*Advanced Materials Research Vols. 47-50 (2008) pp 181-184*
*online at http://www.scientific.net*


# Cellular Morphology, Peel Strength and Thermal Conductivity for wood/NR and Expanded EPDM Laminates for Roofing Applications

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**Keywords:** Natural rubber, Laminates, Composites, Blowing agent, Fiber, Adhesion, Roofs

**Abstract.** Melt-laminates of wood/NR and expanded ethylene-propylene-diene-rubber (EPDM) were prepared for rubber composite roofs. The ethylene-propylene bound OBSH (EPR-*b*-OBSH) blowing agent was introduced as an effective blowing agent in the EPDM layer and the foam property results were compared with the pure OBSH blowing agent. It was found that the EPR-*b*-OBSH gave expanded EPDM layer with a larger number of more uniform cells. The optimum concentration for EPR-*b*-OBSH blowing agent for achieving the maximum peel strength and for lowering the thermal conductivity of the wood/NR-*e*EPDM laminate was 3.0phr.

## Introduction

Roofs made from polymer materials have also been attention recently because of easy process-abilities, good mechanical properties, favorable environmental profiles and cost savings. Some polymer composite roofs are produced in forms of sandwich structure with foamed or expanded polymers cores, which have low density, low thermal conductivity property, and reduce the air temperature and the energy demand for air conditioning system. Most recent work by Yamsaengsung and Sombatsompop [1] developed a fully bonded ply roof from a hard-surface NR roof through ebonite-NR vulcanizates incorporated with wood sawdust particles. The ebonite wood sawdust/NR roofs were then UV-protected by melt-laminating with EPDM layer. The experimental results suggested that the interfacial strength and mechanical properties of the wood/NR-EPDM roofs were greatly improved by a co-crosslinking reaction at the interface through the use of N-(β- Bis-(3-triethoxylpropyl) tetrasulfan (Si69) as a chemical coupling agent within the wood/NR layer. The main goal of this work was to improve the thermal insulation properties of the wood sawdust/NR-EDPM laminating roofs by foaming the EPDM layer through the use of two different forms (pure and modified) of 4,4′oxybis(benzenesulfonylhydrazide) (OBSH). The physical foam characteristics of the expanded EPDM (*e*EPDM) layer, the peel strength and thermal conductivity of the wood/NR-*e*EPDM laminates were studied.

## Experimental

***NR and compounding ingredients:*** The natural rubber (NR-STR5L) was supplied by Siam United Rubber Co., Ltd (Bangkok, Thailand). The natural rubber formulations were 100 phr rubber, 33.4phr zinc oxide (ZnO), 13.4phr stearic acid, 3.4phr mercaptobenzothiazole (MBT), 1.4phr diphenylguanidine (DPG), 0.1phr UV stabilizer, 0.1phr anti-oxidant and 20.0phr sulfur.
***Wood sawdust particles:*** Wood particles were obtained from carpentry and wood-working processes, which supplied by V.P. Wood Co., Ltd. (Bangkok Thailand) and fixed at 40phr.
**Precipitated silica:** The commercial silica used was precipitated amorphous white silica-type Ultrasil VN3 (PSi). The silica content used in this work was fixed at 45phr.
**Chemical coupling agents:** N-(β- Bis-(3-triethoxylpropyl) tetrasulfan (Si69) was used as the coupling agent which were supplied by Shin Etsu Chemical Co., Ltd. (Tokyo, Japan).

**EPDM rubber and compounding ingredients:** EPDM (ESPRENE 505) based on 5-ethylidene-2-norbornene with 49.8 % ethylene and 10.0% diene, was supplied by Siam United Rubber Co., Ltd (Bangkok, Thailand). The formulations were 100phr rubber, 5.0phr ZnO, 1.5phr stearic acid, 0.6phr MBTS, 0.6phr DPG and 2.5phr sulfur.

**Chemical blowing agents:** Two forms of 4,4′ oxybis(benzenesulfonylhydrazide) or pure OBSH and modified OBSH (EPR-*b*-OBSH) were used. The physical and thermal characteristics of both are given in Table 1. The content of the OBSH were varied from 0.0 to 11.0phr. Since the EPR-*b*-OBSH blowing agent had 25% lower OBSH content than pure OBSH, the content of OBSH in the EPR-*b*-OBSH had to be compensated when added into the EPDM compound.

**Surface treatment of wood sawdust particles by Si69 coupling agents**

A Si69 solution was prepared by mixing the silane coupling agent of 0.5wt.% of the wood content with ethanol, then sprayed onto the dried wood particles and mixed with a high-speed mixer. The mixtures were oven-dried for 24 h at 80$^{o}$C until a constant weight was achieved.

