Evaluation of Suitability of Lemongrass Powder as an Alternative Antioxidant for Dry Rubber Composites

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Abstract

Ageing of natural rubber based products leads to reduction in various properties and hence deterioration in quality of the final products. The use of antioxidants (AOs) in rubber based compounding has been introduced to mitigate this issue up to a certain level. The commercially available AOs tend to generate hazardous substances which can affect the eco systems adversely. This study was carried out to probe the possibility of using lemongrass (*Cymbopogon citratus*) root stem and leaf powder (LGP) as an alternative natural AO for natural rubber based dry rubber composites. Cure and ageing properties of composites containing commercially available AOs such as N-isopropyl-N'-phenyl-p-phenylenediamine (IPPD), 2,2,4-trimethyl-1,2-dihydroquinoline (TMQ) and 2-2'-methylenebis [6- (1-methylcyclohexyl)-P-cresol] (WSP) were compared with those of the composite produced with LGP. The dry rubber composite produced without any AO was used as the control. The comparison revealed that the LGP incorporated composite generates better processing safety than that prepared with IPPD and higher cure rate index than that prepared with TMQ, which are considered as ubiquitous AOs in the industry. Additionally, the former composite provided superior qualities not only with respect to cure characteristics, but also in regard to retention of tensile strength and elongation at break than the WSP incorporated composite. It can be concluded that it is possible to substitute the phenolic type AO WSP with LGP in dry rubber composites.

***Keywords**: Lemongrass, antioxidants, cure characteristics, tensile properties, thermal ageing*

1. Introduction

Antioxidants (AOs) play a vital role in retarding the undesirable reactions taking place between the organic matters with atmospheric oxygen. The aforementioned reactions can cause degradation of the properties of a rubber product which will eventually affect the quality of the product adversely. Addition of AOs to polymers depends on the chemical composition of the base material and the extent of exposure to the atmosphere. Generally, for most unsaturated polymers such as natural rubber 0.5-2 phr of AOs are added during the process (Thomas *et al.,*2000). The categorization of AOs can vary according to its main functional groups, staining/non-staining nature and according to different test methods.

The main three classes of AOs used for this study falls into the paraphenylene diamines (PPDs), trimethyl-dihydroquinolines (TMQs) and phenolics (e.g. WSP) which are commonly used in the industry for rubber based applications. PPDs are known as the most powerful AOs which exert a high level of staining effect and scorching on rubber compounds. TMQs on the other hand exhibits a lesser staining effect than the PPDs and provide more or less similar AO activity to the rubber product. However, phenolic type AOs which are generally known as non-staining AOs have lower activity in comparison to PPDs.

The use of commercially available synthetic AOs cause numerous undesirable effects, short term and long term adverse health effects on human beings as well as disturbs the ecosystem equilibrium, e.g. 6PPD affects embryonic development of zebrafish (Xu *et al*., 2022). As a result several studies based on natural AOs have been conducted during the past to probe the possibility of substituting commercially available AOs with natural sources (Zhao *et al.,* 2022, Sukatta *et al*., 2021, Öncel *et al*., 2019). This study was based on a novel approach where the suitability of lemon grass powder (LGP) was accessed in the areas of cure characteristics and physico-mechanical properties with the incorporation into rubber compounds. The phenolic substances present within the LGP plays the major role in the action of AO property (Soliman *et al.,* 2017).

2. Materials and Methods

Materials

The crepe rubber (1X) used for this study was supplied by the Rubber Research Institute of Sri Lanka, Dartonfield, Agalawatta and the other ingredients were sourced from the local market. The control natural rubber (NR) composite was prepared without any AO whereas in the other composites IPPD, TMQ, WSP and LGP were used.

Methodology

Mixing of the ingredients in the formulation given in Table 1 to produce the five NR composites was conducted using a laboratory scale two-roll mill.

