

# Modification of Asphalt Cement Properties and Chemical Properties by Polypropylene and Cellulose Additives

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## Abstract

The study investigated the physical and rheological properties of asphalt binder modified by polypropylene and cellulose materials. The tests were conducted in the study; conventional tests such as; penetration, softening point and ductility, and unconventional tests such as rotational viscosity and dynamic shear rheometer (DSR). From the conventional tests it found that the hardness of modified asphalt binder with the addition of polypropylene and cellulose materials up to 5%. As a result of increased the hardness the softening point of modified asphalt improved compared with base asphalt binder. The rheological property of modified binders enhanced at low temperatures and high temperatures, as the results of DSR test showed that the  $G^*$  were improved. The addition of a different percentages of polypropylene and cellulose fibers to base binder had remarkable influence of resistant to permanent deformation (high temperatures rutting and low temperature fatigue).

## Keywords

Polypropylene, Cellulose, Physical and Rheological Properties, Conventional Tests, Modified Asphalt Binder, Dynamic Shear Rheometer and Rutting and Fatigue Parameters

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## 1. Introduction

Asphalt paving roads show limitation on temperature, cracked when the temperature is low and softens when the temperature is high. Besides, high loading weight and hard traffic will destruction the roads earlier than usual and cause expenses to repair and maintenance [1]. Asphalt cement has the characteristics of low-temperature cracking and high temperature rutting. Moreover, fatigue, rutting and aging are some of the distresses of Asphalt cement causes limited in its manufacturing application. On the other hand, asphalt roads undergo important distresses not only under temperature, but also with increasing heavy traffic loads [2]. Therefore, the modification of Asphalt cement is essential to improve the properties of asphalt cement using materials which can play the role such as polypropylene and cellulose to achieve the best asphalt performance [3].

There are many researchers looking for the reasons to modify bituminous materials. Lewandowski mentioned that the main reasons to modify bituminous materials with different type of additives could be summarized as follows [4]: To obtain softer blends at low service temperatures and reduce cracking, to reach stiffer blends at high temperatures and reduce rutting, to improve fatigue resistance of blends.

Serfass and Samanos [5] concluded that the addition of fibers to asphalt concrete improved the fixation of the asphalt binder in the mix. This relates to less bleeding and improved skid resistance over unmodified mixtures of the same design. Fiber modification also allowed for an increase in film thickness, resulting in less aging and improved binder characteristics. The addition of fibers also resulted in the reduction of temperature susceptibility of asphalt mixtures. Adding fibers enables developing mixtures rich in bitumen

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asphalt binder, and therefore displaying high resistance to moisture, fatigue, and cracking.

Polypropylene fibers were used in a 1993 study by Jiang et al. [6] in an attempt to reduce reflection cracking in an asphalt overlay. Although crack intensities were less on the fiber modified overlay sections, a reduction or delay in reflection cracking was not observed. Sections in which the concrete was cracked and sealed before the overlay were found to have less reflection cracking when fibers were used in either the base or binder layers

In a separate study, a fracture mechanics approach was used to evaluate the effects of fiber reinforcement on crack resistance [7]. Polyester and polypropylene fibers were used to modify mixtures that were then tested for modulus of elasticity, fracture energy, and tensile strength. Fracture energy in modified samples increased by 50 to 100 percent, implying increased toughness. Elasticity and tensile strength results were not significantly affected.

Bahia studied the effect of polymer modification using scanning electron microscope images. The results showed that the modified asphalt concrete mixtures have better binder-aggregate adhesion, which led to increase in its toughness [8]. The main advantage of using polymer

technology is to improve the adhesion properties between the binders and aggregate. Due to ever increasing traffic volume and loading, there is a continuing effort by pavement community to improve asphalt binder. Mechanical properties of asphalt binder can be adjusted to help extend the service life of asphalt pavement as well as minimize the potential pavement distress. The addition of polymer to asphalt binder can reduce potential rutting and fatigue cracking as well as increase cohesion and decrease temperature susceptibility. However, the addition of polymer to asphalt binder increase viscosity and stiffness thereby reduces the workability of asphalt mixture [9].

