PHYSICO - MECHANICAL PROPERTIES OF NBR/SBR BLENDS

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ABSTRACT

Three compatibilizing agents (maleic anhydride, styrene maleic anhydride and styrene butyl acrylate) have been added to NBR/SBR blends to improve the phase morphology and interfacial adhesion, thereby improving the physical & mechanical properties of the blend. The physico-mechanical properties such as rheological properties, tensile stress and sorption properties were examined, for different formulae of NBR/SBR blends. The sorption of toluene into non-compatibilized NBR/SBR blends was studied. Pure SBR and 50/50 blend NBR/SBR showed the minimum sorption and the highest mechanical properties. Three different compatibilizers where added at a different levels to 50/50 NBR/SBR blend and their effect on swelling diffusion coefficient, transport mechanism, tensile stress and elongation at break were studied. It observed that the rheological properties are not affected with the kind of compatibilizer or its doses. the best value of tensile strength and elongation percentage at the break can be obtained when add 0.6 phr of maleic anhydride, 4 phr of styrene maleic anhydride and 4 phr of styrene butyl acrylate as a compatibilizer with NBR/SBR 50/50. Less sorption of toluene is showed with high amount of maleic anhydride or styrene maleic anhydride as a compatibilizer, and also when used butyl acrylate as a compatibilizer at 2 phr. Diffusivity reduced at the higher doses of either maleic anhydride or styrene maleic anhydride in the blend and it increased at a dose of 6 phr of styrene butyl acrylate as a compatibilizer.

Keywords: rubber, maleic anhydride, styrene maleic anhydride, styrene butyl acrylate.

1. INTRODUCTION

Blending of elastomers used to enhance the performance of rubber products. Blending of rubbers is the easiest method to create new polymeric materials with superior properties. Chemical differences between various types of rubbers can sometimes prevent them from blending well. Use different mixing techniques or special additives such as certain process aids, homogenizers, or compatibilizers [1] can achieve a better blend. Compounding of elastomers is the operation of bringing together all the ingredients required to mix a batch of rubber compound. Compounded elastomers have many unique characteristics not found in other materials, such as high elasticity and abrasion resistance. Blending of two incompatible polymers such as SBR and NBR yields a material with poor mechanical properties, adding suitable compatibilizer can solve this problem. Using of compatibilizers help in reduce the macroscopic in-homogeneities and improve the morphological stability by decreasing the interfacial tension [2-6]. In the present paper NBR/SBR blend have been prepared with different formula in the presence and absence of the compatibilizer. It was observed that the blend of 50/50 NBR/SBR has the best physico-mechanical properties. Then the blend of 50/50 NBR/SBR with different amount of the three examined compatibilizers were investigated.

2. EXPERIMENTAL

Materials

I. Rubber:

- **Acrylonitrile Butadiene Rubber (NBR):** Acrylonitrile content: 38%, Moony viscosity a 100°C: 45, Specific gravity: 1.17, Ash content: 0.5% (Bayer Chemicals).
- **Styrene Butadiene Rubber (SBR):** Styrene content: 33.5%, Moony viscosity a 100°C: 38, Specific gravity: 0.98, Ash content: 0.5% (Bayer Chemicals).

II. Plasticizers: Dibutyl Phthalate (DBP): Boiling point 220-248°C and specific gravity 0.983 –0.989, (Aldrich Company).

III. Curing Agent: Sulfur: Pale yellow powder of sulfur element, purity 99.9%, melting point112°C, specific gravity of 2.04-2.06.

IIII. Compatibilizers:

- **Maleic Anhydride:** Molecular formula $C_4H_2O_3$, Mwt. 98.06g/mol, m.p 52.6°C, B.p. 202°C, density 1.48g/cm^3 (Aldrich Company).
- **Styrene Maleic Anhydride:** Molecular formula $C_{10}H_{10}O_3$, sp. gr. 1.05-1.34 at °C, white powder (Aldrich Company).
- **Styrene Butyl Acrylate: Butyl acrylate:** Mwt. 128.17 g/mol, specific gravity 0.9015, viscosity 0.9 cp, boiling point 147°C, melting point -64°C (Aldrich Company). **Styrene:** Colorless oily liquid, Mwt. 104.15g/mol, m.p 52.6°C, density 0.909 g/cm^3, viscosity 0.762 cp, melting point -300°C, boiling point 145°C (Aldrich Company).

2.2 Preparation of Rubber Blends

The used formulae in preparing the rubber blends are shown in Tables 1, 2, 3 and 4. Weighed amounts of the compounding ingredients with the exception of

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vulcanizing agent and accelerators were first introduced into a mixer for 20 minutes for the mastication of each ingredient, and the rubber blend produced was later transferred to a two-roll mill which converted it from an irregularly shaped mass to suitable sheets. Then introduced the vulcanization agent, accelerator, compatibilizer in appropriate ratios as shown in Tables 1, 2, 3 and 4. After compounding, rubber stocks were left overnight (24h) before vulcanization.

Table-1. Composition of Different Rubber Blends without Compatibilizers.

Sample No.	1	2	3	4	5
NBR	100	75	50	25	0
SBR	0	25	50	75	100
DBP (phr)	3	3	3	3	3
Peroxide (phr)	3	3	3	3	3

Table-2. Composition of NBR/SBR (50/50) Blends with Maleic Anhydride.

