EFFECT OF AGING ON THE RHEOLOGICAL PROPERTIES OF THE MODIFIED ASPHALT-RUBBER SYSTEM USING MICROWAVE TECHNIQUE

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The present work aimed to study the effect of aging on asphalt–rubber system after 12 month of the preparation process. The rheological properties of Qaiyarah asphalt were improved with using reclaimed rubber tires treated with anhydrous aluminium chloride. The reclaimed rubber tire samples in 0.5 and 1.0 % of the total amount of the asphalt (the amount of used anhydrous aluminium chloride catalyst was 0.06, 0.12, 0.25, 0.5, 1 %, respectively) were added to Qaiyarah asphalt sample. The reaction components were introduced into a microwave oven operating at 360 W power for 5, 10 and 15 minutes, respectively. The rheological properties including ductility, penetration, softening point and penetration index were measured before and after aging experiments. It was found that the modified asphalt exposed to atmospheric conditions has lower damage in comparison with untreated one and the longer treatment time was used the less crack and the larger resistance were achieved.

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Introduction

Asphalt is an extremely complex mixture and contains a large number of hydrocarbons with variable molecular weight ranging between 400 and 5000 g mol^{-1}. Its viscosity increases very much with decreasing temperature . Asphalt components interact with each other by strong physical and chemical forces resulting in its homogeneous look. .[1] The asphalt properties can be improved by different ways, e.g. by mixing with polymers additives which is one of the most common modification method.[2] Polymeric materials lead to hardening of the asphalt, improves its performance at low temperatures and resistance toward higher temperatures and improve its adhesion and cohesion properties as well.[3]

The rheological properties of asphalt change due to oxidation of hydrocarbons when it is exposed to the sunlight. The formation of free radicals leads to reactions that produce higher molecular weight components than those existed in the starting asphalt. This results in increasing the hardness and softening point and in reducing the values of ductility and penetration.[4]

The asphalt samples modified with rubber tires and styrene-butadiene rubbers after 12 months of aging time showed that prolonging the period of aging reduces the ductility, hardening and polymer breaking.[4] Saleh studied the effect of asphaltene additives on the properties of Begi and Qaiyarah asphalts. The increase in homogeneity for both asphalt types and improvement in ageing specifications for Qaiyarah asphalt[5] could be detected after 18 months of aging compared with untreated asphalt

Anjan and Veeragavan studied mixing of various additives with asphalt to improve its the mechanical properties.[6] Alfi modified the properties of Begi and Qaiyarah asphalts by using polymers such as polystyrene resins, melamine-formaldehyde and poly(methyl methacrylate) with or without aluminium chloride as the catalyst using microwave technique.

Changes in the rheological properties were proved to be advantageous, e.g. excellent values of flow and stability could be detected.[7]

Experimental

A Tokiwa 900 W microwave oven (500 MHz, Germany) was used for the ductility and penetration measurements according to the ASTM D5-83[8] and the ASTM D36-70 standards.[9] Softening point measurement were conducted in accordance with the specifications of ASTM D5-85 standrd.[10]

Qaiyarah crude asphalt was produced in Al-Qaiyarah refinery in Iraq. Reclaimed rubber tires, produced by Babylon Tyres Industry, were ground into ~1.0 mm granules (isoprene rubber content was ~40 %).e.[11] Anhydrous aluminum chloride was supplied by Fluka.

Thermal gravimetric analysis of rubber

One gram of reclaimed crushed rubber tyres granule, s (~1 mm in size) placed into a ceramic crucible and covered with aluminium foil, were heated between 50-600 °C (with 50 °C intervals). The results are given in the Table 1 and Fig 1.

Rubber preparation

Reclaimed rubber tires were thermally crushed before interaction with asphalt material in an electric oven at 350 °C for one hour and cooled at room temperature before crushing it in a mortar.

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Table 1. Thermal analysis of reclaimed rubber tires.

Weight (g)	Temperature (°C)
1*	0
0.9996	50
0.9962	100
0.9852	150
0.9532	200
0.9223	250
0.8962	300
0.6493	350
0.6100	400
0.4640	450
0.1652	500
0.0523	550
0.0094	600

*Net weight

Modification of the rheological properties of asphalt by catalytic treatment

A 250 g of asphalt sample was heated up to 100 °C then crushed and mixed with either reclaimed rubber tyre granules (0.5 and 1.0 %) or anhydrous aluminium chloride (0.06, 0.12, 0.25, 0.5 and 1.0 %). The prepared samples were heated in the microwave oven for 5, 10 and 15 min. The ductility,[8] penetration,[9] softening point[10] and penetration index[12] of the sample were measured.