## 2. Experimental Methods

### 2.1. Materials

Asphalt cement 60/70 penetration grade bituminous was produced and supplied from Durah refinery, Iraq. The physical and chemical compositions of the asphalt cement are listed in Table 1. The polypropylene and cellulose materials were used in this study as a modifier of base asphalt cement, the main properties of it are listed in Table 2.

**Table (1).** The physical and chemical compositions of the 60/70 asphalt cement.

Test	Standard	Value	Specifications
Penetration at 25°C, 100 g (0.10 mm)	ASTM D5	63	SCRB, 2007 [10]
Specific Gravity at 25°C	ASTM D70	1.03	-----
Rotational Viscometer (RV) at 135°C			
Dynamic Shear Rheometer, $G^*/\sin\delta$ , at 10 rad/s, test temperature of 64°C	AASHTO T 316-06	0.85	3.0 Pa.s, max
Ductility (cm) at 25°C	AASHTO TP5	1.42	1.00 kPa, min
Flash point, °C			
Rolling Thin Film Oven Test (RTFOT)	ASTM D113	100	≥100
Dynamic Shear Rheometer, $G^*/\sin\delta$ , at 10 rad/s, test temperature of 64°C	ASTM D92	235	230°C, min
Chemical Composition	ASTM D2872	0.62	1%, max
Asphaltenes (%)		3.68	2.2 kPa, min.
Resins (%)		12.2	
Saturates (%)		30.8	
Aromatic (%)		8.1	
		48.9	

**Table (2).** The main properties of polypropylene and cellulose.

Material	Properties	Value
Polypropylene	Specific gravity	0.91 g/cm <sup>3</sup>
	Specific surface area	260 m <sup>2</sup> /kg
	Young's modulus	5.5-7 Gpa
	Tensile strength	350 Mpa
	Melting point	160°C [17]
	Specific gravity	0.91 g/cm <sup>3</sup>
	Fiber thickness	18 Micron
	Average fiber length	2.1 mm
Cellulose	Diameter	0.016 mm
	Density	1.1
	Surface area	25000 cm <sup>2</sup> /g
	Fiber count at 0.90 kg/m <sup>3</sup>	1.44 billion
	Fiber tensile strength	620-900 MPa

### 2.2. Preparation of Modified Asphalt Cements

The modified asphalt cements were prepared at 150 °C. The modifier quantities (1, 3 and 5 wt.%) of additives were added into the base asphalt binder. The binders were mixed at 5000 rpm speed for about 90 min using a high shear mixer, in order to get the better dispersion of PP and CL in base binder.

### 2.3. Characterizations

The physical properties of the asphalt cement, including penetration, softening point, ductility at 25°C and viscosity were tested according to ASTM D5, ASTM D 36, ASTM D

113 and ASTM D 4402, respectively. The tests were performed three times and the average values of the results were used.

Rotational viscosity test was conducted according to AASHTO T 316-06 "Standard Method of Test for Viscosity Determination of Asphalt" [11]. To have adequate mixing and pumping capabilities, Superpave specification (AASHTO M320) requires the binder to have a rotational viscosity less than 3.0 Pa.s at 135°C. If the rotational viscosity value is higher than 3.0 Pa.s for an asphalt binder, special handling procedure is required to ensure proper mixing and pumping capabilities while using that binder. The viscometer can also be used to develop temperature-viscosity charts by running the viscometer at both 135°C and 165°C to determine laboratory mixing and compaction temperatures.

A Dynamic Shear Rheometer (DSR) was used for dynamic rheological measurements, a plate of 25 mm diameter with 1 mm thickness and a plate of 8 mm diameter with 2 mm thickness, for all binders according to AASHTO TP5. Frequency sweeps test from 10 to 75°C, at a fixed applied frequency of 1.159 Hz (10 rad/s).

The direct tension tester (DTT) is used in the Superpave system to measure low temperature tensile strain at -5°C test temperature. Larger failure tensile strain indicates more ductile asphalt cements and, therefore more resistance to cracking.

