# The Effect of Accelerator on Mechanical and Swelling Properties of NBR/EPDM/BIIR Composite

*Pengaruh Akselerator pada Sifat Mekanik dan Pembengkakan dari Komposit NBR/EPDM/BIIR*

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**Abstract- Composite is a material made from two or more materials. Composite has better properties than a single material. Additive or compatibilizer is usually needed to make a homogeneous composite. This research used Nitrile Butadiene Rubber (NBR) and Ethylene Butadiene Monomer Rubber (EPDM) as the primary matrix with Bromobutyl Rubber (BIIR) as the compatibilizer. The blending of composite rubber was done using the two-roll mill. This research studied the effect of various accelerators (DPG, MBT, CBS, TMTD, and ZDEC) in NBR/EPDM/BIIR composite. The mechanical and swelling properties of the composite were studied using tensile strength tester and swelling test. The CBS gives the highest tensile, elongation at break, and tear strength. CBS gives the lowest swelling percentage. CBS is the proper accelerator for improving the mechanical and swelling properties of the NBR/EPDM/BIIR composite.**

***Keywords: NBR, EPDM, BIIR, accelerator, compatibilizer***

***Abstrak - Komposit adalah material yang terbuat dari dua bahan atau lebih. Komposit memiliki sifat yang lebih baik daripada material tunggal. Aditif atau kompatibiliser terkadang dibutuhkan untuk membuat komposit yang homogen. Penelitian ini menggunakan Nitrile Butadiene Rubber (NBR) dan Ethylene Butadiene Monomer Rubber (EPDM) sebagai matriks utama dengan Bromobutyl Rubber (BIIR) sebagai kompatibilisernya. Pencampuran komposit karet dilakukan menggunakan two-roll mill. Penelitian ini mempelajari pengaruh berbagai akselerator (DPG, MBT, CBS, TMTD, dan ZDEC) pada komposit NBR/EPDM/BIIR. Sifat mekanik dan swelling komposit dipelajari dengan menggunakan alat uji kekuatan tarik dan uji swelling. CBS memberikan kekuatan tarik, perpanjangan putus, dan kekuatan sobek tertinggi. CBS memberikan persentase pembengkakan (swell) terendah. CBS adalah akselerator yang tepat untuk meningkatkan sifat mekanis dan sifat pembengkakan komposit NBR/EPDM/BIIR.***

***Kata kunci: NBR, EPDM, BIIR, akselerator, kompatibiliser***

## I. INTRODUCTION

Composite is one of the interesting topics because of its excellent properties. Composite is a material comes from two or more materials that stronger than a single material. The weakness of a single material can be improved by the superiority of another material. The rubber composite is popular in the industry because of its formability and its mechanical properties [1]. This research used some rubbers to make composites. Nitrile butadiene rubber (NBR) is a synthetic rubber with high resistance to solvent and oil but it has poor resistance from heat aging. Ethylene propylene diene monomer rubber (EPDM) is a synthetic rubber obtained by polymerizing ethylene and propylene with a small amount of a non-conjugated diene, which has good aging characteristics, good chemical resistance, and poor solvent resistance. Bromobutyl Rubber (BIIR) has low gas permeability, good oxidative and thermal stability, excellent chemical resistance properties and moisture barrier [2].

The blending or mixing of composite usually needs an additive to make the homogeneous composite. The differences in polarity, solubility, and poor adhesion between phases can make an inhomogeneous composite thus affect the mechanical properties. Some compatibilizers used in the rubber blending is methacrylate butadiene, styrene [3], maleic anhydrate [4], BIIR, silica [5], and bentonite [6]. The previous research shows that BIIR could be a good compatibilizer for rubber composite. Rozik *et al.* found that BIIR could improve the mechanical and electrical properties of EPDM composite [7]. NBR/EPDM composite can be made compatible by the addition of a third elastomer as a compatibilizer. BIIR in this research plays as the compatibilizer for making compatible composite. The NBR/EPDM composite can have excellent mechanical properties and resistance to oil, that suitable for making seal, gasket, transmission belts, hoses [1].

Accelerator in rubber blends plays an important role in increasing the curing rate. Many types of accelerator gives different effects on rubber composite [8][9]. Some previous research had been studied about the effect of some types of the accelerator on rubber composite, such as in NR/CR composite [10], EPDM/NR composite, [11] and NBR composite [9]. Based on the author's knowledge, there has been very limited

research about the effect of the different accelerators on the NBR/EPDM composite using BIIR as a compatibilizer.

