# Weight loss by soil burial degradation of green natural rubber vulcanizates modified by tapioca starch

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ABSTRACT - The weight loss of soil degraded natural rubber vulcanizates modified by tapioca starch was investigated. The samples were prepared by melt compounding using a Haake internal mixer at different tapioca starch loading of 0, 5, 10, 20, 40, and 60 phr. The samples were exposed to soil burial testing for duration of 7, 14, 21 and 28 days. Then, the weight loss was measured using the difference in weight before and after the testing. The mass reduction was observed to be proportionately increased with the increment of tapioca starch loadings and prolonged soil burial duration. The rate of degradations observed was supported with morphological characteristics of the vulcanizates. This study is highly significant towards the development of green natural rubber composites by incorporation of tapioca starch.

# 1. INTRODUCTION

To date, there are growing interest in the use of natural fillers such as starch [1] in rubbers and their blends. The benefits of these fillers include low cost, easy availability, sustainable sources and a greener choice to our environment. Starch is one of the biopolymer substances most widely found in nature and mostly consists of amylose and amylopectin. There are several works of reinforcing elastomers with tapioca starch [2-3], but only a few addressed the degradation of natural rubber vulcanizates. Thus, the aim of this study is to assess the potential utilization of tapioca starch as biodegradability agent in natural rubber formulations. This study is part of our research work to produce biodegradable natural rubber based composites which proven to have diverse applications from general household products to engineering components.

# 2. RESEARCH METHODOLOGY

# 2.1 Materials

Natural rubber (NR) with commercial trade name of 'SMR20' was purchased from Felda Global Ventures Holdings Bhd (FGV). The NR was masticated using a two-roll mill for about 10 min at 30 °C prior to compounding. Carbon black was supplied by Lembaga Getah Malaysia whereas tapioca starch (TS) was

purchased from Polyscientific Enterprise Sdn Bhd. Other compounding ingredients such as sulfur, zinc oxide, stearic acid were purchased from Systerm Classic Chemical Sdn. Bhd. Tetramethylthiuram disulfide (Perkacit-TMTD) was purchased from Aldrich Chemistry, while 6PPD was supplied by Flexys America, USA. All of the other compounding chemicals were used as received without further purification steps.

# 2.2 Sample preparation

The NR was compounded using a Haake internal mixer working at 60°C and a rotor speed of 60 rpm for 7 minutes according to ASTM D-3192. The TS loading was varied (Table 1) and the recipe was based on semi-EV curing [1]. Then, the compounds were subsequently molded into sheets at 160°C and 150 kgf using a hot press model GT7014-A from GoTech [4].

Materials	Compound (phr) <sup>a</sup>
Natural rubber	100
Carbon Black	50
Tapioca starch	0 /5 / 10/ 20 / 40 / 60
<sup>a</sup> Parts per hundred	0/3/10/20/40/00

<sup>b</sup> Tetramethylthiuram disulfide

°(1,3-dimethylbutyl)-N'-phenyl-p-phenylenediamine

# 2.3 Soil degradation test

Biodegradability of the samples in soils was measured from percentage weight loss of the samples [5]. In this study, samples prepared in accordance to ASTM D-412 type C were weighed and buried in natural soil outdoors, approximately at a depth of 10 cm below the surface. Five samples were removed every week for different burial duration of 7 days, 14 days, 21 days and 28 days. After removal, samples were washed in distilled water and dried at 60°C in a vacuum oven for at least 24 hours. The weight of each specimen was measured before  $(W_1)$  and after  $(W_2)$  degradation. The weight loss (WL)was calculated using Equation 1.