### 3. Results and Discussion

#### 3.1. The Physical Properties Test

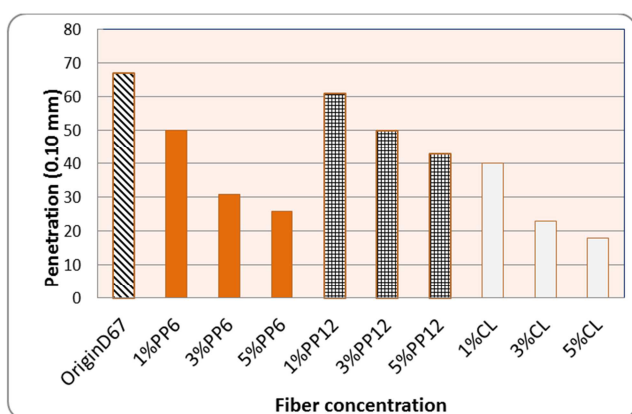


Figure (1). Penetration values of modified and unmodified asphalt cements.

The influences of adding fiber on physical properties of modified asphalt binders samples were instigated. Figure 1 is representing the relationship between the penetration values and fiber concentration. It is observed that a decrease in penetration values with increasing fiber concentration, this

dramatically decreased was as a result of the base asphalt cement becomes more stiff with increase the concentration of the fiber. Furthermore, it noted from figure 2 that the addition of fiber leads to enhance the softening point of modified asphalt binders. This is due to the solid phase of fiber which increase the hardness of modified asphalt cements. In addition, it was recognized that 1%. wt of modifier was the best performance of penetration, softening point and ductility values.

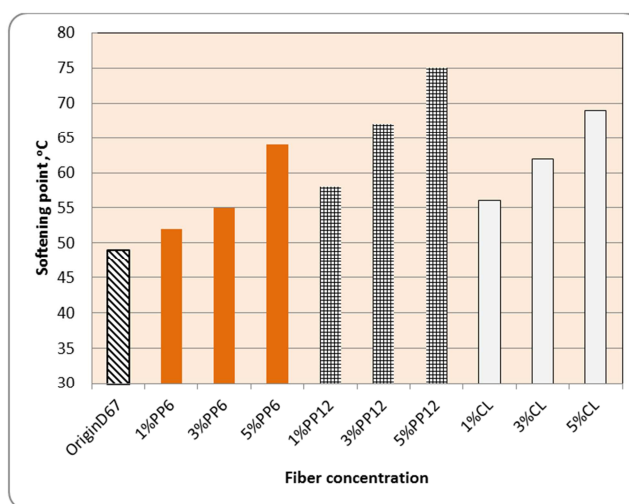


Figure (2). Softening point values for modified and unmodified asphalt cements.

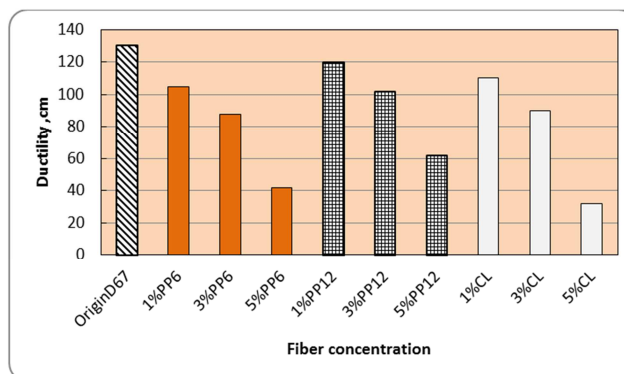


Figure (3). Ductility of modified and unmodified asphalt cements.

The viscosity of the base binder becomes greater with the addition of polypropylene and cellulose at test temperatures (135, 155 and 165°C). However, the modified asphalt cement samples showed significant improvement compared with the origin asphalt cement as shown in Figure 4. The increase in viscosity is a result of the hardening effect of additives. The increased of viscosity of modified asphalt binders might be due to the better dispersion of the additives in the base binder which lead to increase the bonding strength through restricting the flow of asphalt made it harder and led to enhance asphalt physical properties.