Effect of ageing time

The effect of ageing time under atmospheric condition was evaluated by measuring the ductility,[8] penetration,[9] softening point[10] and penetration index[12] values. The measurements were repeated after 12 months.

Results and Discussion

Thermal treatment of reclaimed rubber tires

The reclaimed rubber tire granules were used as additive to modify the properties of asphalt which was thermally treated at 350 °C for 1 h. The weight loss of rubber sample at various temperatures is given in Table 1 and Fig.1.

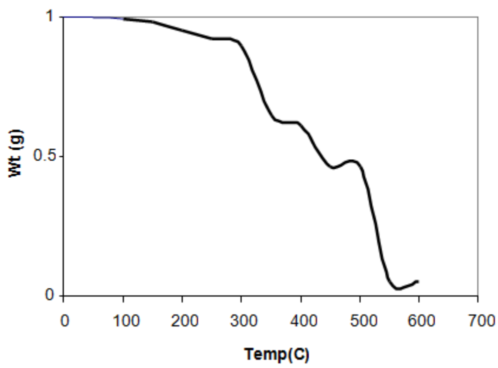


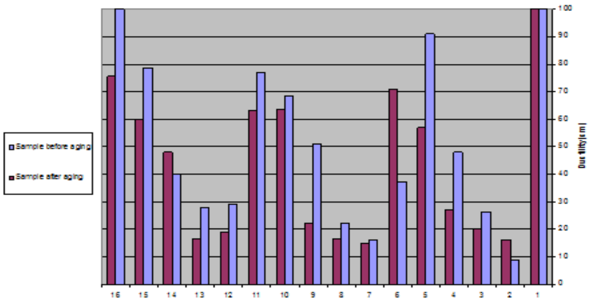
Figure 1. The weigh loss of reclaimed rubber tires at differnet temperatures

Modification of the rheological properties of asphalt

The properties of asphalt modified by reclaimed rubber tire granules in presence of catalytic amount of anhydrous aluminium chloride are given in Tables 2 and 3. The asphalt modified in this way has considerable resistance to ageing. Using more than 1 % of reclaimed rubber tires provides heterogeneity in the produced asphalt mixture. The 360 W power of microwave oven used was to be selected in order to ensure homogeneous dissolving of the components. The use of higher power caused hardening of the sample, decrease in ductility and penetration, and increase in the softening point.

The anhydrous aluminium chloride ($AlCl_3$) was selected as a Lewis acid catalyst in order to ensure appropriate conditions to react the appropriate components with each other.[14,15]

(a)



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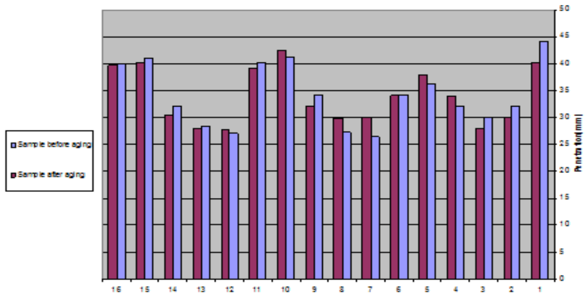

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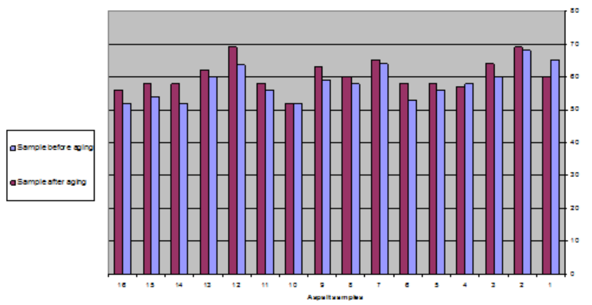

Figure 2. The rheological properties of reclaimed tires rubber before and after ageing of modified asphalt (0.5%) with different percentage of anhydrous aluminium chloride at (360) watt for different time intervals. (a) ductility of asphalt samples; (b) penetration of asphalt samples; (c) softening point of asphalt samples.

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Table 1. Rheological properties of modified asphalt (0.5 % of reclaimed tires rubber with different percent of anhydrous aluminum chloride) samples heated at 360 W microwave power for different time intervals, before and after aging.

