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Effect of carbon black-silicon dioxide powder dual filler on rubber isolator properties

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***Abstract* - Silicon dioxide powder (SP) is a waste material from silicon metal industry. The effects of partial blend of carbon black (CB) by silicon dioxide powder (SP) on the cure characteristics, mechanical, dynamic and endurance properties were investigated. Results showed that maximum torque (M_H) and differential torque (M_H-M_L) were decreased, while cure time (t_{90}) and scorch time (t_{S2}) were increased as increment of SP content. For the blend ratio of CB/SP 50:30 phr is showed similar in mechanical properties when compared to in the case of CB only (60:00 phr). The dynamic properties revealed that better elasticity and lower heat buildup with increasing SP content. It is confirmed by the lowering of C and tanδ. The endurance properties of rubber isolator are importance parameter to prove that addition of SP can act as alternative filler in rubber industry. This is due to enhance the number of cycles until to fracture of rubber isolator.**

Index Terms - Natural rubber; Silicon dioxide; Silane coupling agent; Industrial Wastes; Rubber isolator.

INTRODUCTION

Gum rubber cannot use in pure form due to soft and low mechanical properties. Therefore, filler is loaded into rubber for enhance the mechanical properties such as hardness, tensile strength, tear strength, abrasion resistance and others. The types of filler always used in rubber for example, carbon black [1-4], silica [5-8], clay [9-12], calcium carbonate [13-15], etc. In recent years there has been a shift in the focus towards the usage of filler from natural resource and industrial waste. S. Attharangsan et al. [16] studied the rice husk ash (RHA) from rice milling industry. The RHA is a byproduct of combustion the rice husk and convert into ash [17-19]. It contains silicon dioxide about 85-90% but low percent yield due to 25% convert into rice husk ash and 75% of organic volatile matter. The other filler also consists of silicon dioxide and is obtained from industrial wastes. The fly ash (FA) is a coal combustion and collected from the exhaust gases by filter in electric power plant [20-22]. It contains silicon dioxide about 45-50%. The disadvantage of FA is low silicon dioxide content. Thus, the new alternative filler is silicon dioxide powder (SP) which proposed in this work. The SP essentially is a silicon dioxide which derived from silicon metal industry. It is an industrial waste but quite different from FA because higher silicon dioxide content (95% or more) and particle size. The SP is covered with silanol groups on its surface and hence leads to agglomeration in rubber compound, same as silica using. Due to environmental problems and then require proper management to eliminate the waste material from Thailand industry. We aim to reuse of SP as a filler in rubber industry because a primary purpose of waste management and also intend to make the value added products such as rubber isolator in automotive industry. This is likely to be used in the coming future.

The purpose of this research, we attempt to use the SP as filler. TESPT or bis-(3-triethoxysilylpropyl) tetrasulfide is used as one of the silane for surface treatment [23-24]. It is utilized to improve the SP-NR interaction and increase the reinforcing efficiency. In this work, we have studied the use of SP in place of some portion of the carbon black (CB) on their cure characteristics and mechanical properties. Furthermore, the rubber isolator which is reinforced base on blend ratio of CB/SP. The endurance and dynamic properties of rubber isolator such as static spring constant (Ks), dynamic spring constant (Kd), dynamic to static coefficient (η), damping constant (C) and loss factor (tanδ) are also carried out which has not been reported elsewhere. This is not only save the material cost but also waste utilization.