This research used five accelerators, i.e, diphenyl guanidine (DPG), 2-mercaptobenzothiazole (MBT), n-cyclohexyl-2-benzothiazole sulfenamide (CBS), tetramethyl thiuram disulfide (TMTD), and zinc diethyldithiocarbamate (ZDEC). The purpose of this publication was to study the mechanical and swelling properties of NBR/EPDM/BIIR composites with some accelerators. This information is important to identify the suitable accelerator for NBR/EPDM composite to achieve more efficient production process.

## II. MATERIALS AND METHODS

NBR (ACN 48.5%, Krynac 4975 F) and EPDM (ENB 8.7%, Keltan 4551 A) were used as the primary matrix in this research. The additives are carbon black (CB) N330 and N774 (OCI, Korea), BIIR (Sigma Aldrich), paraffinic oil (Indrasari), paraffin wax (Antilux 654 A), n-(1,3-dimethylbutyl)-N'-phenyl-p-phenylendiamine (6PPD, Starchem), stearic acid Aflux 42 M (Rhein Chemie), zinc oxide (ZnO, Indoxide), sulfur (Miwon), diphenyl guanidine (DPG, Shandong Sianxian), 2-mercaptobenzothiazole (MBT, Bayer), n-cyclohexyl-2-benzothiazole sulfenamide (CBS, Kemai), tetramethyl thiuram disulfide (TMTD, Shandong Sianxian), and zinc diethyldithiocarbamate (ZDEC, Shandong Sianxian).

The rubber composites with the formulation in Table 1 were mixed using a two-roll mill. Then, the composite compounds were cured using a hydraulic press at 160ºC.

Table 1. The formulation of NBR/EPDM/BIIR composites

| | **Composite** | | | | |
|---|---|---|---|---|---|
| | ***NEB 1*** | ***NEB 2*** | ***NEB 3*** | ***NEB 4*** | ***NEB 5*** |
| **Materials** | ***Amount, phr*** | | | | |
| NBR | 75 | 75 | 75 | 75 | 75 |
| EPDM | 25 | 25 | 25 | 25 | 25 |
| BIIR | 5 | 5 | 5 | 5 | 5 |
| Zinc Oxide | 5 | 5 | 5 | 5 | 5 |
| Stearic acid | 1 | 1 | 1 | 1 | 1 |
| HAF CB N330 | 30 | 30 | 30 | 30 | 30 |
| SRF CB N774 | 30 | 30 | 30 | 30 | 30 |
| Paraffinic Oil | 10 | 10 | 10 | 10 | 10 |
| Paraffinic Wax | 0.5 | 0.5 | 0.5 | 0.5 | 0.5 |
| 6 PPD | 5 | 5 | 5 | 5 | 5 |
| DPG | 2 | 0 | 0 | 0 | 0 |
| MBT | 0 | 2 | 0 | 0 | 0 |
| CBS | 0 | 0 | 2 | 0 | 0 |
| TMTD | 0 | 0 | 0 | 2 | 0 |
| ZDEC | 0 | 0 | 0 | 0 | 2 |
| Sulfur | 1.5 | 1.5 | 1.5 | 1.5 | 1.5 |

*phr: part per hundred resin (primary matrix, NBR and EPDM)

The tensile strength, elongation of break, and the tear strength of the NBR/EPDM/BIIR composite were studied using the tensile strength tester Kao Tieh. The tensile strength and elongation at break were tested according to ISO 37 and the tear strength was tested according to ISO 34. The test piece of tensile strength and tear strength test are shown in Figure 1 and Figure 2.

Figure 1. Test piece of the tensile strength and the elongation at break test

Figure 2. Test piece of the tear strength test

The accelerated aging of the NBR/EPDM/BIIR composite was done according to ISO 188 using an air circulating oven (Memmert) at 100 °C for 72 h. The mechanical properties of the NBR/EPDM/BIIR composite were observed by the tensile strength tester Kao Tieh. The results of unaged composites were compared with the aged composites, to study the aging resistance. The mechanical properties retention of NBR/EPDM/BIIR was calculated using Equation (1).

$$Retention, \% = \frac{unaged\ sample - aged\ sample}{unaged\ sample}\ x\ 100 \qquad (1)$$

The swelling properties of the composite were tested according to ISO 1817 with IRM 903 for 72 h at ambient temperature. The swelling percentage was calculated using Equation (2). Where $w_1$ is the final mass and $w_0$ (g) is the initial mass (g).

$$Swelling\ percentage, \% = \frac{w_1 - w_0}{w_0}\ x\ 100 \qquad (2)$$

## III. RESULTS AND DISCUSSION

### A. *Mechanical Properties*

NBR/EPDM/BIIR compounds were vulcanized using a hydraulic press at 160 °C. The tensile strength, elongation at break, and tear strength before and after aging of the composite are shown in Figure 1 until Figure 3.