Table 1. Formulation of NR composites

Ingredient	Function	Parts per hundred parts of rubber (phr)
Crepe rubber (1X)	Rubber	100
ZnO	Activator	5
Stearic acid	Co-activator	2
TMTD	Secondary accelerator	0.3
TBBS	Primary accelerator	1.5
$CaCO_3$	Filler	40
White oil	Paraffinic Processing oil	4
Sulphur	Vulcanizing agent	2.5
WSP/TMQ/IPPD/LGP	Antioxidant	1

Analysis of cure characteristics

The cure characteristics of the composites, minimum torque (M_L), maximum torque (M_H), scorch time, 90% cure time and delta cure (M_H-M_L) were evaluated using a Rubber Process Analyzer (D-RPA 3000) according to ASTM D6601 at 150 ^{0}C.

The cure rate index (CRI) was calculated according to the following formula

$$CRI = \frac{100}{\text{Cure time} - \text{Scorch time}}$$

Preparation of test specimens

Test specimens required to carry out tensile properties before and after ageing were prepared using a hydraulic hot press according to the respective cure times obtained from the D-RPA.

Analysis of tensile properties

The Instron tensile testing machine was used to measure the tensile properties namely, tensile strength and elongation at break of the vulcanisates at room temperature (27 ± 2 ^{0}C) at a grip separation rate of 500 mm /min. in accordance with ISO 37: 2017

Analysis of resistance to thermal ageing

The rubber vulcanisates were aged at 70 ± 2 ^{0}C for 72 hrs in an air circulating oven at 100 ^{o}C (Sanyo Gallenkamp, UK) according to ISO 188: 2011. Tensile strength and elongation at break were evaluated after ageing.

3. Results & Discussion

Cure characteristics

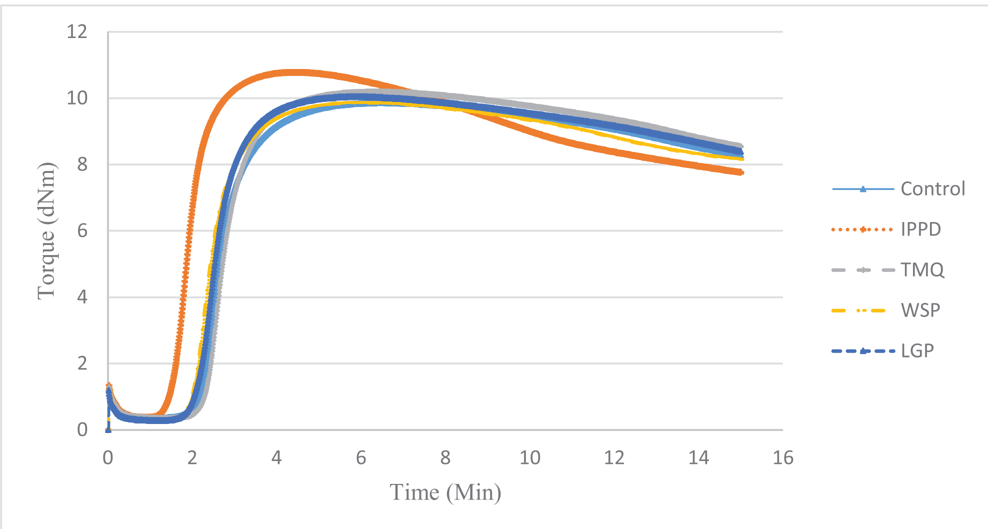

Figure 1. Rheographs of the NR composites

The cure curves of all the composites are illustrated in Figure 1. According to the curves it can be seen that the composite with LGP indicates similar characteristics to those of the composites containing TMQ and WSP as well as to those of the control.