Sample No.	6	7	8	9
NBR/SBR (50/50)	100	100	100	100
Maleic Anhydride (phr)	0.1	0.3	0.6	0.9
DBP (phr)	3	3	3	3
S (phr)	2.5	2.5	2.5	2.5

Table-3. Composition of NBR/SBR (50/50) Blends with Styrene Maleic Anhydride.

Sample No.	10	11	12	13
NBR/SBR (50/50)	100	100	100	100
Styrene Maleic Anhydride (phr)	2	4	6	8
DBP (phr)	3	3	3	3
S (phr)	2.5	2.5	2.5	2.5

Table-4. Composition of NBR/SBR (50/50) Blends with Styrene Butyl Acrylate.

Sample No.	14	15	16	17
NBR/SBR (50/50)	100	100	100	100
Styrene Butyl Acrylate	2	4	6	8
DPB (phr)	3	3	3	3
S	2.5	2.5	2.5	2.5

2.3 Rheometric Characteristics

Rheology is the science of deformation and flow of mass. Rheometer is equipment helps to choose the right material and its appropriate dose to meet the end requirements of the final product in the rubber industry. For the prepared samples the cure characteristics M_L (the minimum torque), M_H (the maximum torque), tc_{90} (the optimum cure time) and ts_2 (the scorch time were determined using a Monsanto Oscillating Disc Rheometer R-100, at 150 ±2 °C in according to ASTM method D-2084 [7-8].

2.4 Mechanical Properties

Tensile strength and elongation at break were measured with a Zwick-1445 tensile tester according to the ASTM D-412-06A (2013) standard testing method using a crosshead speed of 500 mm/min and at 25 °C. For this experiment standard dumbbell, shape specimens as shown in Figure 1 were cut from a 2 mm thick molded sheet. Each vulcanized sheet was cut out at four individual dumbbell shaped samples using a steel die of a constant width 4mm.

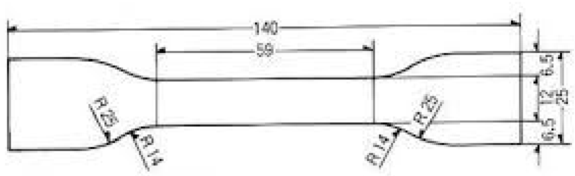

Figure-1. Standard Dumbbell Shape Specimen for Tensile Strength.

2.5 Swelling

First, cured test samples of 20x20x2 mm dimension were accurately weighted ($w_\circ$) then immersed in cylindrical beakers containing toluene at room temperature (25°C). At regular intervals, the tested samples were removed from the toluene and dried rapidly by filter papers to remove the excess amount of the solvent present on the samples' surface. Then weighed again (*wt*) at different time intervals, every time removed from the toluene in less than 40 seconds to minimize the error due to evaporation of toluene from the sample. The weighing was continued till equilibrium swelling was achieved. These procedures were further repeated on rubber samples, which contain different amounts of different compatibilizers maleic anhydride, styrene maleic anhydride and styrene butyl acrylate [9-10].

The percentage of swelling weight for different blend ratios was calculated by using the following equation (1):

$$\%\text{Swelling} = [(w_t - w_\circ) / w_\circ] \times 100 \qquad (1)$$

Where, $w_\circ$ is the initial weight of the blend sample and w_t is the weight of the blend sample at certain time (*t*) [11].

2.5.1 Diffusion Coefficient (D)

The diffusion coefficient of a solvent molecule through a polymer membrane was calculated using equation (2), Fickian's second law of diffusion [9-10].

$$D = \pi \left(\frac{h\theta}{4Q_\infty} \right)^2 \quad (2)$$

Where h is the vulcanizate thickness, θ is the slope of the initial linear portion of the plot of the molar percentage uptake (Qt), which expresses the absorption data of toluene into NBR/SBR blends, against $\sqrt{t}$, and $Q\infty$ is the equilibrium absorption. The molar percentage uptakes (Qt) of toluene per gram of NBR/SBR blends were calculated using the following equation (3):

$$Qt = \frac{\text{Mass of toluene absorbed}}{(\text{Molecular weight of toluene / Initial mass of the blend})} \text{ x } 100 \quad (3)$$

Then, the molar percentage uptake (Q_t) at any particular temperature was plotted against the square root of time ($\sqrt{t}$) [9-10].

2.5.2 Transport Mechanism

In order to study the mechanism of transport phenomenon the sorption data have been fitted into the equation (4).

$$\log\left(\frac{\mathrm{Qt}}{\mathrm{Q\infty}}\right) = \log(\mathrm{K}) + \mathrm{n}\log(\mathrm{t}) \quad (4)$$

Where Qt is the swelling quotient at time t, and $Q\infty$ the equilibrium swelling. K is a constant which depends on the polymer morphology and the polymer-solvent interaction the value of n determines the mode of transport of toluene through the NBR/SBR blends.

First, for normal Fickian mode of transport dominates when n approximately equals 0.5 the rate of polymer chain relaxation is higher compared to the diffusion of penetrating liquid. Second case when n equals 1 the transport approaches non-Fickian behavior where chain relaxation is slower than the liquid diffusion, so it indicates relaxation controlled transport. The third case when n lies between 0.5 and 1, the mechanism of transport called anomalous transport behavior. Finally when the solvent penetration is much below the polymer chain relaxation it is possible to record the n because it is less than 0.5, this mode is named "Less Fickian", or "Quasi Fickian". This mechanism indicates that the solvent diffuses slowly, through the swollen matrix and free spaces in the blend [9].