The accelerators used in this research come from various accelerator groups. The diphenyl guanidine (DPG) from guanidine group, 2-mercaptobenzothiazole (MBT) from thiazole group, n-cyclohexyl-2-benzothiazole sulfenamide (CBS) from sulfenamides group, tetramethyl thiuram disulfide

(TMTD) from thiuram group, and zinc diethyldithiocarbamate (ZDEC) from dithiocarbamate group.

The mechanical properties of composites are very important to be studied because it almost always getting loading on their application. The tensile strength described the ability of a material to resist a force that tends to pull it apart. The elongation described the ductility, elasticity, and flexibility of the material. The tear strength shows the force required to start or to continue to tear.

Figure 1 shows that the CBS and MBT as the accelerator give the highest tensile strength. The DPG gives the lowest tensile strength, while TMTD and ZDEC give tensile strength among them. The high tensile strength represents the homogeneous blend. CBS from the sufenamide accelerator could improve the compatibility of the composite.

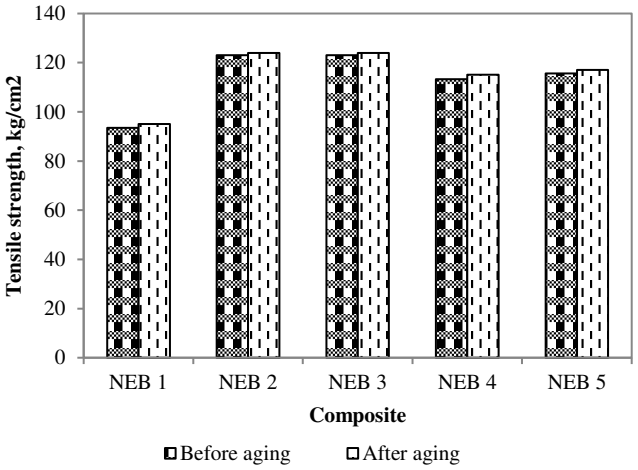


Figure 1. The tensile strength of NBR/EPDM/BIIR composites

Figure 2 the elongation at break of the composites. CBS produces the highest elongation at break, while ZDEC produces the lowest elongation at break. DPG, MBT, and TMTD produce a composite with the elongation at break among them. The elongation at break shows the maximum change in length that can be reached by the composite before rupture/failure. It represents the ductility of the material. The low elongation means the composite is brittle and it is avoided in industry. The high elongation means the material is ductile and easy to process.

Figure 3 shows the tear strength of the composite. CBS and DPG produce high tear strength, while ZDEC produces the lowest tear strength. Tear strength described the force needed to make the composite tear/fail. The high value of tear strength shows the better composite.

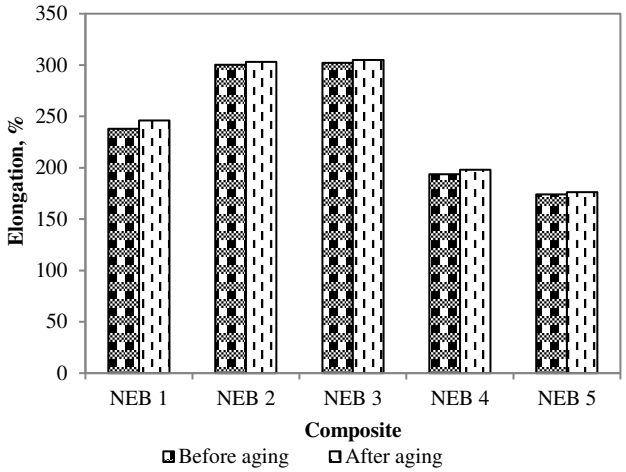


Figure 2. The elongation at break of NBR/EPDM/BIIR composites

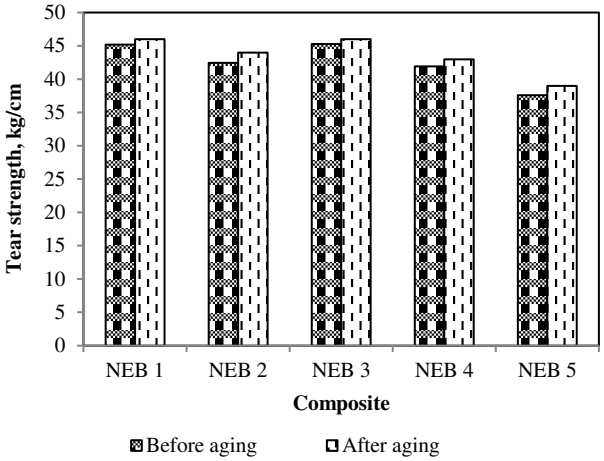


Figure 3. The tear strength of NBR/EPDM/BIIR composites

The accelerated aging test is important to determine the resistance of composite to heat or aging. The rubber composite is mostly used in above room temperature and being exposed to heat. So, the mechanical properties after aging need to be studied. The mechanical retention of the NBR/EPDM/BIIR composite is described in Table 2.