EXPERIMENTAL

I. Materials

The natural rubber (RSS#3) was obtained from Thai Hua Rubber Public Co., Ltd (Rayong, Thailand). TESPT or Si-69 (bis-(3-triethoxysilylpropyl) tetrasulfide) uses as a silane, it was manufactured by Evonik Industries (Essen, Germany) for surface treatment of silicon dioxide powder (SP). Carbon black (N-774) was manufactured by Thai Tokai Carbon Product Co., Ltd. (Chonburi, Thailand). The new alternative filler in this work was SP (average particle size 0.12 µm and specific surface area 14.16 m2/g); it was a waste material from the production of silicon metal industry (Rayong, Thailand). Plasticizer was vivatec 500, supplied by H&R Chem Pharm (Chonburi, Thailand). ZnO (white seal grade) was an activator and supplied by Thai-Lysaght Co., Ltd. (Pathum thani, Thailand), the co-activator was stearic acid which purchased from Chemmin Corporation (Bangkok, Thailand). The antioxidant was TMQ (2,2,4-trimethyl-1,1-dihydroquinoline) and obtained from Kawaguchi Chemical Industry (Japan). The antiozonant was paraffin wax, TBzTD (tetrabenzylthiuram disulfide), NOBS (n-oxydiethylene-2-benzothiazole sulfonamide) and DPG (1,3-diphenylguanidine) use as a rubber accelerator and sulphur were purchased from Sunny World Co., Ltd. (Bangkok, Thailand).

II. Formulations and compounding

Formulations of rubber isolator were showed in Table 1. The content of TESPT and DPG used in formulation were calculated based on the specific surface area of SP according to Eq.1-2 [25],

TESPT content (phr) = 0.00053 × Q × CTAB (1)

DPG content (phr) = 0.00012 × Q × CTAB (2)

where Q was the SP content (phr) and CTAB was the specific surface area of SP (14.16 m2/g). Compound was prepared in a kneader of 3 liters. The natural rubber (RSS#3) was masticated with ZnO and stearic acid in the mixer for 2 minutes. Then, TMQ, Wax, vivatec 500 and rubber filler such as carbon black and SP were also added and mixed with TESPT for 4 minutes. After mixed for further 2 minutes, the masterbatch was dumping out. The rubber accelerator TBzTD, NOBS, DPG and sulphur were added on a two roll mill.

TABLE I
FORMULATIONS OF THE CB/SP RATIO FILLED NATURAL RUBBER

Ingredients	CB:SP (60:0 phr)	CB:SP (50:10 phr)	CB:SP (40:20 phr)	CB:SP (30:30 phr)	CB:SP (0:60 phr)	CB:SP (50:30 phr)
RSS#3	100	100	100	100	100	100
Carbon black (N-774)	60	50	40	30	-	50
Silicon dioxide powder (SP)	-	10	20	30	60	30
Vivatec 500	5	5	5	5	5	5
ZnO	5	5	5	5	5	5
Stearic acid	1	1	1	1	1	1
TMQ	1	1	1	1	1	1
WAX	1	1	1	1	1	1
TBzTD	0.8	0.8	0.8	0.8	0.8	0.8
NOBS	2	2	2	2	2	2
TESPT (Si-69)	-	0.08	0.15	0.23	0.45	0.23
DPG	-	0.02	0.04	0.05	0.10	0.05
Sulphur	1.6	1.6	1.6	1.6	1.6	1.6

III. The cure characteristics and mechanical properties of rubber isolator

The compound was tested at 150°C for 30 minutes with a MDR (model MDR2000, Alpha Technologies, U.S.A). The cure characteristics such as the maximum torque (M_H), the differential torque (M_H-M_L), cure time (t_{90}) and the scorch time (t_{S2}) were determined. The rubber mechanical properties such as hardness (HS), tensile strength (TS), 100% and 300% modulus (M100 and M300), elongation at break (EB), tear strength (TR) were also determined. A universal tensile testing machine (model AG-IS, Shimadzu, Japan) was used for measure tensile strength, elongation atbreak,100% and 300% modulus at 23 ± 2°C with a dumbbell shaped samples, the testing speed was 500 mm/min follow ASTMD412 method. The tear strength was measured follow ASTM D624 method using a type-C die. The hardness test was investigated using a hardness tester (model GS-719H, Teclock, Japan) follow ASTM D2240 method.