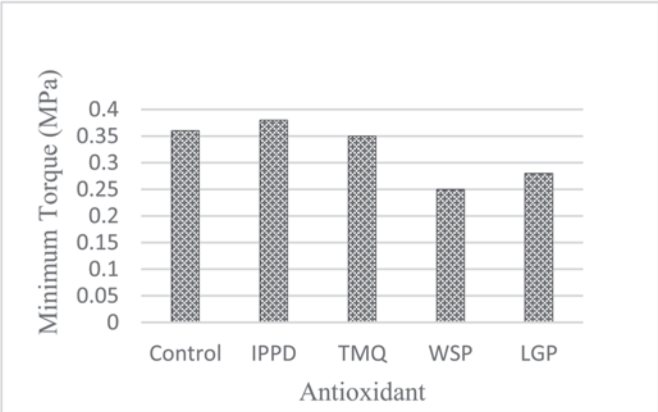

Figure 2. Minimum torque of the NR composites

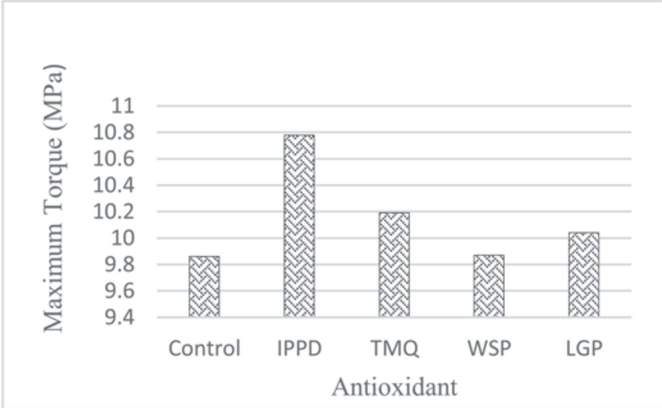

Figure 3. Maximum torque of the NR composites

The minimum torque which indicates the stock viscosity and processability of the unvulcanized rubber composite shows greater values for the control composite and the ones with staining AOs (IPPD and TMQ) and lower values for WSP and LGP incorporated composites (Figure 2). The LGP incorporated composite shows lower viscosity and enhanced processability in comparison to the composites containing IPPD and TMQ. The highest maximum torque can be observed in the compound with IPPD and the lowest being the composite with WSP (Figure 3). The maximum torque of the LGP containing composite is in between the TMQ containing composite and the WSP containing composite. This indicates that the LGP containing composite provide better results than the WSP containing composite in regard to the maximum torque enabling a reasonable level of hardness for the vulcanizate.

Further, the LGP incorporated composite reveals better scorch safety compared to that of IPPD and WSP incorporated composites (Figure 5). Figure 4 indicates that the composite prepared with IPPD cures faster than all the other composites and is in agreement with the results of cure rate (Figure 7). When considering the cure rate, the LGP incorporated composite exhibits the second highest value indicating a better efficiency of curing than the control, TMQ and WSP containing composites.

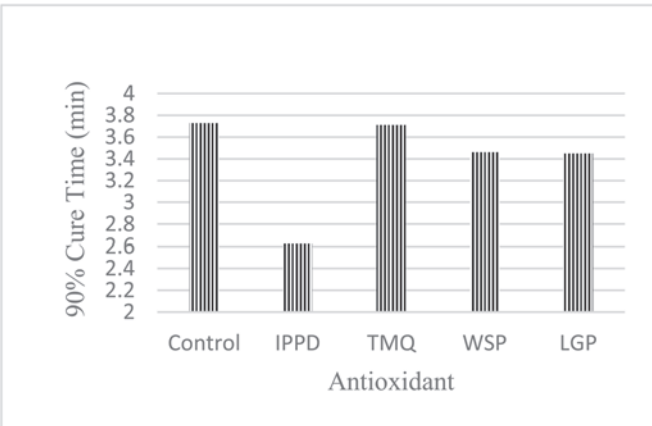

Figure 4. Cure time of the NR composites

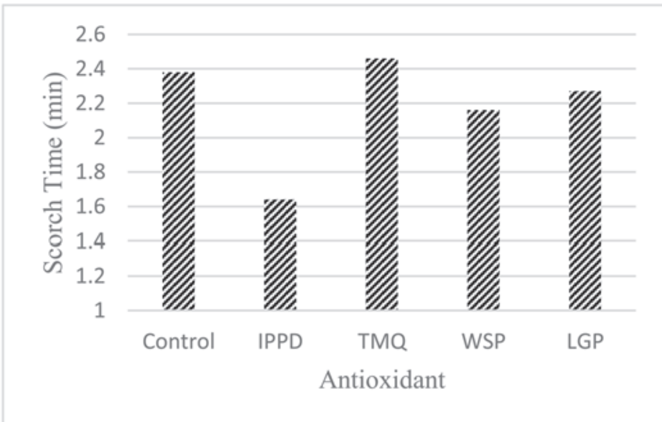

Figure 5. Scorch time of the NR composites

Delta cure (M_H-M_L) of a rubber compound can be related to its crosslink density and hence the results illustrated in Figure 6 reveal that the LGP incorporated composite has a higher crosslink density than the WSP incorporated composite.