3. RESULTS AND DISCUSSIONS

3.1 Rheometric Characteristics

Minimum torque (M_L), Maximum torque(M_H), scorch time (t_{S2}), and optimum cure time (t_{C90}) obtained from the experimental data for the different samples are shown in Table-5 the values of minimum torque of the different samples with different kind and doses of compatibilizer are ranging from (1.25 to 1.32), while the values of maximum torque are ranging from (2.5 to 2.7). Minutes to $x\%$ of torque increase; Optimum cure time" obtained from the experimental data for the different samples are ranging from 10 to 14 while the scorch time ranged from (2.8 to 3.4). From the previous results, we can conclude that the presence of compatibilizer does not much affect the rheometric properties of the obtained samples.

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Table-5. Values of M_H, ML,t_{C90}andt_{S2}for50/50 NBR/SBR with Different Doses of Compatibilizers.

Blend	Compatibilizer	phr	M_H	M_L	t_{C90}	t_{S2}
75/25 NBR/SBR	-	-	2.5	1.32	11	3
50/50 NBR/SBR			2.5	1.25	12	2.8
25/75 NBR/SBR			2.7	1.3	10	2.8
50/50 NBR/SBR	Maleic Anhydride	0.1	2.57	1.3	10	3
		0.3	2.62	1.31	11	3
		0.6	2.7	1.3	10	3.2
		0.9	2.6	1.26	10	3.2
	Styrene Maleic Anhydride	2	2.52	1.28	14	3
		4	2.6	1.3	12	3.2
		6	2.6	1.31	12	3
		8	2.53	1.26	12	3.4
	Styrene Butyl Acrylate	2	2.6	1.31	11	3.3
		4	2.63	1.27	12	3.1
		6	2.54	1.28	11	3
		8	2.6	1.32	13	3

3.2 Tensile Strength and Elongation

The dependence of the tensile strength and elongation percent on the amount of NBR in the blend is illustrated by Figures (2 - 3) It is clear that, the tensile strength and elongation percentage at the break increases as the amount of NBR increases until it reaches its optimal value at 50/50 NBR/SBR ratio and then it decreases with increasing of SBR amount in the blend. It is also clear that the 50/50 NBR/SBR blend gives the maximum value of tensile strength and elongation percentage so that this blend possesses the superior mechanical properties. As a result, this blend formula was chosen for further investigations.

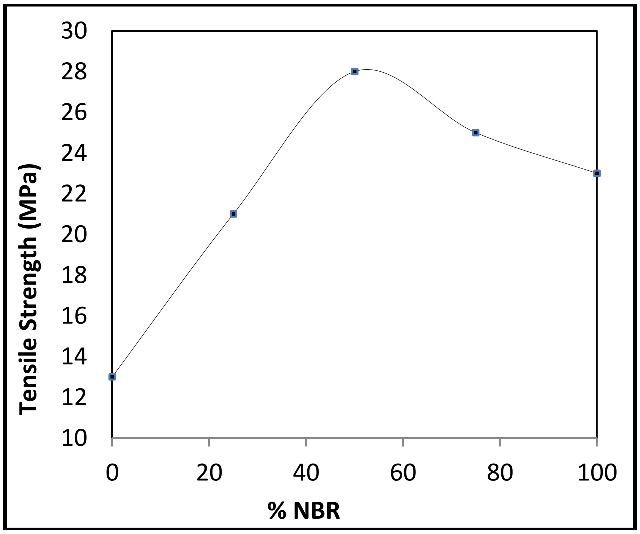

Figure-2. Tensile strength for Different Formula of NBR/SBR Blend without Compatibilizers.

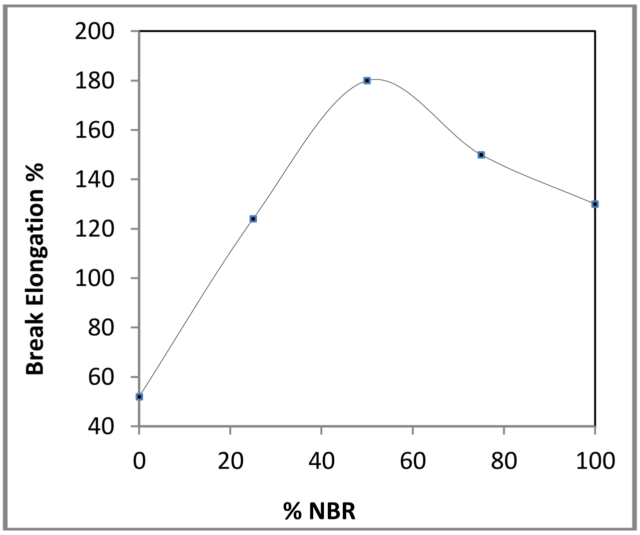

Figure-3. Elongation at Break % for Different Formula of NBR/SBR Blend without Compatibilizers.

The effect of loading the NBR/SBR 50/50 sample with different amount of maleic anhydride (0.1, 0.3, 0.6 and 0.9) phr on the tensile strength and elongation percentage at break is illustrated in Figures 4-5. The tensile strength and elongation percentage verifying with the different doses of maleic anhydride, it shows its superior value at 0.6 phr of maleic anhydride.