**Preparations of NR and EPDM layers**

The NR layer was prepared by masticating on a laboratory two-roll mill (Yong Fong Machinery Co., Ltd., Samutsakorn, Thailand) for 5 min before compounding with the desired contents of the vulcanizing chemicals for a further 25 min. The EPDM layer was prepared by masticating on a laboratory two-roll mill for 5 min before compounding with the desired contents of the vulcanizing chemicals and chemical blowing agents for further 15 min. 5.0mm thick of the wood/NR and *e*EPDM sheets were prepared using the laboratory two-roll mill with a controlled temperature of 45$^{o}$C, the thickness of the rubber sheets being determined by controlling the nip gap. The rubber sheets were kept at 25$^{o}$C with 50 % humidity prior to further use.

**Production of wood/NR and expanded EPDM laminates**

The unvulcanized wood/NR and EPDM layers of 5.0mm thick were thermally laminated using a LAB TECH hydraulic press (Type LP-S-20) at a lamination pressure of 15 MPa, and lamination temperature of 140$^{o}$C for a cure time of 13 min, these lamination conditions being experimentally suggested by our previous work [1] which gave the optimum peel strength between the layers as a result of co-crosslinking reaction at the interface. The laminates model is shown in Fig. 1.

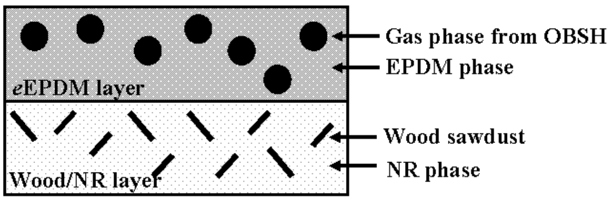

Fig.1 Wood/NR and expanded EPDM laminates

Table 1 Physical and thermal properties of OBSH blowing agents used

| Properties | OBSH | EPR-*b*-OBSH |
|---|---|---|
| OBSH content (phr) | 100 | 75 |
| EPR content (phr) | 0 | 25 |
| Physical appearance | Yellow powder | White granules |
| Decomposition temperature ($^{0}$C) | 140.5 | 147.4 |
| Supplier | Bayer Thai Co.,Ltd (Bangkok Thailand) | A.F.Goodrich Chemical Co.,Ltd |

**Characterization**

**Expanded EPDM layer**

**Foam characteristic:** The SEM micrographs of razor cut surfaces of *e*EPDM were obtained using a JEOL-JSM-6400 at 15kV accelerating voltage.

**Wood/NR-*e*EPDM laminates**

**Peel strength:** Rectangular samples 25 mm wide and 200 mm long were cut from the laminates. The *e*EPDM side was coated by a thin aluminum sheet using a neoprene-rubber adhesive to avoid elongating deformation during the test. A floating roller apparatus (ASTM D3167 (2004)) was used for determining the peel strength which were obtained within 100 mm peel distance and using the test speed at 200 mm/min. The peel strength ($P$) was calculated using Eq. 1 [2].

$$P = \frac{F}{w} \qquad (1)$$

where $F$ is the peel force (N), and w is the sample width (mm)

**Thermal conductivity:** The thermal conductivity of the wood/NR-*e*EPDM laminates followed the ASTM C 518-04 (2004) standard by the Nepzsch apparatus [3].

**Results and Discussion**

Fig. 2 shows SEM micrographs of expanded EPDM layer with OBSH and EPR-*b*-OBSH at different blowing agent contents. The number of foam cells increased with increasing OBSH content and the cell morphologies of *e*EPDM by EPR-*b*-OBSH is well-dispersed. It is believed that the EPR coating on OBSH particles may act as an effective surfactant in the EPDM matrix which would result in better dispersion and compatibility, and more uniform dispersion of the OBSH particles during compounding. Since EPR is a flexible and un-crosslinked molecule, it could enhance a stabilization of the cell structure of the EPDM foam by allowing the EPDM matrix to foam freely without collapsing and merging of the foam cells under the heat. It was observe a decrease in the average cell size of *e*EPDM samples at the OBSH loadings of 9.0-11.0phr. This was because the cell size had reached equilibrium, this being known as the gas containment limit [4], where the cell walls made contacts to each other during their expansion. At these excess loadings (9.0-11.0phr), there may be a loss of $N_2$ gas either by diffusing through the cell wall or by destroying the cell structure [5].