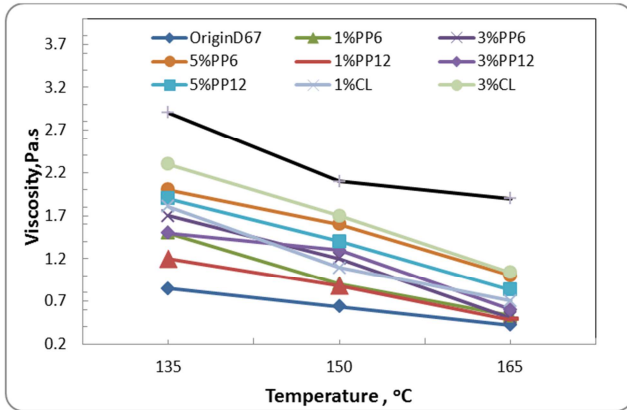


Figure (4). Viscosity of modified and unmodified asphalt cements.

### 3.2. Rheological Properties

The complex modulus ( $G^*$ ) alongside temperature variations for the original asphalt binder and polypropylene and cellulose modified asphalt binders is represented in Figure 4. It is observed that an increase in  $G^*$  with the addition of modifier to asphalt up to 5% of the base asphalt. From Figure 5 the modified asphalts with 5% of fiber has higher complex modulus among the binders which means it has greater resistance to the permanent deformation, meanwhile the base binder has the lowest  $G^*$  value.

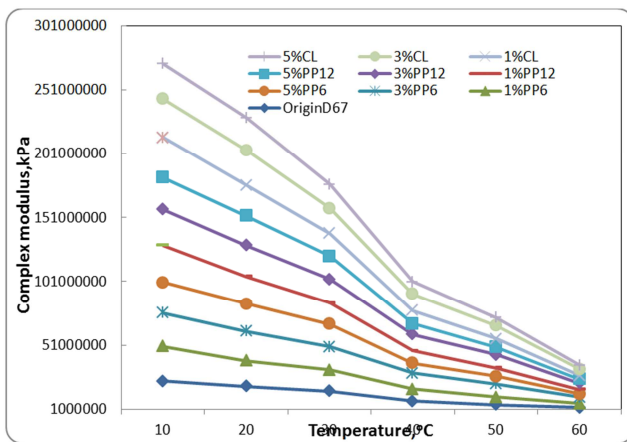


Figure (5). The complex modulus ( $G^*$ ) against temperature.

### 3.3. SHRP Rutting and Fatigue Parameters

The resistance of asphalt binder against rutting and fatigue cracking (permanent deformation) was measured by DSR tests. The DSR tests were conducted on asphalt binders for different temperatures, namely 45-75°C increment of 10°C to measure the rutting resistance. Likewise, fatigue-cracking tests were also conducted at temperatures, specifically at 10, 15, 25, and 35°C. The rutting and fatigue-cracking tests utilized the constant loading frequency of 1.59 Hz (10 rad/s), as identified by the strategic highway research program (SHRP).

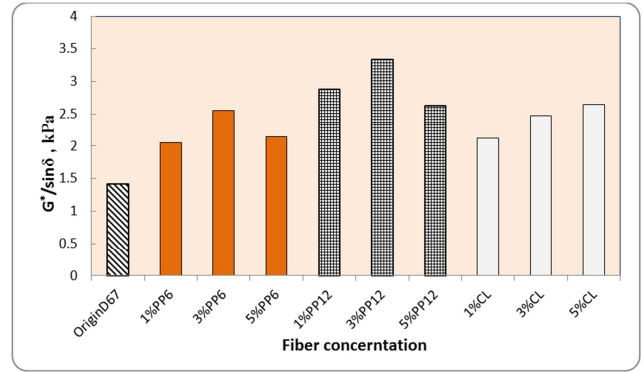


Figure (6). The rutting parameter of modified and unmodified asphalt binders.

