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Multivariate Modeling of Structure–Performance Relationships in Polybutadiene-Based Elastomeric Composites Containing Recovered Carbon Black

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Introduction

Recovered carbon black (rCB), obtained from the pyrolysis of end-of-life tires [1], can be used as a filler in elastomeric composites. The properties of these materials are influenced by microstructural factors, such as filler dispersion, interfacial interactions, and crosslink density. Multivariate artificial neural networks (ANNs) enable the simultaneous modeling of different physical, mechanical, and rheometric responses while accounting for their potential interrelationships.

Objectives

To develop and evaluate multivariate ANNs for the simultaneous prediction of nine properties of polybutadiene (BR) composites containing rCB, investigating the predictive performance and stability of different network architectures through cross-validation and independent experimental validation.

Metodology

A total of 72 samples from six formulations containing 0–50 phr of rCB were used [2]. The rCB content was used as the input variable of the multivariate ANN, with nine properties as outputs. Crosslink density was determined from equilibrium swelling measurements using the Flory-Rehner equation [3]. The networks were developed in MATLAB, trained using normalized data and Bayesian regularization [4], and evaluated through leave-one-formulation-out (LOFO) cross-validation, stability analysis, and external experimental validation at 12, 23, and 37 phr.

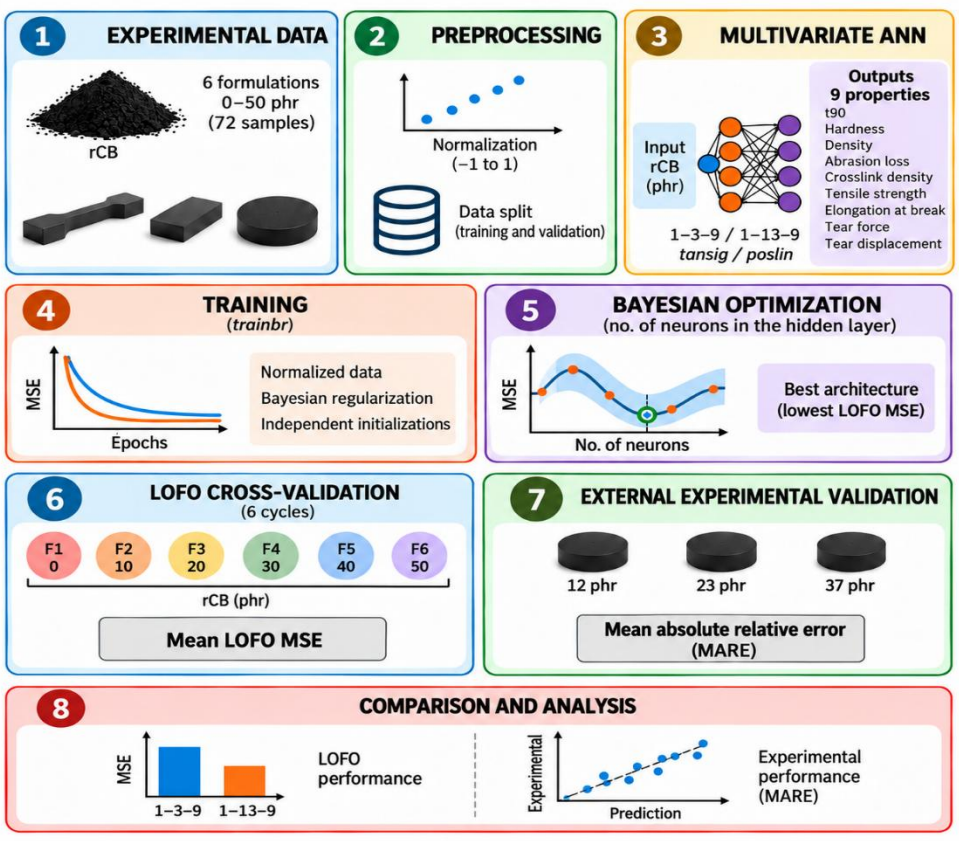


Figure 1. Stages of the development and validation of the multivariate ANNs.

Results and Discussion

The 1-13-9 architecture achieved a lower mean LOFO MSE (0.0571) than the 1-3-9 architecture (0.0615). However, the 1-3-9 architecture exhibited greater stability and lower mean absolute relative errors in the independent experimental validation, including hardness (3.33%), abrasion loss (4.81%), and tensile strength at break (4.30%).

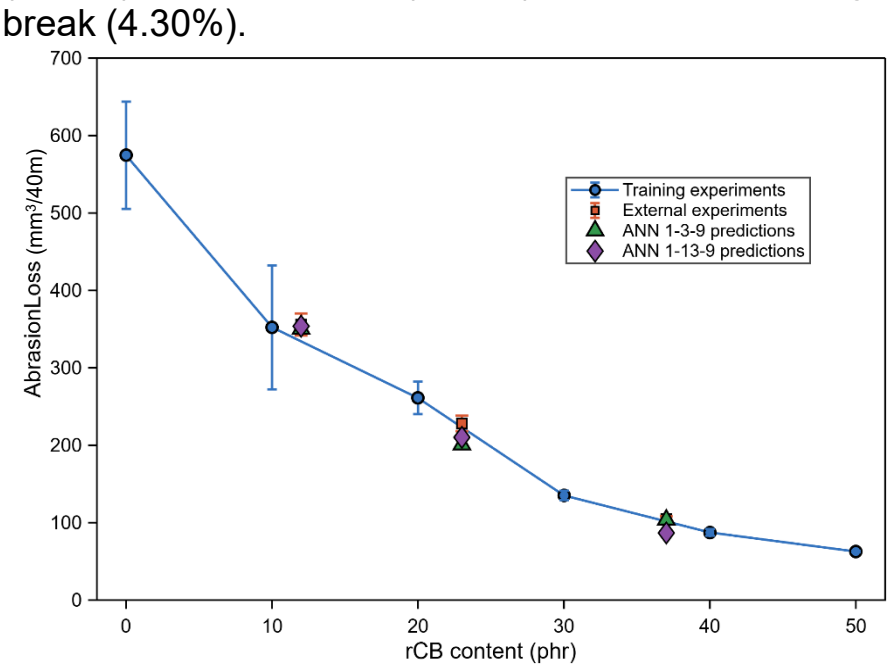


Figure 2. Comparison of the training experimental data, external experimental validation results, and predictions from the 1-3-9 and 1-13-9 ANN architectures for abrasion loss.

Table 1. ANN performance in LOFO cross-validation and external experimental validation.

Property/Metric	RNA 1-3-9 *tansig*	RNA 1-13-9 *poslin*
External validation(MARE)		
Hardness	**3,33%**	3,39%
Abrasion loss	**4,81%**	8,42%
Tensile strength	**4,30%**	5,87%
Displacement at tear	**0,70%**	0,88%
LOFO cross-validation (MSE)		
Mean normalized MSE	0,0615	**0,0571**

Conclusions

The multivariate approach demonstrated potential for the simultaneous prediction of properties of BR/rCB composites. Although Bayesian optimization identified architectures with lower mean LOFO errors, the 1-3-9 network exhibited greater stability and better relative predictive performance in independent experimental validation. These findings highlight the importance of jointly considering prediction errors, training stability, and generalization ability when selecting ANN architectures for the development of elastomeric composite formulations.

References and Acknowledgments

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