of Architecture, Kasetsart University, Bangkok, Thailand 10900  
⁴ Department of Materials Science and Technology, Nagaoka University of Technology, Niigata-ken, Japan 940-2188  
⁵ Green Materials for Industrial Application Research Unit, Faculty of Science, Chulalongkorn University, Bangkok, Thailand 10330  
E-mail: K.Boonkerd@gmail.com

Abstract. This research aimed to study the influence of type and content of blowing agents on the properties of rubber foam NR/EPDM/EVA. Supercell DP, supercell RC 720, and EW5 were used here as a blowing agent. The content of the blowing agent was varied from 3 phr to 4 and 5 phr. The results showed that supercell RC 720 and OBSH were not suitable for this trinary rubber foam due to the absence of even porous structure, while the supercell DP gave the even porous structure throughout the sample and the lowest density. It was found that increasing supercell DP loading led to the increase in pore size, thermal conductivity but the decrease in density, tensile strength, and elastic recovery of rubber foam. Owing to the lowest thermal conductivity, it can be inferred here that supercell DP at 5 phr was suitable to produce rubber foam from NR blended with EPDM and EVA for a ceiling board application.

1. Introduction  
Nowadays, global warming leads to an increasing in the atmosphere temperature every year. The ceiling board is used to reduce the heat transfer from the outside. The insulator should have some specific properties such as lightweight, dimensional stability, low thermal conductivity, and ozone resistance. The one with those excellent performances is rubber foam [1]. Rubber foam is well known as a ceiling board owing to its low thermal conductivity. Mostly, rubber foam is produced from synthetic polymer, in turn, they are high cost and non-renewable materials [2]. Natural rubber (NR) is known as an environmental friendly material and assures excellent mechanical properties [3-4]. According to the world's top in 2019, Thailand was the top country which exported NR about 31.5% of NR consumption in the world. To promote the usage of NR, this research is aimed to develop NR-based foam for use as a thermal insulator board. However, NR has low weathering and solvent resistance due to the presence of a double bond and consisting only of hydrocarbon, respectively [5]. To overcome these disadvantages and reach some specific requirements, many researches have been conducted by blending NR with other specialty rubber such as ethylene propylene diene monomer rubber (EPDM), butadiene rubber (BR). EPDM is known for good aging resistance to weather materials including oxygen, ozone, UV, water, and heat, however, it has fair tensile strength [6]. Therefore, blending NR with EPDM rubber gain many interests from researchers owing to the combination of their beneficial properties [6-9]. On contrary,
some researchers found that the blend showed phase separate, and cure-rate mismatching because of the amount of diene, polarity, and solubility [7-8,10-11]. Moreover, from our preliminary study, rubber foam from NR blended with EPDM showed considerable shrinkage, thus having a high density which was improper for using as a thermal insulator. Therefore, ethylene vinyl acrylate (EVA) known as rubber-like material and has high dimensional stability, UV resistance, and aging ability is also used here [12-13]. In the previous work, it had been reported that the type and content of the blowing agent also played an important role in the density, structure, and properties of foam [14-17]. In addition, Charoeythornkhajornchai et al., noticed that the increasing of blowing agent improved cure rate and thermal expansion coefficient [18]. Furthermore, the effect of type of blowing agents including 4,4-oxybis-(benzenesulfonyl hydrazide) (OBSH) and Azodicarbonamide (ADC) on the expanded natural rubber was investigated. The results showed that OBSH was more effective for forming natural rubber foam than ADC. With the presence of OBSH, it was observed that foam density and crosslink density decrease with the increase of blowing agents [14].

NR/EPDM/EVA blends are expected to give low density, dimensional stability, good mechanical properties, UV and ozone resistance. There were many studies between blends of two or more than two elastomers, however, the blending of NR/EPDM/EVA has not been widely implemented. Therefore, in this work, rubber foam was prepared from the blending NR/EPDM/EVA for applying as a ceiling board insulator. For this report, the effects of blowing agents including Dinitrosopentamethylenetramine (supercell DP), modified Azodicarbonamide (supercell RC270), and 4,4-oxybis-(benzenesulfonyl hydrazide) (EW5) at various contents from 3 to 5 phr on the properties of blend foam were evaluated. The properties of the obtained foam including density, hardness compression set, tensile strength, thermal conductivity, and optical microscope of the obtained rubber foam were reported.