The value of  $G^*/\sin \delta$  is regularly used to define the rutting resistance of the asphalt binder at high temperatures. The Superpave technique requires  $G^*/\sin \delta = 1$  kPa as the minimum rutting parameter for an unaged sample [12], [13]. Figure 6 shows that the lowest value of  $G^*/\sin \delta$  was obtained by the unmodified asphalt binder and that the 5% cellulose has the highest value of  $G^*/\sin \delta$ . A greater value of  $G^*/\sin \delta$  indicates a pavement with better permanent deformation resistance [13]. However, the 7% sample shows a different behavior as the value of  $G^*/\sin \delta$  slightly decreased. Overall, it can be observed that all asphalt binders have  $G^*/\sin \delta$  values larger than 1.0 kPa at 65°C. On the other hand,  $G^* \cdot \sin \delta$  is regularly used to define the fatigue resistance of asphalt binder at low temperature. The Superpave method requires a limitation at 5000 kPa for the maximum fatigue-cracking parameter [14]. Figure 7 shows that all asphalt binders including the base asphalt have fatigue-cracking parameters greater than the required Superpave value at all test temperatures ranging from 10 to 35°C. Moreover, it is observed that the decrease in the value of  $G^* \cdot \sin \delta$  by increasing the nano content up to 5% persists up to 25°C. Figure 8 shows failure tensile strain of modified and unmodified asphalt binders.

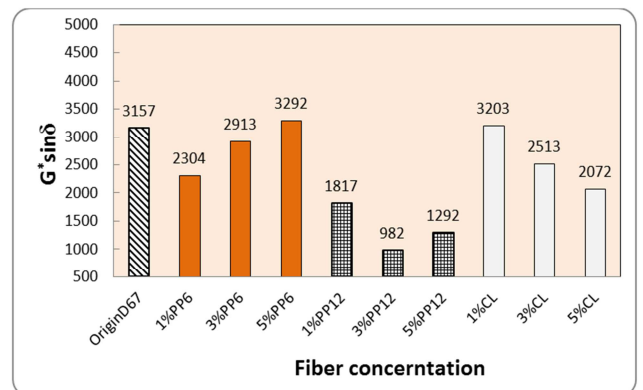
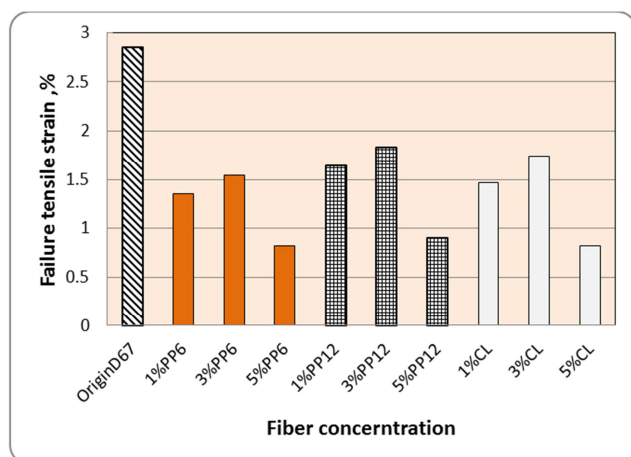


Figure (7). The fatigue parameter of modified and unmodified asphalt binders.



**Figure (8).** The failure tensile strain of modified and unmodified asphalt binders.

Referring to Figure 8 it is observed that failure tensile increased with increasing of fiber concentration up to 5%. The minimum percent of failure tensile strain is 1.0% as required by SUPERPAVE [15].

### 3.4. Chemical Composition of Modified Asphalt

Asphalt components can be separated by using the solubility of the various molecules in different solvents. The standard (ASTM D 4124) test method [16], used in this work, separates out the solid (Asphaltenes) as n-heptane insoluble the remaining groups of resins and oils, referred to as (Petrolenes) are composed of:

-Saturates: liquid material that is not absorbed on calcined F-20 alumina absorbent on percolation in a n-heptane solvent.

Saturates and naphthene -aromatics are liquid and provide a softening or plasticizing effect on the polar aromatic and asphaltenes which are solid and have hardening effects [17]. In general, asphaltenes produce the bulk of the asphalt,

aromatics contribute to adhesion and ductility, and saturates influence flow and viscosity properties [18].

-Naphthene -Aromatics: liquid material that is adsorbed on calcined F-20 alumina in the

presence of n-heptane, and desorbed by toluene.

-Polar -Aromatics: Solid material desorbed from calcined F-20 alumina absorbent, using toluene and trichloroethylene solvents.