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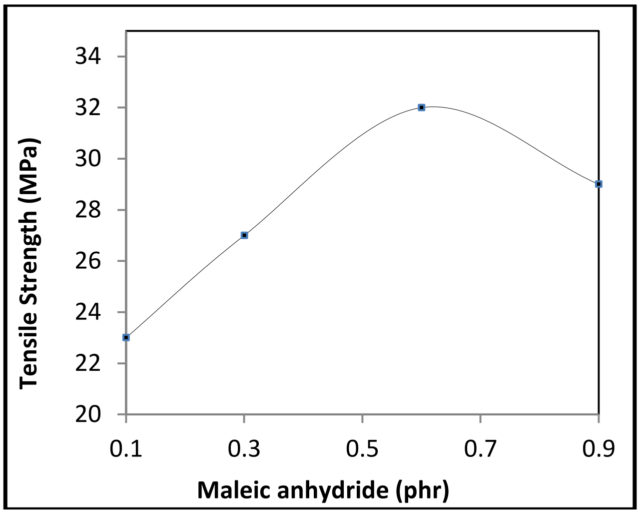

Figure-4. Tensile Strength of 50/50 NBR/SBR Blend with Different amount of Maleic Anhydride.

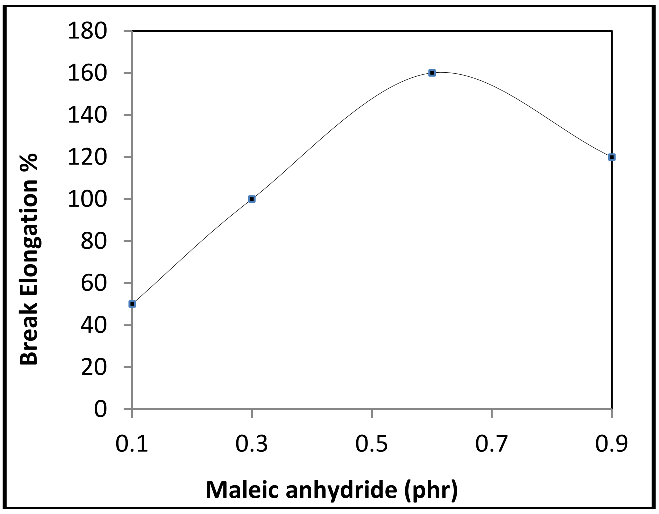

Figure-5. Break Elongation % of 50/50 NBR/SBR Blend with Different amount of Maleic Anhydride.

The variation of tensile strength and elongation at break % with the different doses of styrene maleic anhydride (2, 4, 6 and 8) phr as a compatibilizer is illustrated in Figures 6-7. From the Figs., it is clear that, the tensile strength and elongation percentage at the break at 4 phr is the superior.

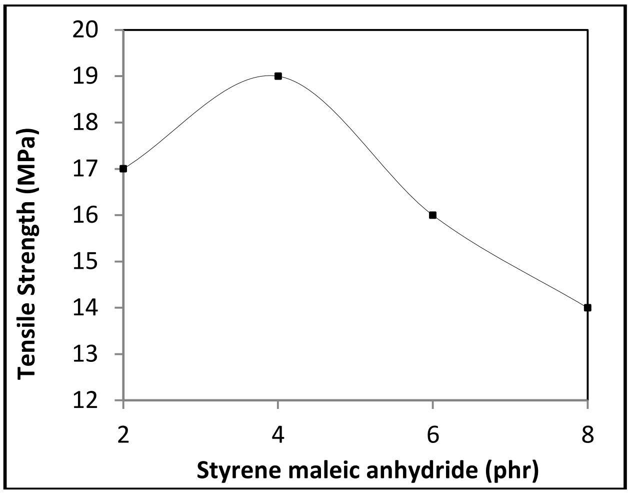

Figure-6. Tensile Strength of 50/50 NBR/SBR Blend with Different amount of Styrene Maleic Anhydride.

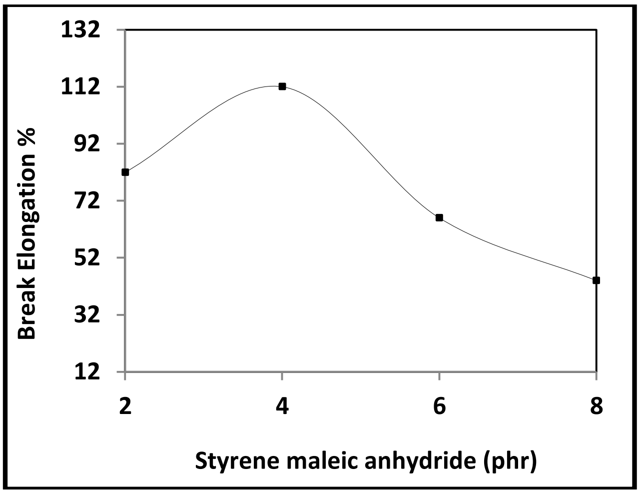

Figure-7. Break Elongation % of 50/50 NBR/SBR Blend with Different amount of Styrene Maleic Anhydride.

The last used compatibilizer is styrene butyl acrylate with different doses (2, 4, 6 and 8) phr. The variations in tensile strength and elongation at break % vs. the different doses of styrene butyl acrylate are shown in Figures 8-9. It is clear that, the tensile strength and elongation elongation percentage at the break increase as the amount of styrene butyl acrylate increases until it reaches its optimal value at 4phr styrene butyl acrylate then it decreases with increasing of styrene butyl acrylate amount in the blend. From the previous results, it is clear that each compatibilizer posses an optimal dose beyond which loss of mechanical properties was observed, this can be explained by the fact of phase separation and disintegration at higher doses takes place [12].