The accelerated aging test leads to an increasing in mechanical properties. The heat makes the atom move faster, thus the curing reaction becomes faster [1] thus lead to the increasing of mechanical properties. Table 2 shows that CBS gives the lowest retention (0.8-1.6%). It means that the composite has good resistance to aging. CBS from sulfenamide group accelerator can produce homogenous blends that lead to excellent mechanical properties.

Table 2. The mechanical properties retention of NBR/EPDM/BIIR composites

| Composite | % retention | | |
|---|---|---|---|
| | *Tensile strength* | *Elongation at break* | *Tear strength* |
| NEB 1 | 1.57 | 3.36 | 1.85 |

| | | | |
|---|---|---|---|
| NEB 2 | 1.08 | 1.28 | 3.61 |
| NEB 3 | 0.81 | 0.99 | 1.62 |
| NEB 4 | 1.57 | 2.18 | 2.55 |
| NEB 5 | 1.15 | 1.34 | 3.72 |

B. *Swelling properties*

Rubber composites are sometimes used in oily, such as seal, gasket application. A swelling test is important to study the resistance of the composite to the solvent. This research uses oil IRM 903 as the solvent. The swelling test is presented in Figure 4 and the swelling percentage of the NBR/EPDM/BIIR composite is presented in Figure 5.

Figure 4. The swelling test of NBR/EPDM/BIIR composites

Swelling of the composite is the presence of pressure between the rubber matrix and the solvent that works to swell or shrink the matrix [12]. Immersion in IRM 903 gives a positive value, which means the mass of the rubber increases (swell) due to the diffusion of the solvent into the rubber matrix. The swelling phenomena shows the solvent that fills in the free volume between rubber-additive. The CBS as accelerator gives the lowest swelling percentage (3.62%). The DPG gives the highest swelling percentage (5.50%). The lower the swelling percentage, the higher the solvent resistance. The low swelling percentage related to the thigh crosslinking that creates less free volume in the rubber matrix, thus the solvent more difficult to penetrate the matrix. Crosslinking is the bonding interaction between rubber and additives. High solvent resistance represents a homogenous composite.

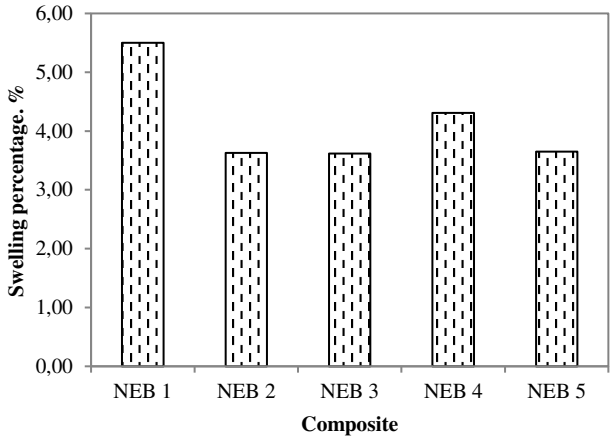


Figure 5. The swelling percentage of NBR/EPDM/BIIR composites

## IV. CONCLUSION

The different accelerators give different composite properties. Accelerators play an important role in the mechanical and swelling properties of NBR/EPDM/BIIR composites. The CBS from the sulfenamide accelerator group gives the highest tensile strength (123 kg/cm$^2$), elongation at break (124 %), tear strength (44 kg/cm), and gives the lowest increasing aging percentage (0.8-1.6%) and swelling percentage (3.62 %). The DPG gives the lowest tensile strength (94 kg/cm$^2$) and elongation at break (238 %). The ZDEC gives the lowest elongation (174 %) and tear strength (38 kg/cm). The MBT and TMTD give the composite properties between CBS, DPG, and ZDEC. From this research, it is known that the CBS is a proper accelerator for improving the mechanical and swelling properties of NBR/EPDM/BIIR composite.

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