Corbet [19] studied by separating of asphalt into its four generic fraction (saturates, naphthene-aromatics, polar-aromatics, and asphaltenes), and observing the change in properties with respect to the variation in composition. In addition, method of characterization was also evolved which serves to identify the average chemical structure within each fraction

To avoid using a single component fraction to judge the behavior of asphalt, the author favors the use of the Gaestel or composition index which was suggested by Brule [20] to summarize the overall behavior of the asphalt in terms of its colloidal stability. There appear to be definite ranges of this index that are commensurate with the rheological character of an asphalt as measured by viscosity and penetration.

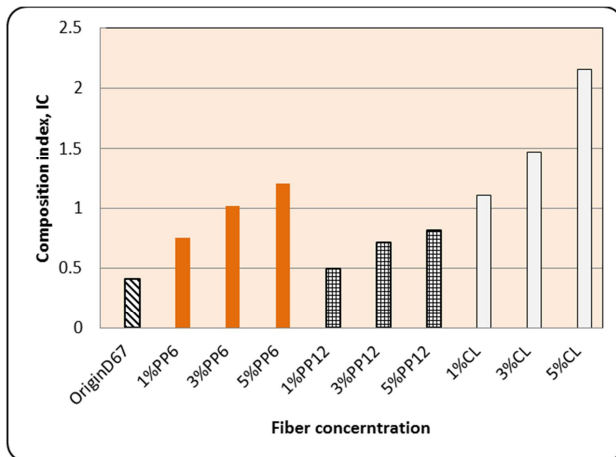
Composition index (IC) as the ratio of the less reactive fractions (Asphaltenes and Saturates) to the more reactive fractions (Naphthene and Polar Aromatics) can be used to evaluate processing as related to performance properties of Asphalt cement [18]. The compositional fractions have been conducted by the device existed in Chemical Engineering Department Laboratories in Baghdad University. The chemical fractions and composition index of various modified asphalt types used in this study are reported in Table 3 below

**Table 3.** Chemical fractions and composition index of various modified asphalt types.

	Asphaltene	Polar -Aromatics	Naphthene -Aromatics	Saturates	Composition index( IC)
OriginD67	12.5	8.4	62.1	16.5	0.41
1%PP6	22.7	15.1	41.8	20.4	0.76
3%PP6	30.6	18	31.2	19.5	1.02
5%PP6	36.4	15.2	30.1	18.3	1.21
1%PP12	17.3	21.6	44.8	15.9	0.50
3%PP12	19.6	16.6	41.1	21.5	0.71
5%PP12	23.4	16.9	37.4	20.9	0.82
1%CL	33.1	18.7	28.6	19.2	1.11
3%CL	38.7	16.4	23.9	20.6	1.47
5%CL	46.9	21.7	9.93	21.3	2.16



Figure (9) exhibits Composition index values of modified and unmodified asphalt binders.



**Figure (9).** Composition index values of modified and unmodified asphalt binders.

It is clearly shown that composition index slightly and extremely increased when fiber concentration increased for modified asphalt with polypropylene and cellulose fibers respectively.

## 4. Conclusion

In this study, an assessment of using polypropylene and cellulose modified asphalt cements was presented according to the conventional and rheological properties.

Test data obtained from the testing program yields the following outcomes:

1. Improved stiffness of the modified binders which means declined their temperature susceptibility. However, the improvement was quite obvious on 5% of additives.
2. The softening point of modified binders improved for modified binders compared with origin asphalt cement.
3. Frequency sweep test shows that all modified binders had an increase in complex modulus value and decrease the phase angle comparing with the base asphalt binder, which lead to enhanced elastic behavior of asphalt.
4. The fatigue and the rutting parameters showed that the both modifiers were able to increase the resistance high temperature rutting and improved low temperature fatigue.
5. Overall, the study of characterization of polypropylene and cellulose materials summaries that modification using additives able to improve the physical and rheological properties of asphalt binder significantly. Moreover, the best performance of modified asphalt binders was 5% of the modifier.

6. The additives modified the chemical composition of the asphalt cements by increased the less active components in the modified asphalt in comparison with the more active components.