2. Experimental

2.1. Materials, Compounding, and Vulcanization

The compound formulation used in this work was given in Table 1. Natural rubber (STR 5L, Rubber Authority of Thailand) was masticated for 5 min in internal mixer (Chareontut, MX500-D75L90) at 50°C. EPDM rubber (ENB 3.9w%, Ethylene 56.7w%, ExxonMobil Chemical Asia Pacific), EVA (N8038 vinyl 18w%, A.F. Supercell Co., Ltd), CaCO₃, Wing stay L, and paraffin oil were added, respectively in total mixing time 14 min. The compound was cooling down to room temperature, then was mixed with crosslink agent and blowing agent such as supercell DP, supercell RC720, and EW5 on two roll mills (Labtech, LRM-110) within 16 min at 60°C. Vulcanization stages were carried out in hydraulic compression (Labtech, LP-5-50) at 160°C for 7 min and the foam occurred freely after removing hydraulic pressure.

| Ingredients                  | F1 | F2 | F3 | F4 | F5 |
|------------------------------|----|----|----|----|----|
| Natural Rubber (NR)          | 50 | 50 | 50 | 50 | 50 |
| EPDM                        | 10 | 10 | 10 | 10 | 10 |
| EVA                         | 40 | 40 | 40 | 40 | 40 |
| Dicumyl Peroxide (DCP)      | 0.7| 0.7| 0.7| 0.7| 0.7|
| Paraffin Oil                | 10 | 10 | 10 | 10 | 10 |
| Wing Stay L                 | 1  | 1  | 1  | 1  | 1  |
| CaCO₃                       | 30 | 30 | 30 | 30 | 30 |
| Supercell DP                | -  | -  | 3  | 4  | 5  |
| EW5                         | 4  | 4  | -  | -  | -  |
| Supercell RC720             | -  | 4  | -  | -  | -  |
2.2. Characterization

The density of rubber foam was calculated by dividing the weight of the sample by the size of the sample. The hardness (shore OO) of rubber foam was determined according to ASTM D2240. For each sample, an average of eight points was reported. The compression set was tested based on ASTM D395 (method B) at 70°C for 22 h. Tensile strength was followed by ASTM D412 with a dumbbell-shaped specimen using a universal testing machine (Tinius Olsen, 5ST). The averages strength of seven specimens was reported. Thermal conductivity (EKO Instrument, HC740, Japan) was tested according to ASTM C518 with temperature 50°C to 20°C control by cooling bath. The characteristics of cell size were elucidated using an optical microscope. The picture was taken at 100x magnification.

3. Results and Discussion

Figure 1 showed the surface of rubber foam when using EW5, RC720 and DP at 4 phr. At the same moulding condition, it was found that the foams prepared by EW5 and RC720 had brownish and rough surface. On the other hand, the foam prepared from DP showed smooth surface. In addition, the cross-sectional morphology of rubber foam prepared from EW5 as shown in Figure 2(F1) indicated that the cell structure was hardly formed, while that of the one prepared from RC720 shown in Figure 2(F2) showed the nonuniform distribution of cell structure with the presence of solid region. However, the rubber foam with the uniform distribution of cell structure could be obtained when using DP as a blowing agent. Figure 2(F4) showed that the rubber foam prepared from DP had the cell size ranging from 0.284 mm to 1.266 mm.

Figure 1. Images of rubber foam surface when using EW5 (F1), RC720 (F2), and DP (F4) at 4 phr

Figure 2. Cross-sectional morphology of rubber foam at 100 magnification when using EW5 (F1), RC720 (F2), and DP (F4) at 4 phr
Considering at the same content of blowing agent, the density as depicted in Figure 3, and the hardness as depicted in Figure 4 of the rubber foam prepared with OBSH and RC 720 were significantly higher.
than the rubber foam prepared with DP. As the aim of this work, the rubber foam will be applied as a ceiling board insulator at which the material with the low density is needed. The suitable blowing agent for this trinary rubber blend was voted to DP. Furthermore, the cell size of rubber foams was slightly increased with the increase of the blowing agent content as shown in Figure 5 indicating that the different amounts of gas were produced during decomposition of DP. Additionally, it was clearly seen that the density of rubber foam was decreased as shown in Figure 6. It was noted that the increase of blowing agent content led to more gas trapped inside the matrix, thus making the foams in having a diverse expansion ratio, as the result, it slightly decreased in hardness (Figure 7). Besides, tensile strength and compression sets of rubber foam with varying content of DP at 3, 4, and 5phr were shown in Figure 8 and 9, respectively. The results showed that the tensile strength and elastic recovery decreased with increasing blowing agent content. It indicated that increasing blowing agent content resulting in more expansion and less materials in a given volume causes the foam with inferior tensile strength and less recovery ability. Moreover, it was observed the thermal conductivity was declined with increases in the blowing agent content. This can be inferred that the rubber foam with the large cell size can perform as an insulator much better than the one with small cell size.

