

Sustainable Natural Rubber Composites Reinforced with Eggshell-Derived CaCO₃/BaCO₃ Hybrid Fillers: Mechanical Performance and Radiation Shielding

Pitisan Banyen¹, Nathapong Sukhawipat^{2,3}, Natsupon Chutithanapanon⁵, Narongrit Sosa⁴,
Chadet Yenchai⁶, Siriprapa Kaewjaeng^{7,8}, Wichain Chailad^{1,*}

¹Department of Materials and Metallurgical Engineering, Faculty of Engineering, Rajamangala University of Technology Thanyaburi, Pathum Thani 12110, Thailand

²Department of Mechanical Engineering Technology, College of Industrial Technology, King Mongkut's University of Technology North Bangkok, Bangkok 10800, Thailand.

³Center of Aircraft and Defence Technology (CADeT), Techno Park, King Mongkut's University of Technology North Bangkok, Bangkok 10800, Thailand

⁴Department of Applied Science and Social, College of Industrial Technology, King Mongkut's University of Technology North Bangkok, Bangkok, 10800, Thailand

⁵Functional Materials and Nanotechnology Center of Excellence, Walailak University, Nakhon Si Thammarat 80160, Thailand.

⁶Department of Nuclear Engineering, Faculty of Engineering, Chulalongkorn University, Bangkok 10330, Thailand.

⁷Department of Radiologic Technology, Faculty of Associated Medical Sciences, Chiang Mai University, Chiang Mai 50200, Thailand

⁸Biomedical Engineering and Innovation Research Center, Chiang Mai University, Chiang Mai, 50200, Thailand

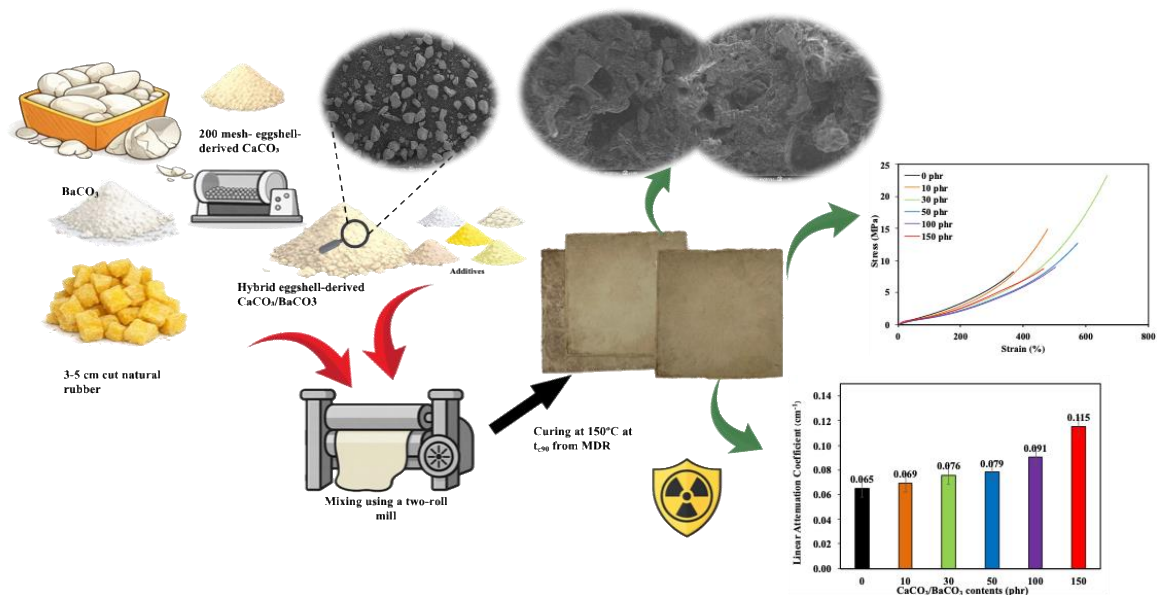
*Corresponding's email: wichain_c@rmutt.ac.th