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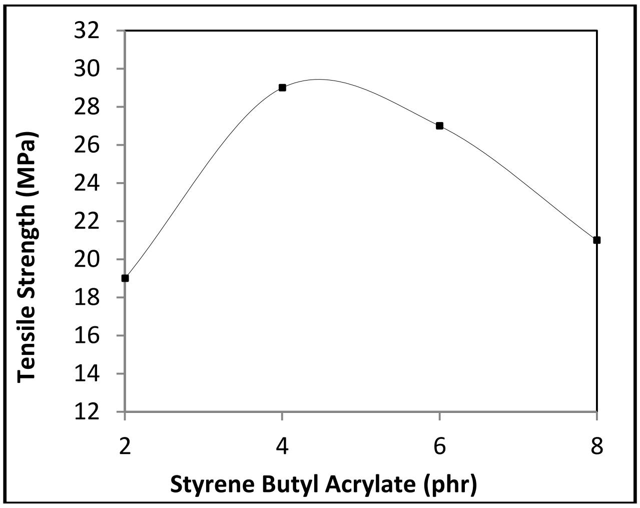

Figure-8. Tensile Strength of 50/50 NBR/SBR Blend Different amount of Styrene Butyl Acrylate.

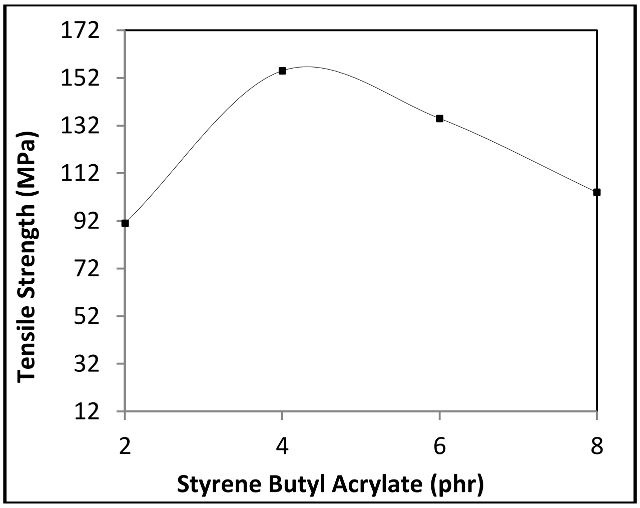

Figure-9. Break Elongation % of 50/50 NBR/SBR Blend with Different amount of Styrene Butyl Acrylate.

3.3 Sorption Characteristic

Figures 10-13 show plots of swelling persentage against the time of exposure to solvent with different percentage of NBR and SBR, and the blend of NBR/SBR (50/50) with different compatibilizers (Maleic anhydride, Styrene maleic anhydrideand Styrene butyl acrylate). Figure-10 showed that the sorption of toluene into NBR/SBR blends without compatibilizer. Pure NBR and 50/50 blend NBR/SBR showed the minimum sorption yet. Pure SBR has the highest value of sorption. The reason for choosing a 50/50 blend to be the target sample for studying the effect of compatibilizer addition was its superior mechanical properties. In Figure-11 after adding different amount of maleic anhydride as a compatibilizer, it was observed that more amount of maleic anhydride lead to less sorption of toluene. The blend of 50/50 NBR/SBR and 0.9 phr maleic anhydride showed the less sorption of toluene. Added of styrene maleic anhydride as a compatibilizer in different amount (2, 4, 6 and 8phr) to the (50/50) NBR/SBR blend are shown in Figure-12. It was observed that more dose of styrene maleic anhydride lead to less sorption of toluene in the blend. The styrene butyl acrylate was added to the blend with different amount (2, 4, 6 and 8). It is obvious from Figure-13 that the swelling of toluene show the less value at the 6 phr dose of styrene butyl acrylate in the blend.

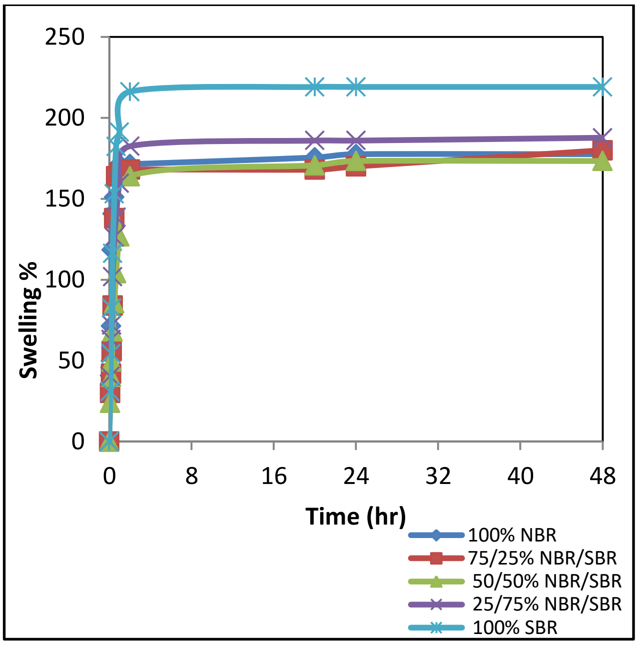

Figure-10. Swelling vs Time for NBR/SBR without Compatibilizer.