Weight Loss (WL) = 
$$\frac{W_1 - W_2}{W_1} \times 100$$
 (1)

## 3. RESULTS AND DISCUSSION

## 3.1 Weight loss

Figure 1 depicts the degradation rate experienced by NR vulcanizates for the effects of tapioca starch loading and soil exposure time. From the results, both factors played significant roles. The samples exhibited pronounced lost in their weight once exposed to almost 2 weeks to the soil degradation which shown by drastic change in the curve. The rate of the degradation started at very low rate of almost 0 %/day to upto 0.21 %/day for sample with 60 phr tapioca starch. The rate of degradation was dramatically increased beyond this point and in some formulations the curve manifested a constant change until 28 days. The degradation rate was accelerated with the amount of tapioca starch present in the samples. It was noted that weight loss was highly proportional to the starch content. As the samples exposed to the moistures, heat and microbes in the soils, the hydrophilic portions of the starch will be degraded, consumed and depicted as weight loss. Therefore, the degradation process of a natural polymer such as tapioca starch is highly complex which involved both climate and biology elements. During the soil burial test, the starch structure is destroyed and the amylopectin and amylase chains degraded [6].



Figure 1 Percentage of weight loss versus time of NR vulcanizates modified by tapioca starch for soil burial degradation test.

## 3.2 Surface morphology

The degradation experienced by samples were explained by the morphological characteristics of the samples exposed to the soil burial testing. Figure 2 shows the morphologies of three selected samples before and after the testing at 500X magnifications via optical microscopy. From the morphology, the white phase is recognized as dispersed tapioca starch in the natural rubber matrix which clumped together during the compounding process. The starch aggregated into larger particles as the loading of starch increased in the samples. The starch aggregates appear smeared with larger sizes in vulcanizates after soil burial testing of 28 days. This was due to the swollen starch particles from reactions with microbes, moistures and other factors in soil. Presence of water promotes the microbe activities which results in molecular degradation of the vulcanizates. The biological degradation process form microbe's enzymes could occur under aerobic and anaerobic conditions, leading to complete or partial removal of components to environment [7].

### 4. CONCLUSION

As the conclusion, it was found that rate of degradation of natural rubber vulcanizates is highly influenced by the loading of tapioca starch into the matrix. Nevertheless, the degradation curves nearly achieved their constant rates after almost 2 to 3 weeks of exposure to soils. This demonstrates the promising potential for sustainable mechanical properties and confirms the biodegradability tendency once in contact with soils. The findings are significant to be exploited for future green rubber composites based products.



Figure 2 The comparison of morphology for before and after exposed to soils for 28 days.

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## REFERENCES

- M. Mazliah, N. Mohamad, A.R. Jeefferie and H.E. Ab Maulod, "cure characteristics and tensile properties of natural rubber vulcanizates modified by tapioca starch," in *Proceedings of Mechanical Engineering Research Day 2016*, 2016, pp. 163-164.
- [2] A.W.M. Kahar and H. Ismail, "High-density polyethylene/natural rubber blends filled with thermoplastic tapioca starch: Physical and isothermal crystallization kinetics study," *J Vinyl* and Additive Technology, vol. 22, no. 3, 2016.
- [3] S. Attharangsan, H. Ismail, M. Abu Bakar and J. Ismail, "Carbon black (CB)/rice husk powder (EHP) hybrid filler-filled natural rubber composites: Effect of cb/rhp ratio on property of the composites," *Polym Plast Technol Eng*, vol.51, no.7, pp.655–662, 2012.

- [4] A.R. Jeefferie, S.H. Ahmad, C.T. Ratnam, M.A. Mahamood and N. Mohamad, "Effects of PEI adsorption on graphene nanoplatelets to the properties of NR/EPDM rubber blend nanocomposites," *J Mater Sci*, vol. 50, pp. 6365 – 6381, 2015.
- [5] Y.H. Lum, A. Shaaban, N.M.M. Mitan, M.F. Dimin, N. Mohamad, N. Hamid and S.M. Se, "Characterization of urea encapsulated by biodegradable starch-PVA-glycerol," *Journal of Polymers and the Environment*, vol. 21, no. 4, pp. 1083-1087, 2013.
- [6] N.I. Miren, A. Carmen, H. Marianella, G. Jeanette and P. Jenny, "Characterization of natural rubber/cassava starch/maleated natural rubber formulations," *Revista Latinoamericana de Metalurgia y Materiales*, vol. 31, no. 1, pp.71-84, 2011.
- [7] K. Leja and G. Lewandowicz G, "Polymer biodegradaion and biodegradable polymers – A review," *Polish Journal of Environmental Studies*, vol. 19, no. 2, pp. 255 – 266, 2010.