$AlCl_3$ wt%	Time, min	Ductility at 25 °C, 5 cm min^{-1}			Penetration at 25 °C, 100g, 5 s			Softening point, ring and ball			Penetration index, PI		
		Before	After	Δ	Before	After	Δ	Before	After	Δ	Before	After	Δ
0	0	+100	+100	0	44.2	40.2	4	65.0	60.0	5	1.662	0.506	1.156
1	5	9.0	16.3	-7.3	32.1	30.1	2	68.0	69.0	-1	1.434	1.456	-0.022
0.5	5	26.4	20.0	6.4	30.0	28.0	2	60.0	64.0	-4	-0.122	0.466	-0.588
0.25	5	48.0	27.3	20.7	32.2	34.0	-1.8	58.0	57.0	1	-0.365	-0.455	0.09
0.12	5	91.0	57.0	34	36.2	38.0	-1.8	56.0	58.0	-2	-0.529	-0.016	-0.513
0.06	5	37.3	71.0	-33.7	34.2	34.1	0.1	53.0	58.0	-5	-1.291	-0.248	-1.043
1	10	16.1	15.0	1.1	26.5	30.0	-3.5	64.0	65.0	-1	0.405	0.782	-0.377
0.5	10	22.0	16.5	5.5	27.2	29.8	-2.6	58.0	60.0	-2	-0.695	-0.135	-0.56
0.25	10	51.0	22.3	28.7	34.2	32.2	2	59.0	63.0	-4	0.044	0.579	-0.535
0.12	10	68.4	63.8	4.6	41.2	42.5	-1.3	52.0	52.0	0	-1.138	-1.073	-0.065
0.06	10	77.0	63.0	14	40.2	39.2	1	56.0	58.0	-2	-0.307	0.052	-0.359
1	15	29.0	19.2	9.8	27.0	27.8	0.8-	63.5	69.0	-5.5	0.303	1.287	-0.984
0.5	15	28.0	16.4	11.6	28.4	28.0	0.4	60.0	62.0	-2	-0.231	0.109	-0.34
0.25	15	40.0	48.0	-8.0	32.2	30.5	1.7	52.0	58.0	-6	-1.629	-0.475	-1.154
0.12	15	78.5	60.0	18.5	41.0	40.2	1	54.0	58.0	-4	-0.696	0.108	-0.804
0.06	15	+100	75.6	24.4	40.0	39.7	0.3	52.0	56.0	-4	-1.201	-0.334	-0.867

Table 3. Rheological properties of modified asphalt (1.0 % of reclaimed tires rubber with different percent of anhydrous aluminium chloride) samples heated at 360 W microwave power for different time intervals, before and after aging.

$AlCl_3$ wt%	Time, min	Ductility at 25 °C, 5 cm min^{-1}			Penetration at 25 °C, 100g, 5 s			Softening point, ring and ball			Penetration index, PI		
		Before	After	Δ	Before	After	Δ	Before	After	Δ	Before	After	Δ
0	0	+100	+100	0	44.2	40.2	4	65.0	60.0	5	1.662	0.506	1.156
1	5	12.2	13.2	-1	26.2	30.0	-3.8	64.0	65.0	-1	0.331	0.782	-0.451
0.5	5	18.7	27.0	-8.3	32.0	30.2	1.8	56.0	60.0	-4	-0.783	-0.108	-0.675
0.25	5	23.3	35.4	-12.1	32.2	32.0	0.2	55.0	60.0	-5	-0.977	0.009	-0.986
0.12	5	31.5	56.2	-24.7	33.7	33.0	0.7	50.0	61.0	-11	-2.000	0.264	-2.264
0.06	5	56.0	50.0	6	34.4	32.0	2.4	57.0	57.0	0	-0.429	-0.579	0.15
1	10	12.0	15.7	-3.7	27.5	25.0	2.5	56.0	64.0	-8	-1.075	0.240	-1.315
0.5	10	19.0	17.3	1.7	33.2	30.5	2.7	58.0	62.0	-4	-0.304	0.284	-0.588
0.25	10	23.0	38.0	-15	31.6	33.7	-2.1	54.0	60.0	-6	-1.229	0.120	-1.349
0.12	10	53.0	33.0	20	34.2	32.0	2.2	57.0	60.0	-3	-0.443	0.009	-0.452
0.06	10	50.0	35.0	15	37.2	36.0	1.2	53.0	59.0	-6	-0.899	0.064	-0.963
1	15	13.0	12.5	0.5	31.2	28.5	2.7	58.0	62.0	-4	-0.430	0.146	-0.576
0.5	15	26.0	20.5	5.5	34.2	31.4	2.8	60.0	60.0	0	0.149	0.004	0.145
0.25	15	17.1	32.0	-14.9	30.5	31.9	-1.4	58.0	60	-2	-0.476	0.005	-0.481
0.12	15	52.0	44.0	8	29.2	30.0	-0.8	55.0	54.0	1	-1.168	-1.326	0.158
0.06	15	40.0	39.5	0.5	35.7	36.0	-0.3	54.0	57.0	-3	-0.985	-0.337	-0.648