Overall, comparison of cure characteristics of all the five natural rubber composites indicates that the LGP incorporated composite has better characteristics than the WSP incorporated composite with respect to maximum torque, scorch time, delta cure and cure rate index.

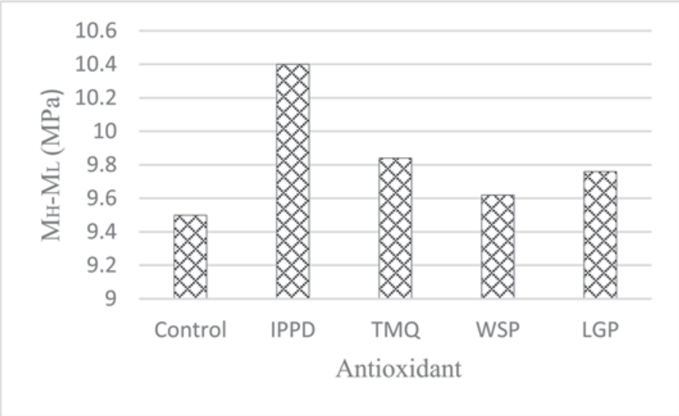

Figure 6. Delta cure of the NR composites

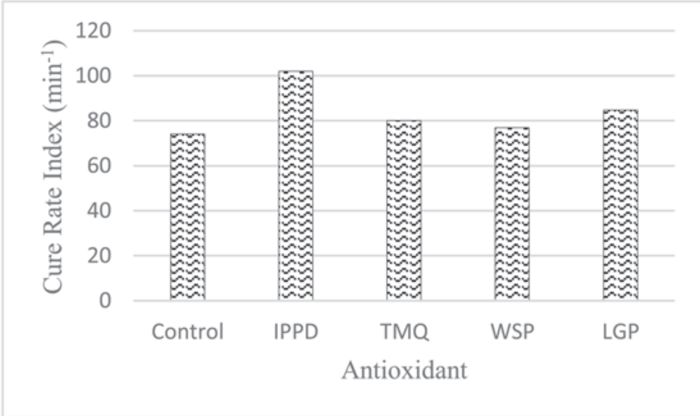

Figure 7. Cure Rate Index of the NR composites

Tensile properties before and after ageing

Tensile strength of the five vulcanizates before ageing indicated almost similar values ranging from 21.2-21.4 MPa (Figure 8). However, with the applied ageing conditions the tensile strength has deteriorated giving the lowest value for the control (14.7 MPa) and the highest value for the TMQ incorporated composite (20.1 MPa). The ageing resistance of the LGP incorporated composite is higher than that of the composite prepared with WSP.

Elongation at break of the vulcanisates before and after ageing is illustrated in Figure 9. There is a variation in elongation at break among the vulcanisates before ageing. The highest and lowest values are shown by the TMQ and IPPD incorporated vulcanisates, respectively. However according to after ageing results, elongation at break of the three vulcanisates produced with TMQ, WSP and LGP is almost similar and the control exhibits the lowest value.

Retention of tensile strength and elongation at break after ageing for 72 hrs is illustrated in Figure 10. The retention values with respect to tensile strength indicate that the vulcanisates produced with the AOs TMQ and IPPD have greater resistance towards thermal ageing followed by LGP and WSP incorporated vulcanisates. The percentage retention values of elongation at break after ageing indicate similar results for TMQ, WSP and LGP incorporated vulcanisates, whilst the IPPD incorporated vulcanisate has generated a superior result (96.4 %).

