DEVELOPMENT OF A THERMOPLASTIC VULCANIZATE FROM NATURAL RUBBER AND POLYETHYLENE AS A COMPETITIVE MATERIAL FOR ROOFING SHEETS

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DECLARATION OF CANDIDATE AND SUPERVISORS

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ABSTRACT

Development of a Thermoplastic Vulcanizate from Natural Rubber and Polyethylene as a Competitive Material for Roofing Sheets

In Sri Lanka, roofing materials are commonly manufactured from clay, metal, plastic, wood, and asbestos, each associated with specific limitations. Despite well-documented health risks, asbestos roofing remains widely used due to its low cost. However, extensive scientific evidence linking asbestos exposure to severe health hazards has prompted the Government of Sri Lanka to initiate measures toward banning asbestos-based roofing materials. This has created an urgent need for safer, durable, and cost-effective alternatives. Thermoplastic vulcanizates (TPVs), produced through dynamic vulcanization in which a rubber phase is selectively crosslinked and dispersed within a thermoplastic matrix, offer a promising solution. In this context, the present study focuses on developing a sustainable TPV roofing material using natural rubber (NR) and polyethylene (PE).

Initially, NR/PE blends were prepared via melt blending using a twin-screw extruder with three polyethylene grades, namely low-density polyethylene (LDPE), linear low-density polyethylene (LLDPE), and high-density polyethylene (HDPE), at varying compositions to identify the most suitable PE grade for roofing applications. Based on mechanical, physical, and water absorption performance, HDPE was identified as the optimal thermoplastic component. Subsequently, TPVs from NR and HDPE were further optimized by varying blend composition and incorporating particulate fillers to improve performance and reduce cost. Six fillers, including dolomite, calcium carbonate, barium sulfate ($BaSO_4$), snobrite clay, talc, and kaolin, were evaluated, with $BaSO_4$ showing the best overall compatibility and reinforcement. Further optimization involved adjusting the loadings of dicumyl peroxide (DCP) as the vulcanizing agent, $BaSO_4$ filler, and a zirconate coupling agent to enhance interfacial adhesion. Mechanical, thermal, and physical properties were evaluated in accordance with international standards, and morphology was examined using scanning electron microscopy. Prototype roofing sheets (40 cm × 40 cm × 0.6 cm) were fabricated by compression moulding and assessed against relevant PVC-based roofing specifications.

NR/HDPE-based TPVs showed higher tensile strength, tear strength, and hardness than those made with LDPE and LLDPE. Although none reached the 25 MPa tensile strength of commercial PVC-based roofing sheets, all exceeded the 3 MPa minimum required for roofing, with impact strength surpassing that of PVC-based roofing sheets. NR/HDPE-based TPV exhibited the lowest water absorption (0.04%), nearly matching PVC-based roofing sheets and significantly better than asbestos sheets. Based on overall performance, HDPE was identified as the most suitable PE grade for NR/PE roofing applications. The 20/80 (NR/HDPE) composition offers the best balance of mechanical strength and impact resistance, making it ideal for roofing applications. The TPV prepared with $BaSO_4$ filler demonstrated the highest overall performance, with optimal results observed at a filler loading of 50 phr and 1 phr of DCP. A zirconate loading of 1.5 phr resulted in optimum mechanical properties and refined phase morphology, indicating enhanced interfacial compatibility between the NR and HDPE phases in the TPV. The prototype sheet prepared using the optimized formulation showed comparable impact strength, elongation at break, density, and thermal conductivity to that of conventional PVC-based roofing sheets.

Keywords: Barium sulfate, Dynamic-blending, High-density polyethylene, Natural rubber, Zirconate coupling agent

DEDICATION

To,

My ever-loving parents

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LIST OF ABBREVIATIONS

Abbreviation	Description
ANOVA	Analysis of Variance
ASTM	American Society for Testing and Materials
$BaSO_4$	Barium Sulfate
CPE	Chlorinated Polyethylene
$CaCO_3$	Calcium Carbonate
CSPE	Chlorosulfonated Polyethylene
DBM	Dibenzoylmethane
DCP	Dicumyl Peroxide
EPDM	Ethylene Propylene Diene Monomer
ENR	Epoxidized Natural Rubber
$FeCl_2$	Ferrous Chloride
GO	Graphene Oxide
HDPE	High Density Polyethylene
HMWPE	High Molecular Weight Polyethylene
IPPD	Isopropyl Paraphenylene Diamine
IRG	Irganox
LDPE	Low Density Polyethylene

LLDPE	Linear Low Density Polyethylene
LNR	Liquid Natural Rubber
MDPE	Medium Density Polyethylene
MBTS	Dibenzothiazyl Disulfide
$Mg(OH)_2$	Magnesium Hydroxide
NR	Natural Rubber
PC	Polycarbonate
PE	Polyethylene
PEm	Maleic Anhydride Polyethylene
PP	Polypropylene
PVC	Polyvinyl Chloride
SBC	Snobrite Clay
SEM	Scanning Electron Microscope
SLS	Sri Lanka Standards
$SnCl_2$	Stannous Chloride
TMTD	Tetramethylthiuram Disulfide
TIN	Tinuvin
TPE	Thermoplastic Elastomer
TPE-A	Polyamide/Elastomer Block Copolymer
TPE-E	Polyester Block Copolymer

TPE-O	Thermoplastic-Elastomer Olefin
TPE-S	Styrene Block Copolymer
TPE-U	Thermoplastic Polyurethane Elastomer Block
TPE-V	Thermoplastic-Elastomer Vulcanizate
TPV	Thermoplastic Vulcanizate
UHMWPE	Ultra High Molecular Weight Polyethylene
ULMWPE	Ultra Low Molecular Weight Polyethylene
WSP	Lowinox
$ZnCl_2$	Zinc Chloride