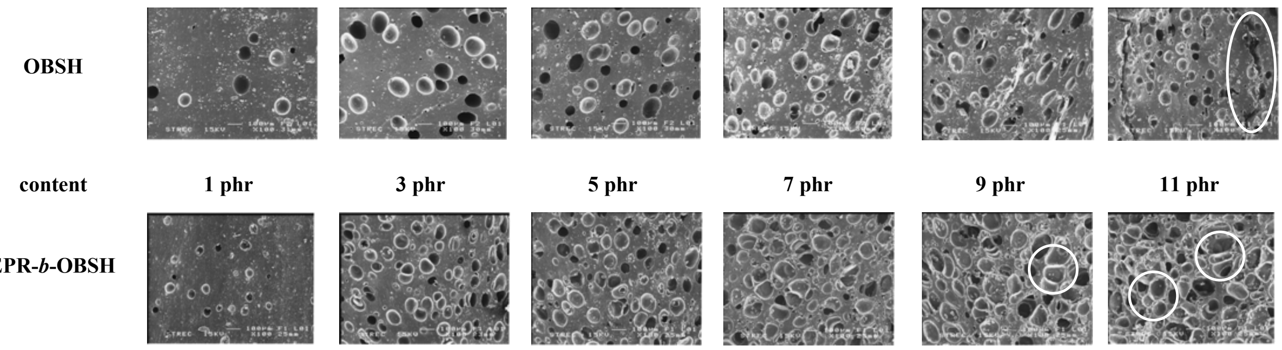


Fig. 2 Effect of OBSH loading on the peel strength of the laminates

Fig. 3 shows the effect of OBSH loading on the peel strength of the laminates. The peel strength increased with increasing OBSH loadings up to 3 phr and then dropped for a loading of 5.0phr, and then slowly declined at higher loadings (7.0-11.0 phr). Previous work [1] suggested that an improved interfacial strength between the layers was associated with co-crosslinking reactions caused by molecular diffusion and entanglement. In relation to this present work, the increase in the peel strength probably resulted from more molecular compaction at the interface as a result of expanding the EPDM layer onto the wood/NR layer. If this was the case, higher molecular diffusion and entanglement of the rubber molecules at the wood/NR and *e*EPDM interface may have taken place and thus increased the peel strength.

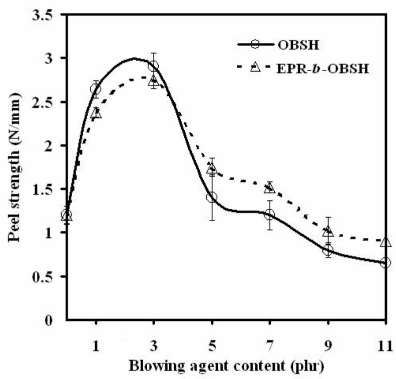


Fig. 3 Effect of blowing agent form and content on peel strength for wood/NR and *e*EPDM laminates

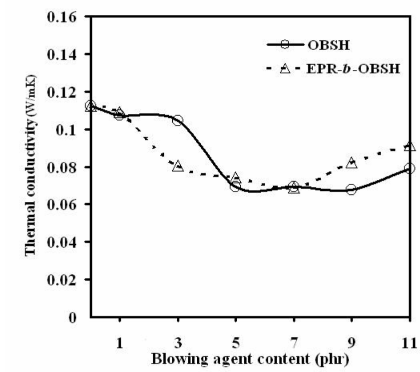


Fig. 4 Effect of blowing agent form and content on thermal conductivity for wood/NR and *e*EPDM laminates

Thermal conductivity of the laminates with both OBSH are illustrated in Fig 4. The thermal conductivity decreased from 0.11 to 0.08 W/mK when adding 5.0phr of pure OBSH or 3.0phr for EPR-*b*-OBSH. The content of blowing agent to obtain the minimum K value for EPR-*b*-OBSH agent was lower than that for pure OBSH agent. This is because of the EPDM samples with EPR-*b*-OBSH having higher expansion ratio with greater number of cells and thus gas phase which usually has very low thermal conductivity. It was interesting to note that at OBSH loadings of 11.0phr, the thermal conductivity values of the laminates tended to increase. This may be due to the effect of gas containment limit as discussed earlier in Fig. 2.

**Summary**

The experimental results indicated that the EPR-*b*-OBSH form was a more effective blowing agent than the pure OBSH form. The expanded EPDM samples with EPR-*b*-OBSH agent had higher number of cell and gave better cell stabilization. The recommended content for EPR-*b*-OBSH blowing agent to achieve the rubber laminates with maximum peel strength (~3 N/mm) with relatively low the thermal conductivity (~0.08 W/mK) was 3.0phr.

**Acknowledgments**

The authors would like to thank the Commission on Higher Education (Research fund for the Centre for Utilizations of Polymers and Natural Resources, CUP-NATURE), the Thailand Research Fund, and the Joint Graduate School of Energy and Environment (JGSEE/THESIS/119) for financial support. The authors would like to thank Dr. C. Thongpin (Silpakorn University, Nakorn-Pathom, Thailand) for her comments and suggestions during preparation of the manuscript.

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