Properties of natural rubber (NR) and wood/NR composites as gamma shielding materials

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Abstract. In this work, gamma shielding, cure characteristics, and mechanical properties of natural rubber (NR) and wood/NR composites was investigated with the addition of lead (Pb) powder for potential use as flexible shielding materials. The content of Pb powder in this work was varied from 0 to 50 part per hundred parts of rubber by weight (phr) in 10-phr increment. The results showed that the increases in Pb contents led to the improvement in gamma attenuation but had negligible effects on cure properties of the composites. The increases in Pb contents reduced overall tensile properties of the NR and wood/NR composites, while the addition of 20-phr wood particles in wood/NR composites increased the tensile strength, elongation at break, surface hardness, and dimensional stability. Hence, the overall properties investigated in this work suggested that the developed NR and wood/NR composites could be used to attenuate low-intensity gamma rays produced after former steps of shielding such as after thermal neutron or beta shielding.

1. Introduction
Gamma rays are relatively high energy and high penetrating electromagnetic waves with their energy higher than 100 keV. Gamma rays are presently used in various applications including gamma imaging [1], elemental analysis based on PIGE [2], and radiosurgery [3]. However, despite all the usefulness of gamma rays, exposure to high doses could fatally harm users and works. The risks include nausea, vomiting, fatigue, skin burns, and even death [4]. In order to reduce risks of too high exposure to gamma rays, the radiation safety principle called “As Low As Reasonably Achievable” or “ALARA”, which includes sufficient management in working time and distance, and effective radiation shielding installation and uses, must be strictly followed in all radiation-related facilities. Particularly for shielding, its importance is specially emphasized in the case of works that need extended period of time or close proximity to gamma sources. In principle, elements with high atomic numbers and densities such as lead (Pb) and Pb-containing materials are usually used as gamma shields due to their high probability of interaction with gamma rays and high energy transfer between gamma rays and nuclei of the heavy elements. Examples of gamma shielding materials are Portland cement with the addition of granulated Pb [5] and PbO-SiO₂ glasses [6].

Natural rubber has great potential to be utilized in such applications as NR is known to have high flexibility, great resistance to abrasion, and also superb toughness. Examples of NR used in radiation shielding applications were boron oxide (B₂O₃)/NR composites as neutron shielding materials [7], and Pb/NR composites as gamma shielding materials [8]. In spite of the advances in radiation shielding
development from NR, there are still rooms for further improvement, especially in terms of dimensional stability and surface hardness. One possible method to achieve these additional properties is to introduce wood particles into the composites. Since wood particles have high rigidity, they could improve dimensional stability and increase stiffness as well as surface hardness. The wood/NR composites were previously used in roofing applications that showed significant improvements in strength, UV protection, and thermal insulation [9], hence, applying wood/NR composites with Pb powders could present new developed materials that are able to utilize in wider radiation shielding applications.

As a result, this work aimed to investigate gamma shielding, cure characteristics, and mechanical properties of NR and wood/NR composites with the addition of Pb powder, in which its contents were varied from 0 to 50 parts per hundred parts of rubber by weight (phr) in 10-phr increments. It should be noted that this work focused on the attenuation of gamma rays caused after the former step of neutron shielding (B$_2$O$_3$/NR composites), hence, the intensity of the incoming gamma rays was not as high as the ones from gamma sources.

2. Experimental

2.1 Materials and chemicals

Natural rubber (STR 20) was used as the main matrix for this work. The vulcanizing formulations, which include their roles, are given in Table 1. It should be noted that types of wood particles should be negligible effects on gamma shielding properties as they mostly contain light elements, however, have potential to improve strength of the materials.