4. Conclusion
To prepare trinary rubber foam from NR blended with EPDM and EVA, the RC720 and EW5 were not suitable blowing agent. The nonuniform distribution of cell structure was obtained when using the above blowing agents. DP was found to be the suitable blowing agent for this trinary rubber foam. It gave the rubber foam the uniform cell structure. The thermal insulating performance was increased with increase in the blowing agent content. However, the opposite effect on the tensile strength and elastic recovery of the rubber foam was observed.
5. Acknowledge
The scholarship has been provided by GAICCE DDP Scholarship from ASEAN University Network/Southeast Asia Engineering Education Development Network (AUN/SEED-net).

6. References
[1] Najib N N, Ariff Z M, Manan N A, Bakar A A and Sipaut C S Effect of blowing agent concentration on cell morphology and impact properties of natural rubber foam 2009 J. Phys. Sci 20 13–25
[2] Puşcă A, Bobancu Ş and Duţă A Mechanical properties of rubber 2010 Bulletin of the Transilvania Uni. of Braşov 3 107–14
[3] Wang N, Hu L, Babu H V, Zhang J and Fang Q Effect of tea saponin-based intumescent flame retardant on thermal stability, mechanical property and flame retardancy of natural rubber composites 2017 J. Ther. Ana. and Cal. 128 1133–42
[4] Jame M E, Roland C M and Erman B 2013 The Science and Technology of Rubber vol 4 (Amsterdam:Elsevier) p 77
[5] Board D 2007 Kirk Othmer Encyclopedia of Chemcail technology (Amsterdam: John Wilet & Sons) p 852-80
[6] Rattanasupa B and Keawwattana W Development of rubber compound based on natural rubber (NR) and ethylene-propylene-diene-monomer (EPDM) rubber for playground rubber Mat. 2007 Kasetsart J.(Nat.Sci) 247 239–47
[7] Chaisuriyathepkul A, Suchiva K and Sae-ou P Effect of mixing conditions on phase morphology of NR/EPDM blends 2013 Adv. Mat. Research 747 467–70
[8] Ghosh A K, Debnath S C, Naskar N and Basu D K NR–EPDM Covulcanization:A Novel Approach 2001 J. Appl. Pol. Sci. 81 800–08
[9] Chandrasekar V C 2010 Rubber as a construction materials chapter 1 p 1–6
[10] Arayaprance W and Rempel G L Properties of NR/EPDM blends with or without methyl methacrylate-butadiene-styrene (MBS) as a compatibilizer 2007 Int. J. Mat Struc. Reli. 5 1–12
[11] Chang Y, Shin Y, Chun H and Nah C Effects of trans-Polyoctylene Rubber (TOR) on the properties of NR/EPDM blends 1998 J. Appl. Pol. Sci. 73 749–56
[12] Kaewsakul W, Kaesaman A and Nakason C Dual phase continuity and phase inversion phenomena in natural rubber/ethylene vinyl acetate (EVA) copolymer blends 2012 e-Pol. 1–13
[13] Miedzianowska J, Masłowski M and Strzelec K Thermoplastic elastomeric composites filled with lignocellulose bioadditives part 1: Morphology, processing, thermal and rheological properties 2020 Materials 13 1598-618
[14] Sombatsompop N and Lertkamolsin P Effects of chemical blowing agents on swelling properties of expanded elastomers 2000 J. Elas. Pla. 32 311–28
[15] Lewis C, Rodlum Y, Misaen B, Changchum S and Sims G L A Effect of compound formulation and processing conditions on properties of extruded EPDM and NR/EPDM foams 2003 Cell. Pol. 22 43–56
[16] Wang B Q, Peng Z L, Zhang Y and Zhang Y X Study on foaming kinetics and preparation of EPDM foams 2006 Plas. Rub. Composit 35 361-67
[17] Kim M, Park C, Chowdhury S R and Kim G Physical properties of ethylene vinyl acetate copolymer (EVA)/natural rubber (NR) blend based foam 2004 J. Appl. Pol. Sci. 94 2212–16
[18] Charoeythornkhajhornchai P, Samthong C, Boonkerd K and Somwangthanaroj A Effect of azodicarbonamide on microstructure, cure kinetics and physical properties of natural rubber foam 2017 J. Cell. Plas. 53 287–303