TABLE II
THE EFFECT OF CB/SP RATIO FILLED NATURAL RUBBER ON MECHANICAL PROPERTIES

CB/SP blend ratio	Hardness (Shore A)	Tensile strength (MPa)	100% modulus (MPa)	300% modulus (MPa)	Tear strength (N/mm)	Elongation at break (%)
60:00 phr	66 ± 0.54	22.31 ± 0.59	4.03 ± 0.03	15.90 ± 0.14	64.52 ± 0.54	430 ± 33
50:10 phr	63 ± 0.62	20.22 ± 0.76	3.46 ± 0.04	14.13 ± 0.16	62.93 ± 0.48	436 ± 32
40:20 phr	61 ± 0.51	19.81 ± 0.61	3.08 ± 0.06	12.84 ± 0.12	58.91 ± 0.59	442 ± 42
30:30 phr	59 ± 0.43	19.40 ± 0.62	2.76 ± 0.04	11.89 ± 0.18	57.83 ± 0.58	455 ± 28
00:60 phr	53 ± 0.50	16.30 ± 0.59	1.96 ± 0.05	8.60 ± 0.15	55.76 ± 0.60	480 ± 40
50:30 phr	66 ± 0.49	20.13 ± 0.57	3.89 ± 0.03	15.45 ± 0.16	62.53 ± 0.62	420 ± 45

TABLE II
THE EFFECT OF CB/SP RATIO FILLED NATURAL RUBBER ON DYNAMIC PROPERTIES

CB/SP blend ratio	Static spring constant Ks (N/mm)	Dynamic spring constant Kd (N/mm)	Dynamic to Static coefficient η	Damping constan C (N.s/mm)	Loss factor tan δ
60:00 phr	72.93 ± 0.64	109.20 ± 0.74	1.497 ± 0.016	0.176 ± 0.001	0.092 ± 0.003
50:10 phr	68.38 ± 0.52	100.10 ± 0.63	1.463 ± 0.012	0.119 ± 0.003	0.074 ± 0.005
40:20 phr	63.18 ± 0.48	90.50 ± 0.68	1.432 ± 0.011	0.094 ± 0.001	0.067 ± 0.002
30:30 phr	59.33 ± 0.52	81.50 ± 0.65	1.373 ± 0.013	0.063 ± 0.002	0.052 ± 0.003
00:60 phr	53.00 ± 0.61	71.50 ± 0.58	1.349 ± 0.013	0.049 ± 0.002	0.032 ± 0.004
50:30 phr	71.30 ± 0.63	108.20 ± 0.49	1.517 ± 0.015	0.177 ± 0.003	0.094 ± 0.003

IV. The dynamic properties of rubber isolator

The rubber isolator was vulcanized by compression molding at 150°C based on the cure time (t90) plus 12 minutes. The dynamic properties of rubber isolator (static spring constant (Ks), dynamic spring constant (Kd), dynamic to static coefficient (η), damping constant (C) and loss factor (tanδ)) were carried out. For Ks testing, it was examined at 23 ± 2°C by using an autograph machine (model AGS-X, Shimadzu, Japan). The testing load was applied in the range of 0-1.50 kN on the rubber isolator but calculation the Ks in the range of 0.50 to 1.50 kN according to Eq.3

$$Ks = \frac{F_2 - F_1}{D_2 - D_1} \quad (N/mm) \qquad (3)$$

where Ks was static spring constant (N/mm)

F1 was load at 0.50 (kN), F2 is load at 1.50 (kN)

D1 was lower displacement (mm), D2 was upper displacement (mm)

In the case of Kd, C and tanδ testing, it was measured with a rubber isolator dynamic characteristic tester (model V-157, Sum Electro Mechanics, Japan) at 23 ± 2°C. A sinusoidal mechanical excitation of the rubber isolator was executed whereby force at 15 Hz with amplitude held constant at 0.5 mm. The Kd, C and tanδ were reported and calculated according to Eq.4-7

$$Kd = \left|K^*\right| \cos\delta \quad (N/mm) \qquad (4)$$

$$Ki = \left|K^*\right| \sin\delta \quad (N/mm) \qquad (5)$$

$$1 = \tan\delta = \frac{Ki}{Kd} \qquad (6)$$

$$C = \frac{Ki}{\omega} \quad (N.s/mm) \qquad (7)$$

where Kd was dynamic or storage spring constant (N/mm)

Ki was loss spring constant (N/mm)

|K*| was absolute value of complex spring constant (N/mm)

tanδ was loss factor

δ was loss angle (rad)

C was damping constant (N.s/mm)

ω was angular velocity (rad/s)

V. The endurance property of rubber isolator

Endurance testing of rubber isolator was tested with an endurance tester (model TV-731, Sum Electro Mechanics, Japan). Rubber isolator was placed on the jig and applied the load of 13 kN at 1.5 Hz. For the evaluation of sample, stop testing when found crack on surface of product.