Table 1. Material formulations of NR and wood/NR composites including chemicals and their roles.

| Ingredient                          | Content (phr*) | Function               |
|-------------------------------------|----------------|------------------------|
| Natural rubber (STR 20)             | 100 (part)     | Main matrix            |
| ZnO                                 | 5.0            | Activator              |
| Stearic acid                        | 2.0            | Activator              |
| Mercaptobenzothiazole; MBT          | 0.5            | Accelerator            |
| Diphenylguanidine; DPG              | 2.0            | Accelerator            |
| Carbon black (HAF N330)             | 40.0           | Reinforcement          |
| Wood particles                      | 20.0           | Filler                 |
| Silane coupling                     | 0.5%wt of wood particles | Coupling agent      |
| UV stabilizer (CHIGUARD 234)        | 0.1            | UV absorber            |
| Anti-oxidant (TY 1076)              | 0.5            | Anti-oxidant           |
| Anti-ozonant (REDEZON 515P)         | 3.0            | Anti-ozonant           |
| Sulphur                             | 3.0            | Cross-linker           |
| Lead powder (Pb)                    | Varying from 0, 10, 20, 30, 40, and 50 | Gamma protective filler |

* phr: parts per hundred parts of rubber by weight

2.2 Preparation of NR composites

The NR was masticated on a laboratory two-roll mill (Yong Fong Machinery Co., Ltd., Thailand) for 5 min. The masticated NR was then compounded with the chemicals given in Table 1. The compounding process was also carried out on the two-roll mill for 25 min to ensure the uniform distribution of all fillers. For wood/NR composites, 20-phr of wood particles was chemically treated with a silane coupling agent and oven-dried at 100°C for 6 h until constant weight was achieved before being mixed with NR [10].

2.3 Cure characteristics

The scorch time ($t_{s1}$), cure time at 90% ($t_{90}$), and torque differences of the NR and wood/NR compounds were evaluated using an oscillating die rheometer (ODR GT 70-70-S2, GOTECH Testing machine, Inc., Taiwan) following ASTM D2048-11 standard testing at the temperature of 160°C. The determined cure times were then for Vulcanizing the NR composites sheets in hydraulic press at 160 kg/cm$^2$ for preparation on NR composite specimen sheet.
2.4 Mechanical and physical properties
The tensile properties including tensile modulus, tensile strength, and elongation at break of the NR and wood/NR composites were investigated using a universal testing machine (Autograph AG-I 5kN, Japan) following ASTM D412-06 standard testing. For hardness measurement, all the samples were tested using a hardness durometer, Shore A (Teclock GS-719G, Japan) according to ASTM D2240-05.

2.5 Gamma ray shielding properties
The gamma ray shielding properties were performed at the Thailand Institute of Nuclear Technology (TINT), in which the incoming gamma rays were produced after thermal neutron shielding with 300-mCi $^{241}$Am/Be as a neutron source and 80-phr B$_2$O$_3$/NR as neutron shielding materials. The gamma detector used in this work was a survey meter (LUDLUM 2200) connected to a set of high voltage power supply, amplifier, and timer/counter.

3. Results and Discussion

3.1 Cure characteristics
Cure characteristics of NR and wood/NR composites are shown in Table 2. The results indicated that adding Pb powder to the composites did not significantly change cure characteristics due to the fact that Pb does not chemically interact with NR matrix. Furthermore, it was observed that wood/NR composites had higher scorch time and cure time compared with NR composites at all Pb contents. This was because wood particles had hydroxyl groups that could absorb activators and accelerators, slowing the chemical vulcanizing process [7]. In terms of the torque differences, the results indicated that increasing Pb contents led to the increases in the values due to the high rigidity of Pb powder.

| Pb content (phr) | Scorch time (s) NR composites | Cure time (s) NR composites | Torque difference (dN m) NR composites | Scorch time (s) Wood/NR composites | Cure time (s) Wood/NR composites | Torque difference (dN m) Wood/NR composites |
|------------------|-------------------------------|----------------------------|-----------------------------------------|-----------------------------------|-------------------------------|-----------------------------------------|
| 0                | 34±2                          | 184±1                      | 74±1                                    | 42±8                              | 222±12                       | 79±1                                    |
| 10               | 35±2                          | 200±2                      | 93±0                                    | 40±5                              | 250±13                       | 70±1                                    |
| 20               | 34±3                          | 205±2                      | 95±0                                    | 38±1                              | 220±2                        | 85±0                                    |
| 30               | 35±2                          | 206±3                      | 96±0                                    | 38±3                              | 219±1                        | 86±1                                    |
| 40               | 34±1                          | 204±2                      | 97±0                                    | 37±1                              | 220±0                        | 87±1                                    |
| 50               | 34±1                          | 208±4                      | 97±0                                    | 37±3                              | 222±2                        | 86±0                                    |