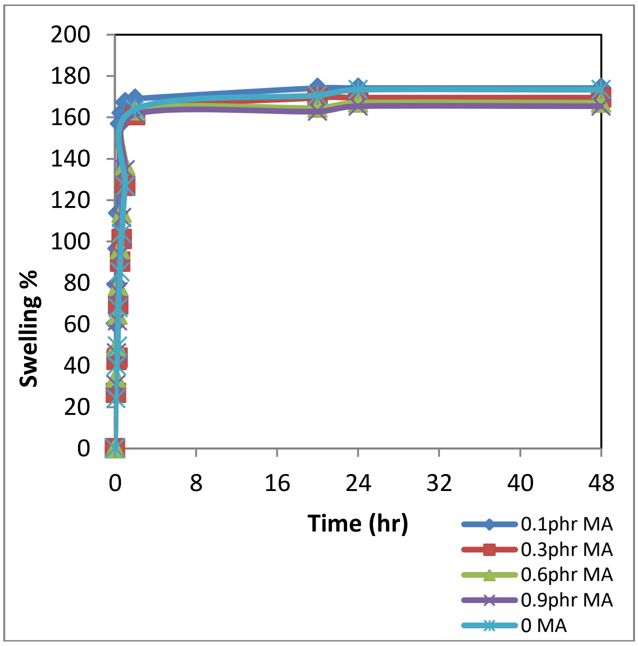

Figure-11. Swelling vs Time for NBR/SBR with Maleic Anhydride.

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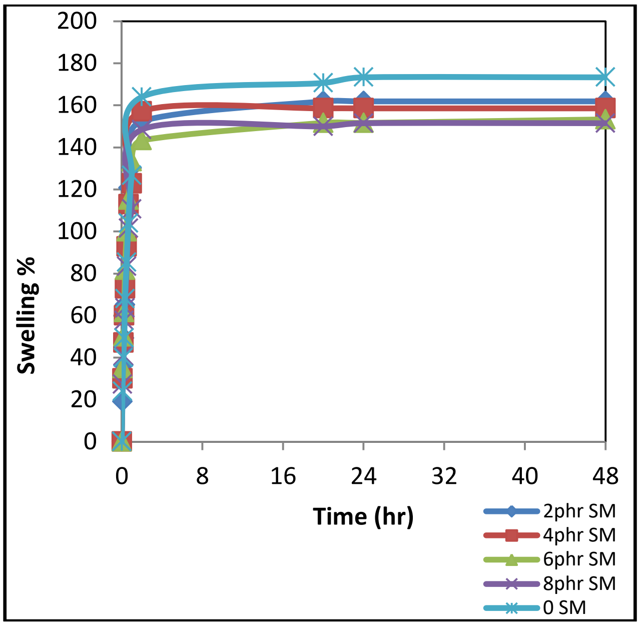

Figure-12. Swelling vs Time for NBR/SBRStyrene Maleic Anhydride.

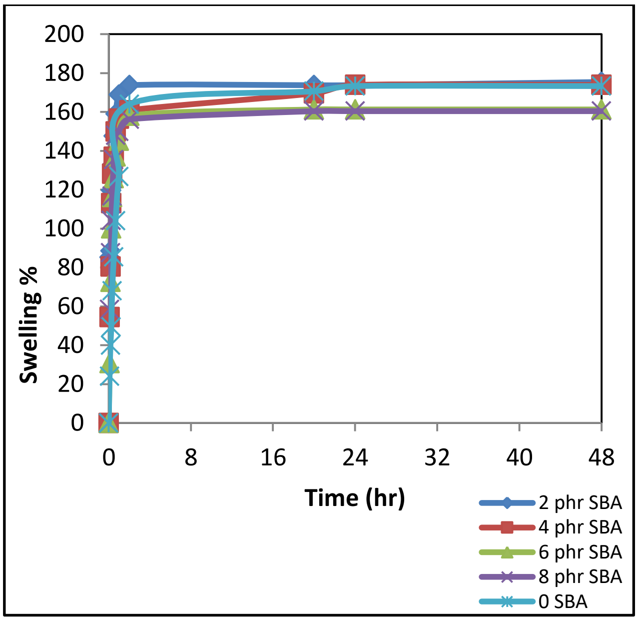

Figure-13. Swelling vs Time for NBR/SBR with Styrene Butyl Acrylate.

3.3.1 Diffusion Coefficient (*D*)

The molar percentage uptake (Q_t) at any particular temperature was plotted against the square root of time ($\sqrt{t}$) as shown in Figures 14 to 17, Figure-14 shows (Qt) vs ($\sqrt{t}$) for NBR/SBR blends without compatibilizer, Figures 15,16,17 show (Qt) vs ($\sqrt{t}$) for a different dose of the using compatibilizer maleic anhydride, styrene maleic anhydride and styrene butyl acrylate consecutively. The Figs. show initial increases in the mass of the absorbed toluene in the NBR/SBR blends until reached to the maximum absorption once the mass of the absorbed toluene remained constant, that is equilibrium as absorption was attained.