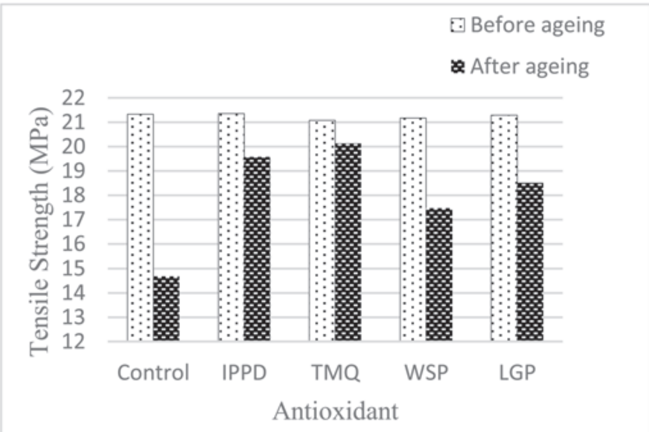

Figure 8. Tensile strength of the NR composites

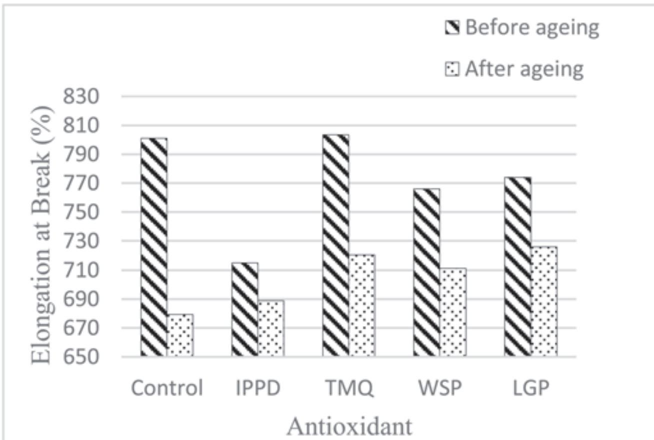

Figure 9. Elongation at break of the NR composites

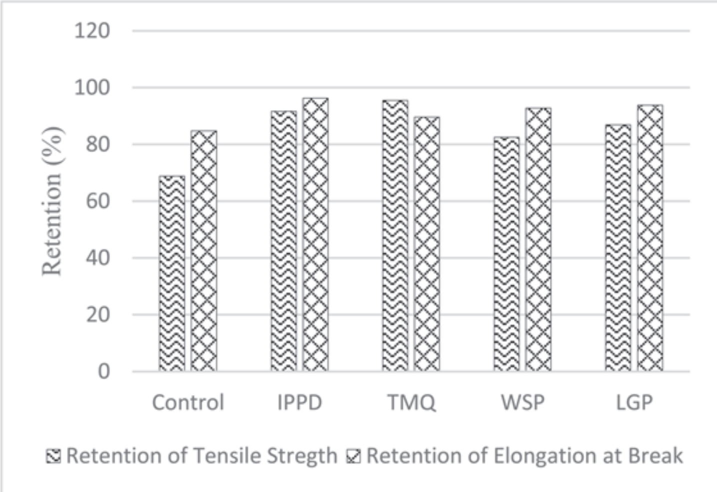

Figure 10. Percentage retention of Tensile strength and Elongation at break of the NR composites after ageing

4. Conclusion

The possibility of using LGP as an alternative AO in NR composites was evaluated in this study. The performance of the LGP incorporated composite was superior to that of the WSP incorporated composite in respect to the cure characteristics maximum torque, scorch time, delta cure and cure rate index. Additionally, the results of tensile strength and elongation at break of the former composite after ageing were higher than those of the latter composite. Further, the LGP incorporated vulcanisate generated favorable results than the control vulcanisate in regard to retention of tensile strength and elongation at break. Hence, it can be concluded from the results generated through the studies on cure characteristics, tensile properties and thermal ageing that LGP has the ability to be substituted as a potential AO for NR composites, especially as an alternative for phenolic AOs such as WSP.

5. References

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