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## References

- [1] Vichitcholchai, N., Panmai, J. & Na-Ranong, N. (2006), "Modification of Asphalt Cement by Natural Rubber for Pavement Construction", Nippon Gomu Kyokaishi (Journal of the Society of Rubber Industry, Japan) 79 (3): 170.
- [2] Muniandy, R., Mahdi, LM, Yunus, R., Hasham, S. & Aburkaba, E. 2013, "Characterization of Organic Montmorillonite Nanoclay Modified Asphalt Binders Using Dynamic Shear Rheometer (Dsr)". Australian Journal of Basic & Applied Sciences 7 (14):
- [3] Zhang Sl, Zhang Zx, Xin Zx, Pal K, Kim Jk, (2010). "Prediction Of Mechanical Properties Of Polypropylene / Waste Ground Rubber Tire Powder Treated By Bitumen Composites Via Uniform Design And Artificial Neural Networks", Mater Design, 31, 1900-1905,.
- [4] Lewandowski, L.H. Polymer Modification Of Paving Asphalt Binders. Rubber Chemistry And Technology, Volume 67, Pp435-447. 1994.
- [5] Serfass, J.P. and J. Samanos. (1996), "Fiber-Modified Asphalt Concrete Characteristics, Applications and Behavior", Journal of the Association of Asphalt Paving Technologists, Vol. 65, p 193-230.
- [6] Jiang, Yi: Rebecca S. McDaniel, (1993), "Application of Cracking and Sealing and Use of Fibers Control Reflection cracking", Transportation Research Record 1388 p150. 159.
- [7] Jenq, Yeou-Shang: Chwen-Jang Liaw, Pei Liu., (1993), "Analysis of crack resistance of asphalt concrete overlays. A fracture mechanics approach" Transportation Research Record n 1388 pp 160-166.
- [8] Bahia, H. And Anderson, D.A., (1995), "Strategic Highway Research Program Binder Rheological Parameters: Background and Comparison With onventional Proerties", Journal of The Transportation Research Board, Volume 1488, pp. 32-39.
- [9] Zhang, F., Yu, J., Wu, S., (2010) "Effect Of Aging On Rheological Properties Of Torage-Stable SBS/Sulfur-Modified Asphalts" J. Materials In Civil Engineering, 182.
- [10] State Cooperative of Roads and Bridges (SCRB), "Standard Specifications for Roads and Bridges", Republic of Iraq, Ministry of Housing and Construction, Department of Design and Studies, Baghdad, Addendum No. 3, 2007.

- [11] AASHTO, (2007), *Standard Specifications for Transportation Materials and Methods of Sampling and Testing*, 5<sup>th</sup> edition, American Association of State Highway and Transportation Officials, Washington, D.C., USA.
- [12] McGennis, RB, Shuler, S. & Bahia, HU (1994), "Background of Superpave Asphalt Binder Test Methods", Federal Highway Administration, Office of Technology Applications.
- [13] Khadivar, A. & Kavussi, A., (2013), "Rheological Characteristics of Sbr and Nr Polymer Modified Bitumen Emulsions at Average Pavement Temperatures", *Construction and Building Materials* 47 (1099-1105).
- [14] Rusbintardjo, G., Hainin, MR & Yusoff, NIM, (2013), "Fundamental and Rheological Properties of Oil Palm Fruit Ash Modified Bitumen", *Construction and Building Materials* 49 (702-711).
- [15] Asphalt Institute, (2003), *Performance Graded Asphalt Binder Specification and Testing*, Manual Series No. 1, (SP-1), Asphalt Institute, Lexington, Kentucky.
- [16] ASTM Standards, (2005), "Roads and Paving Materials", Annual Book of the American Society for Testing and Materials Standards, Section 4, Vol. 04-03.
- [17] Corbett and H.E. Schweyer, *Composition and Rheology Considerations in Age Hardening of Bitumen*, Proceedings of the Association of Asphalt Paving Technologists, Vol. 50, 1981.
- [18] Krebs, R. D. and R. D. Walker, (1971), *Highway Materials*, McGraw-Hill, New York, NY.
- [19] Corbett, L.W., (1970), "Relationship between Composition and Physical Properties of Asphalt", Proceedings of the Association of Asphalt Paving Technologists, vol.39.
- [20] Brule, B., (1986), "Relationship Between Composition, Structure and Properties, of Road Asphalts: State of Research at the French Central Road Research Laboratory", Proceedings of the 67th Annual Meeting of the Transportation Research Board, Jan.