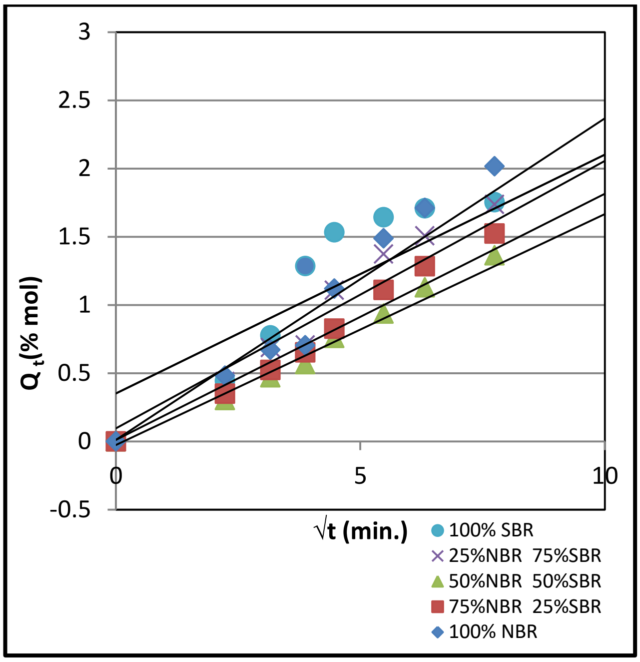

Figure-14. Plot of (Qt) vs ($\sqrt{t}$) for NBR/SBR without Compatibilizers.

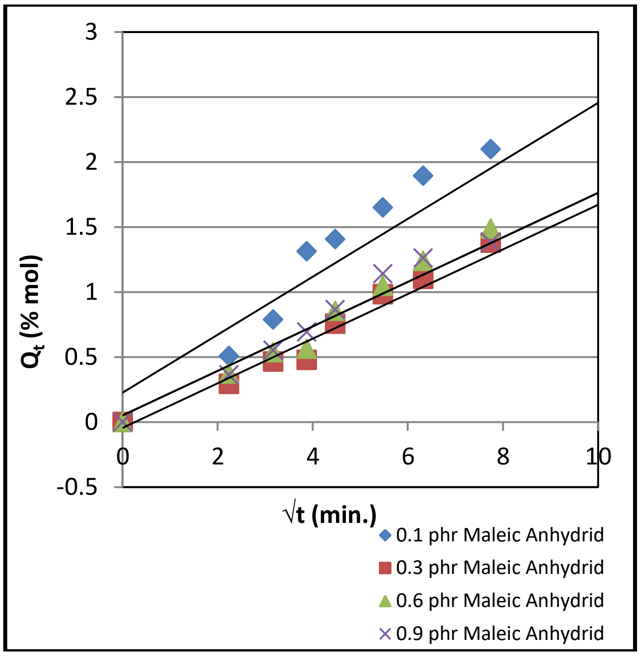

Figure-15. Plot of (Qt) vs ($\sqrt{t}$) for NBR/SBR with Maleic Anhydride.

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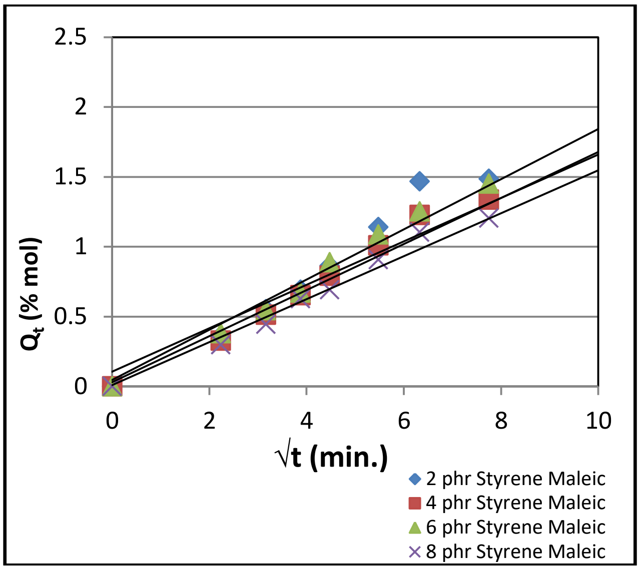

Figure-16. Plot of (Qt) vs ($\sqrt{t}$) for NBR/SBR with Styrene Maleic Anhydride.

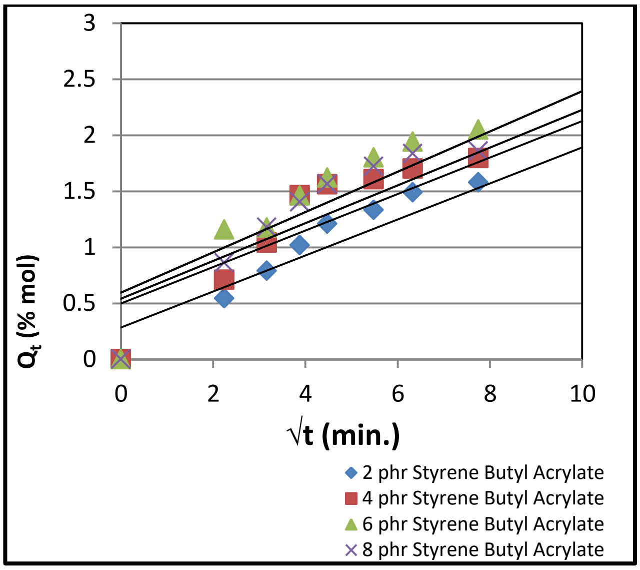

Figure-17. Plot of (Qt) vs ($\sqrt{t}$) for NBR/SBR with Styrene Butyl Acrylate.

The diffusion coefficient of a toluene through NBR/SBR was calculated as shown in section 2.5.1. Table-6 showed the diffusion coefficient for the different examined formula of blends.