3.2 Mechanical properties
The results of mechanical properties are shown in Table 3, which suggested that increases in Pb contents reduced overall tensile properties of both NR and wood/NR composites, while the addition of 20-phr wood particles in wood/NR composites increased the tensile strength, elongation at break, surface hardness, and dimensional stability. The reduction in tensile properties caused by adding Pb powder were due to the poor interfacial compatibility of fillers.

| Pb content (phr) | Tensile modulus (MPa) NR composites | Tensile strength (MPa NR composites) | Elongation at break (%) NR composites | Hardness (Shore A) NR composites | Tensile modulus (MPa) Wood/NR composites | Tensile strength (MPa Wood/NR composites) | Elongation at break (%) Wood/NR composites | Hardness (Shore A) Wood/NR composites |
|------------------|-------------------------------------|-------------------------------------|--------------------------------------|---------------------------------|------------------------------------------|------------------------------------------|------------------------------------------|-------------------------------------|
| 0                | 3.7±0.0                             | 14.4±0.9                            | 316±19                               | 74±1                            | 3.7±0.0                                  | 15.4±0.3                                | 460±10                                  | 76±1                                |
| 10               | 3.6±0.0                             | 11.0±1.2                            | 262±26                               | 70±2                            | 4.0±0.0                                  | 14.0±1.0                                | 391±24                                  | 75±1                                |
| 20               | 3.6±0.0                             | 9.2±2.4                             | 228±52                               | 70±1                            | 3.7±0.0                                  | 13.3±0.5                                | 411±11                                  | 75±1                                |
| 30               | 3.6±0.0                             | 7.8±0.9                             | 197±22                               | 71±1                            | 3.6±0.1                                  | 12.1±0.5                                | 387±29                                  | 75±1                                |
| 40               | 3.5±0.0                             | 7.8±1.4                             | 213±34                               | 70±2                            | 3.4±0.1                                  | 11.0±0.6                                | 372±8                                   | 75±1                                |
### 3.3 Gamma shielding properties

Figure 1. Shown the gamma shielding properties of NR and wood/NR composites, Fig.1a indicates the values of linear attenuation coefficient ($\mu$) of samples with different Pb contents while Fig.1b indicates the gamma transmission of samples with 50-phr Pb content at different thicknesses. The overall results implied that increasing Pb contents resulted in the improvement of shielding abilities as seen by the increases in the $\mu$ values. Furthermore, it was observed that the NR composites had slightly higher $\mu$ than the wood/NR composites due to the dilution effect of wood particles in wood/NR composites that reduced the capability of Pb powders to attenuate gamma ray. However, after considering gamma transmission shown in Fig.1b, negligible differences in gamma attenuation were observed at thicker sheets of samples, indicating similar gamma shielding properties between the two composites and great potential for wood/NR composites to be used as flexible gamma shielding materials.

| Pb content (phr) | Tensile modulus (MPa) | Tensile strength (MPa) | Elongation at break (%) | Hardness (Shore A) | Tensile modulus (MPa) | Tensile strength (MPa) | Elongation at break (%) | Hardness (Shore A) |
|------------------|-----------------------|------------------------|-------------------------|-------------------|-----------------------|------------------------|-------------------------|-------------------|
| 50               | 3.5±0.1               | 6.4±1.3                | 178±33                  | 71±1              | 3.5±0.0               | 10.9±0.2               | 368±8                  | 76±1              |

### 4. Conclusions

Flexible gamma shielding materials based on NR and wood/NR composites with addition of Pb powder were developed. The results showed that wood particles improved overall mechanical properties and dimensional stability of the composites, but negligibly affected gamma shielding properties. Furthermore, the Pb powders effectively acted as gamma protective fillers as seen by the improvement in gamma attenuation with increasing Pb content. Based on the experimental results in this work, it could be concluded that NR and wood/NR composites had great potential to be used as flexible and effective gamma shielding materials.

### 5. References

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