RESULTS AND DISCUSSION

I. The cure characteristics of rubber isolator

Figure 1 is a rheograph which represents the effect of CB/SP ratio filled natural rubber on cure characteristics. The test result shows that maximum torque (M_H) and differential torque (M_H-M_L) are significantly decreased with increasing SP content. This is due to the higher average particle size and lower surface area of SP than CB. As the increasing partial blend of CB by SP leading to gradually reduced stiffness. Therefore, the SP increases mobility of the segment of natural rubber chain and affect to flow easily in the mold. Moreover, it also can be seen that an increase in SP content increased the cure time (t_{90}) and scorch time (t_{S2}) of the rubber compound. This is known that the SP has silanol groups on its surface and caused to adsorption of rubber accelerator [26]. So, it makes the cure retardation. It should be noted that the advantage of SP usage is scorch safety. From all test results, especially in the case of CB/SP (50:30 phr) shows M_H and M_H-M_L are similar to that observe when CB only (60:00 phr) is filled into natural rubber. It is indicated that can provide equivalent cure state in the rubber compound, as see previously cure curves. In particular, M_H-M_L value can be the evidence to support the degree of crosslinking in the rubber compound [27]. This tends to the higher reinforcement of vulcanized rubber. Whereas, t_{90} and t_{S2} of CB/SP (50:30 phr) is slower than CB only (60:00 phr). It is the same tendency observed when increase SP content.

II. The mechanical properties of rubber isolator

The influence of CB/SP blend ratio on mechanical properties of natural rubber vulcanizates is showed in Table 2. It can be showed that hardness (HS) is decreased with increasing SP content in the partial blend of CB/SP ratio. This is possibly due to the particle size of SP bigger (or lower specific surface area) than CB, as mentioned earlier. Thus, it provides lower interaction to natural rubber than CB. As expected, the filler reinforcement mainly depends on the particle size level. This explains why natural rubber filled with CB only (60:00 phr) enhances the hardness more than filled with SP only (00:60 phr). Moreover, the effect of CB/SP ratio on tensile strength (TS), tear strength (TR), 100% modulus (M100) and 300% modulus (M300) are also evaluated. Test results are indicated that TS, TR, M100 and M300 show decreasing trend as SP content increased. A decreasing trend of TS and TR, it may be due to significant reduction of crosslink density in the rubber chain. The M_H-M_L is an evidence to support this explanation because of measures the degree of crosslink density. For the results of M100 and M300, it is believed that correspond to M_H value [28]. The natural rubber vulcanizates show decrease not only hardness but also stiffness when increase the SP content. This shows clearly result in reduction of both modulus (M100 and M300) [29]. Whereas, elongation at break (EB) is gradually increased with increasing SP content. It is contributed to the SP particles and leading to enhance rubber chain mobility. Because at higher SP content, there is more gap between rubber and filler compare to CB which becomes more elastic and flexible. In addition, natural rubber filled with CB/SP (50:30 phr) provides the similar mechanical properties which is compared to that in the case of CB only (60:00 phr). This is attributed to the same content of concentration of crosslink density, as showed in the vulcanization curves. From all the test results of CB/SP dual phase filler have been studied, it is clear that SP can partial replacement to CB in the ratio of 50:30 phr.