It can be seen from the results in Tables 2 and 3 or Figures 2 and 3 that most of the modified asphalt samples that were subjected to ageing do not show substantial changes in their important properties. It is an advantageous result indicating that the samples of modified asphalts resisted large stresses and cracks. It ensures long operational life which does not alter importantly with aging. Using reclaimed rubber tires in this form as additives improves the mechanical properties via increasing the durability, reducing stresses and thermal cracking and increasing resistance towards forming grooves.[16,17] This phenomenon might be the direct consequence of the presence of high amount of carbon black in reclaimed rubber which can increase the life and resistance of rubber materials against oxidation and sunlight.

The sulfur compounds, that exist as original components in the mixture of Qaiyarah crude asphalt in a relative high percentage (about 7 %), play an important role in the resistance of asphalt towards oxidation because these compounds act as free radical scavenger,[1,18,19] and antioxidants thereby these can prevent the asphalt against free-radical accelerated oxidation processes.

Based on the results given in Tables 2 and 3 some samples were selected for engineering test for making paving asphalt. Some other samples characterized with high softening point and low values of penetration and ductility qualified in the production of moisture insulation material and some samples were tested as flattening asphalt.

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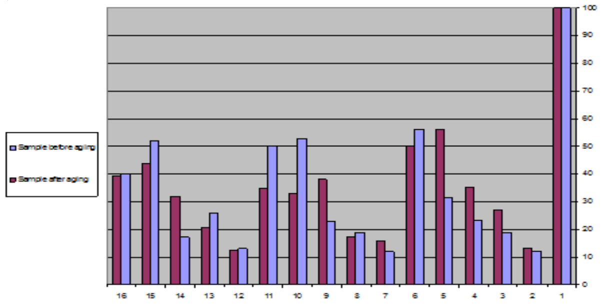
(a)



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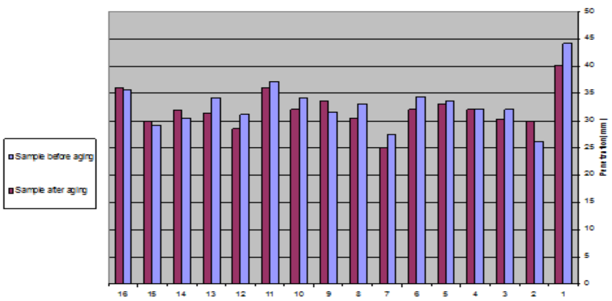

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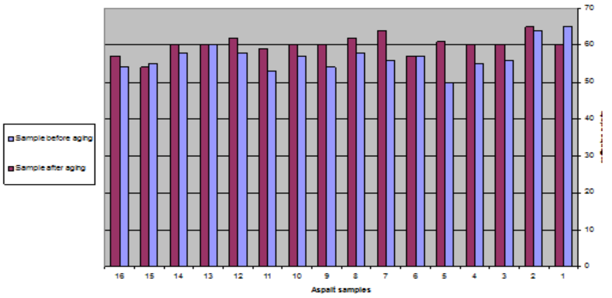

Figure 3. Rheological properties before and after ageing of modified asphalt (1 %) of reclaimed tires rubber with different percent of anhydrous aluminum chloride, at (360) watt for different time intervals. (a) ductility of asphalt samples; (b) penetration of asphalt samples; (c) softening point of asphalt samples.

The values of penetration, ductility and softening point of the asphalt which can be used for mastic insulator[20] to moisture, for flatness[21] and for paving[22] can be found in the appropriate American Standards (ASTM) descriptions.

Conclusion

The microwave energy is used to reduce the modification time from hours to minutes by making modified rubber asphalts in the presence of aluminium chloride or other Lewis acid catalysts. This microwave treatment decreased the amount of evolved pollutant gases during the treatment and thereby reduced the environmental pollution.

The thermal sensitivity was found to be correlated with the Penetration index (PI) values between +2 and -2. This shows the increasing thermal stability of the end products.

In addition, using of small amount of reclaimed rubber tyres into asphalt makes the system to be more homogenous and improves the asphalt resistance against weather conditions.

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