Abstract

This study investigates the use of hybrid fillers based on eggshell-derived calcium carbonate (CaCO_3) and barium carbonate (BaCO_3) for improving the mechanical and X-ray shielding properties of natural rubber (NR) composites. This work proposes a sustainable hybrid filler approach that combines waste-derived eggshell CaCO_3 with high atomic number BaCO_3 to simultaneously enhance mechanical performance and X-ray attenuation efficiency in NR composites. The hybrid filler was prepared by ball milling CaCO_3 (200 mesh) and BaCO_3 at a fixed weight ratio of 50:50, while SEM/EDX analysis was used to examine the morphology and elemental distribution of the hybrid filler prior to incorporation into the NR matrix at loadings ranging from 0 to 150 phr. At low to moderate filler contents, the NR composites exhibited improved mechanical performance. Tensile strength increased and reached a maximum value of 23.3 MPa at 30 phr, while tear strength showed the highest value of 27.9 N/mm at 10 phr. At higher filler loadings, both properties gradually decreased, which was attributed to filler agglomeration and the formation of stress concentration regions within the NR matrix, as observed from SEM analysis. In contrast, hardness continuously increased from 28.5 to 58.4 Shore A with increasing filler content, indicating reduced mobility of the polymer chains due to the reinforcing effect of the hybrid filler. The incorporation of the hybrid filler also significantly enhanced X-ray attenuation performance. The linear attenuation coefficient (μ) increased from 0.13 cm^{-1} for neat NR to 1.3 cm^{-1} at 150 phr, mainly due to the presence of high atomic number BaCO_3 , which improves photon interaction within the composite. In addition, neutron attenuation performance improved with increasing filler loading. The μ values for fast neutrons increased from 0.065 to 0.115 cm^{-1} , while thermal neutron attenuation reached a maximum value of 1.31 cm^{-1} at 100 phr. Overall, filler contents in the range of 10–30 phr provided a more balanced combination of mechanical properties and structural integrity, whereas higher filler loadings were more effective for X-ray shielding applications. The developed composites show potential for use in flexible X-ray shielding sheets, medical protective materials, and radiation-related industrial applications.

Keywords: Natural rubber composites, Egg shell–derived calcium carbonate, Barium carbonate, Hybrid filler, Lead-free radiation shielding

Graphical abstract



References

- Ardiansyah, A., Armynah, B., Heryanto, H., Sukaryo, S.G., Abdul Karim, M.K., Tahir, D., 2024. X-ray radiation shielding: Enhanced performance with BaCO_3 nanocrystal-infused composite polyester/PVA/ BaCO_3 exhibiting medicine capsule-like structure. *Mater. Chem. Phys.* 319, 129366. <https://doi.org/10.1016/j.matchemphys.2024.129366>
- Armart, K., Boontueng, P., Mutuwong, C., Prasertsri, S., Pancharee, S., Kaewkhao, J., Thongjerm, P., Wonglee, S., Pornroongruengchok, W., Saenpoowa, J., Kobdaj, C., 2025. Development of the thermal neutron shielding efficiency and its secondary radiation of natural rubber composites with a multilayer structure. *Radiat. Phys. Chem.* 236, 112944. <https://doi.org/10.1016/j.radphyschem.2025.112944>
- Barrera, C.S., Tardiff, J.L., 2022. Static and dynamic properties of eggshell filled natural rubber composites for potential application in automotive vibration isolation and damping. *J. Clean. Prod.* 353, 131656. <https://doi.org/10.1016/j.jclepro.2022.131656>
- Longkaew, K., Gibaud, A., Tessanan, W., Daniel, P., Phinyocheep, P., 2023. Spherical CaCO_3 : Synthesis, Characterization, Surface Modification and Efficacy as a Reinforcing Filler in Natural Rubber Composites. *Polymers (Basel)*. 15, 4287. <https://doi.org/10.3390/polym15214287>
- Muhammad, N.A., Armynah, B., Tahir, D., 2022. High transparent wood composite for

effective X-ray shielding applications. Mater. Res. Bull. 154, 111930.

<https://doi.org/10.1016/j.materresbull.2022.111930>

Tangboriboon, N., Takkire, R., Sangwan, W., Changkhamchom, S., Sirivat, A., 2019. BIO-CACO₃ FROM RAW EGGSHELL AS ADDITIVE IN NATURAL RUBBER LATEX GLOVE FILMS. Rubber Chem. Technol. 92, 558–577.

<https://doi.org/10.5254/rct.19.81489>