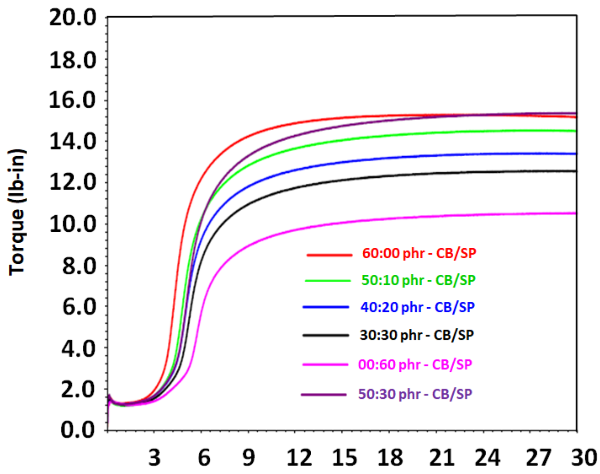

FIGURE I
THE EFFECT OF CB/SP RATIO FILLED NATURAL RUBBER ON CURE CHARACTERISTICS

III. The dynamic properties of rubber isolator

Figure 2 shows the relationship between compressive force and displacement with different CB/SP ratio. The change in the slope of each CB/SP ratio is static spring constant (Ks) and directly related to HS value; as showed before. Ks of rubber isolator is decreased with increasing SP content. This is led to a decrease in stiffness of rubber isolator. In addition, the variation of dynamic spring constant (Kd) with frequency sweep from 0.1 Hz to 100 Hz at different CB/SP ratio is illustrated in Figure 3. Kd of rubber isolator is also decreased with increasing SP content; same as Ks results. Meanwhile, the Kd of all blend ratio increases when frequency increases, rapidly at low frequency, slowly at high frequency. It can be explained by the rubber molecules will resist quickly motion in high frequency state more than low frequency state. Then, the Kd depends on stiffness and frequency of the applied displacement. Furthermore, as seen from Table 3 is summarized the dynamic properties of rubber isolator obtained from rubber isolator dynamic characteristic tester at 15 Hz with amplitude held constant 0.5 mm. As a result, the increment of SP content in blend ratio of CB/SP has a significant effect on the Ks, Kd and dynamic to static coefficient (η). The Ks, Kd and η appeared to decrease with increasing SP content. It is indicated that the SP reinforced natural rubber enhances elastic property more than CB. This is mainly due to the SP has a particle size bigger than CB, so the addition of SP in the rubber isolator plays the important role to provide better flexibility. Moreover, damping constant (C) and loss factor (tanδ) are always used for determining the damping and hysteresis (or heat buildup) of the rubber isolator, respectively. The decreasing trend of C and tanδ when gradually increases the SP content in the rubber matrix. It is an evidence to show that low damping and heat buildup of the rubber isolator. This is possibly due to it enhances the mobility of molecular chains of natural rubber. Hence, the partial blending of CB by SP introduces an amorphous phase. From our research, we believe that natural rubber filled with CB/SP (50:30 phr) is the optimum loading and same the dynamic properties when compared to filled with CB only (60:00 phr).