Table-6. Diffusion Coefficient for the Different Examined Formula of NBR/SBR Blends.

NBR/SBR	Compatibilizer	Amount of Compatibilizer	D
100/0	-	-	3.394E-08
75/25		-	3.334E-08
50/50		-	1.927E-08
25/75		-	2.21E-08
0/100		-	3.814E-08
50/50	Maleic anhydride	0.1	3.631E-08
50/50		0.3	1.961E-08
50/50		0.6	1.647E-08
50/50		0.9	1.626E-08
50/50	Styrene maleic anhydride	2	1.845E-08
50/50		4	1.66E-08
50/50		6	1.65E-08
50/50		8	1.386E-08
50/50	Styrene butyl acrylate	2	3.515E-08
50/50		4	3.489E-08
50/50		6	3.286E-08
50/50		8	3.297E-08

It is clear from Table-5. pure NBR showed a lower diffusivity compared to SBR. The blend of 50/50 NBR/SBR showed the least diffusivity. On adding maleic anhydride the diffusivity decreases gradually, that is to say, the diffusion coefficient value is inversely proportional to the amount of maleic anhydride, the same trend was observed with maleic styrene. On using styrene butyl acrylate the diffusivity decreased to its minimum value at 6 phr then began rising again. The values of n and

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K were obtained from the plot of (Qt / $Q\infty$) against log *t* as shown in Table-7.

Table-7. Values of *K* and *n* for Different Examined Samples of NBR/SBR.

NBR/SBR	Compatibilizer	Amount of Compatibilizer	K	N
100/0	-	-	0.192752	0.427
75/25		-	0.101391	0.868
50/50		-	0.102094	0.819
25/75		-	0.253513	0.574
0/100		-	0.138038	0.785
50/50	Maleic anhydride	0.1	0.390841	0.52
50/50		0.3	0.142561	0.66
50/50		0.6	0.218776	0.57
50/50		0.9	0.188799	0.606
50/50	Styrene maleic anhydride	2	0.076208	0.875
50/50		4	0.179887	0.626
50/50		6	0.218776	0.589
50/50		8	0.163682	0.63
50/50	Styrene butyl acrylate	2	0.731139	0.338
50/50		4	0.366438	0.449
50/50		6	0.159221	0.465
50/50		8	0.462381	0.469

When the sample is only SBR its show anomalous transport behavior, the same behavior is shown with NBR/SBR blend (75/25, 50/50, 25/75). The transport of toluene through the NBR is described by Less Fickian mechanism, the value of obtained *n* is 0.427. The values of n obtained from the blend of 50/50 NBR/SBR with different amount of Maleic anhydride (0.1, 0.3, 0.6, and 0.9) shows anomalous transport behavior. The same behavior is shown when use a different amount of Styrene maleic anhydride(2, 4, 6, 8). It's clear when styrene butyl acrylate used the less than 0.5 that is mean solvent penetration is much below the polymer chain relaxation and the solvent diffuses slowly this is "Less Fickian", or "Quasi Fickian".

4. CONCLUSIONS

a) The morphology of blends is dependent on the rheological properties of the components, and rheological properties are not affected with the kind of added compatibilizer or its doses.

b) At the break, the tensile strength and elongation percentage decrease as the amount of NBR decreases until it reaches its optimal value at 50/50 NBR/SBR ratio, this blend possesses the superior mechanical properties. NBR/SBR 50/50 compatibilization with different amount of maleic anhydride affects the tensile strength and elongation percentage and shows its superior value at 0.6 phr of maleic anhydride. Different doses of styrene maleic anhydride with NBR/SBR 50/50 are shown the best values of the tensile strength and elongation percentage with 4 phr of styrene maleic. It is recommended to add 4 phr of styrene butyl acrylate as a compatibilizer with NBR/SBR 50/50 because it shown the best value of tensile strength and elongation percentage at the break.

c) Pure SBR has the highest value of sorption, and 50/50 blend NBR/SBR showed the minimum sorption yet. After adding maleic anhydride as a compatibilizer to 50/50 NBR/SBR, it was clear that more amount of maleic anhydride showed the less sorption of toluene. The same for styrene maleic anhydride as a compatibilizer more dose of styrene maleic anhydride lead to less sorption of toluene in the blend.

d) It is recommended to add 6 phr dose of styrene butyl acrylate as a compatibilizer to the blend because it shown the less value of sorption in toluene.

e) The diffusivity of SBR is higher than that of NBR and the blend of 50/50 NBR/SBR showed the less value of

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D. it was observed that 0.1 phr of maleic anhydride showed the highest *D*. Then less diffusivity at the higher dose of maleic anhydride. Dose of styrene maleic anhydride in the blend lead to less *D*. A dose of 2 phr of styrene butyl acrylate as a compatibilizer increasing the diffusivity.

f) The values of n and K were obtained from the plot of (Qt / $Q\infty$) against log t, SBR shows anomalous transport behavior, which is the same behavior is shown with NBR/SBR blend (75/25, 50/50, 25/75). The transport of toluene through the NBR is described by Less Fickian mechanism. The blend of 50/50 NBR/SBR with different amount of Maleic anhydride shows anomalous transport behavior. The same behavior is shown when use a different amount of Styrene Maleic. when styrene butyl acrylate used the less than 0.5 that is mean solvent penetration is much below the polymer chain relaxation and the solvent diffuses slowly this is "Less Fickian", or "Quasi Fickian".

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