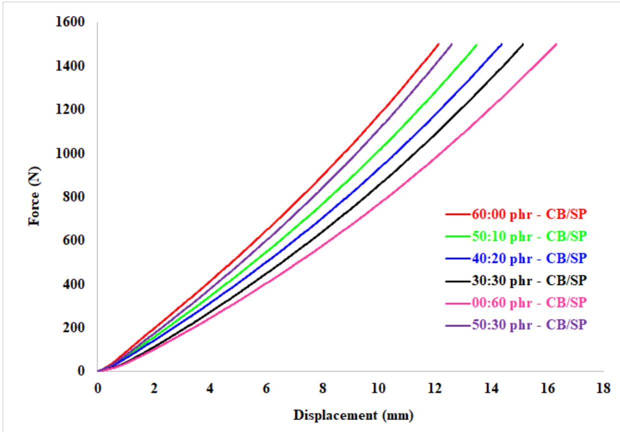

FIGURE II
THE RELATIONSHIP BETWEEN COMPRESSIVE FORCE AND DISPLACEMENT WITH CB/SP RATIO FILLED NATURAL RUBBER

IV. The endurance property of rubber isolator

The aim of this part is focus on rubber product evaluation. An experimental investigation to comparatively study the blend ratio of CB/SP on endurance property of rubber isolator. It can be seen in Figure 4, the number of cycles until to fracture of rubber isolator is rapidly decreased with increasing SP content in blend ratio of CB/SP. According to the theory of rubber reinforcement. It can be explained by two reasons. The first one is probably due to HS of rubber isolator. This attribute to the microsize particles of SP bigger than CB. So, it is easily deformed and influence to breakdown as the HS decrement. The second reason, it is affected from stiffness of rubber isolator. The SP has strong SP-SP interaction due to silanol group on its surface and hence, a decrease in SP–rubber interaction [30]. This result in decreases in crosslink density and stiffness. These reasons are supported by the value of HS and modulus (M100 and M300) which is also illustrated in Table 2. In order to aid in the endurance property, we found that rubber isolator filled with CB/SP (50:30 phr) enhances the number of cycles until to fracture more than CB/SP (50:10 phr). It is significantly change from 295,182 to 319,022 cycles until to fracture, so increased by 8.07%. However, the relatively small difference in the number of cycles until to fracture of the rubber isolator containing with CB/SP (50:30 phr) compared to CB only (60:00 phr) due to same HS and stiffness level. This implies that the increment of SP by 25% content in blend ratio causes significant enhance endurance property.

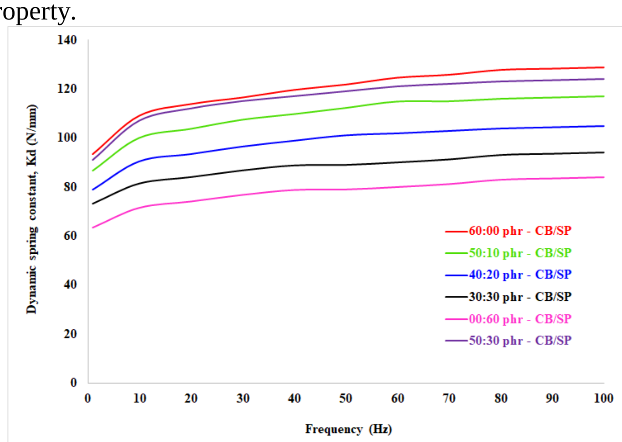

FIGURE III
THE RELATIONSHIP BETWEEN DYNAMIC SPRING CONSTANT AND FREQUENCY WITH CB/SP RATIO FILLED NATURAL RUBBER

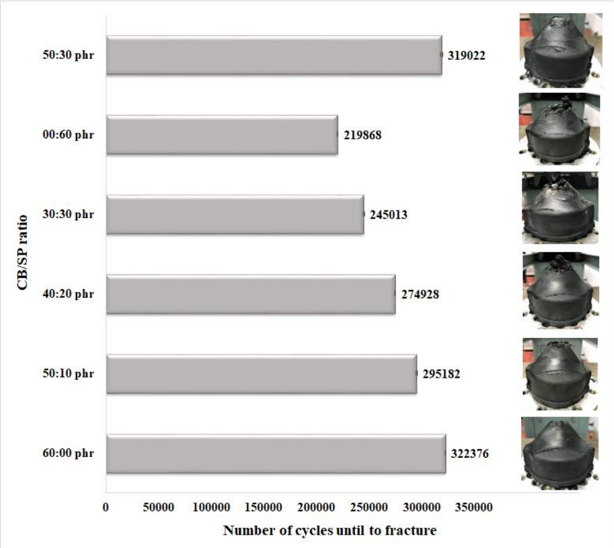

FIGURE IV
THE EFFECT OF CB/SP RATIO FILLED NATURAL RUBBER ON ENDURANCE PROPERTIES

CONCLUSIONS

This work is aimed to use the SP as filler. It can be concluded that SP is affected to cure characteristics, mechanical, endurance and dynamic properties. The partial CB replacement by SP imparts to reduce stiffness and cure retardation. As expected, the mechanical properties such as HS, TS, TR, M100 and M300 are decreased steadily whereas EB is increased with increasing SP content due to microsize particles. However, for dynamic properties of rubber isolator, the increment of SP result in decrease the Ks, Kd, η, C and tanδ. This is indicated that enhance the elastic property and reduce heat buildup. At the same time, the endurance property of rubber isolator is also carried out. Most of the rubber isolators show a more damage when SP increased. It may be attributed to decrease stiffness of the rubber isolator. Then, it is possible to use the SP as alternative filler when an increase of SP by 25% content in blend ratio for improve all rubber properties.

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