

Evaluation of Pyrolysis Polypropylene Modified Asphalt Paving Materials

Abdul-Rahim I. Al-Hadidy

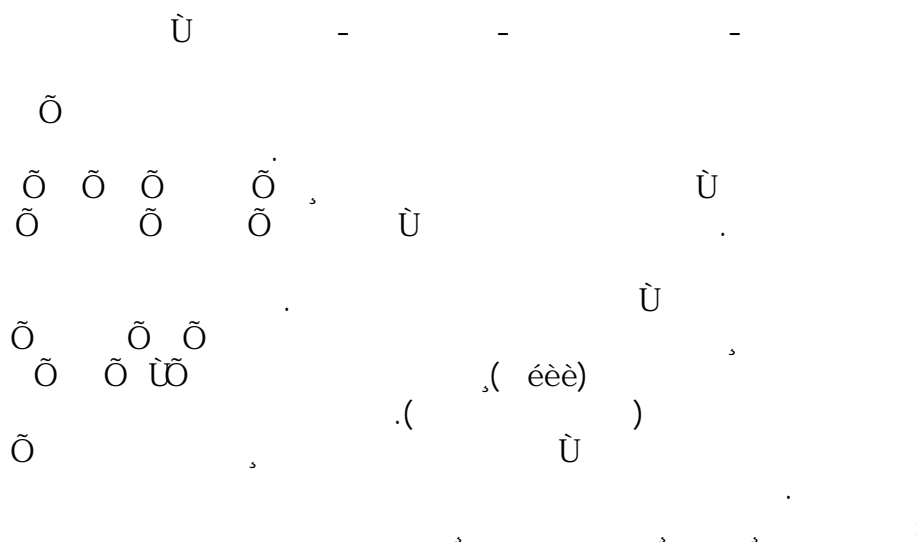
Assistant Lecturer, College of Engineering,
Civil Dept., Mosul University, Mosul-Iraq

Abstract

The main objective of this research study is to investigate the potential use of pyrolysis polypropylene as modifiers in hot mix asphalt paving mixtures. Seven different blends including conventional mix were subjected to binder testing such as rheological tests, as well as to some other tests related to the homogeneity of the system.

Optimum asphalt content was obtained by Marshall method and used in all the modified mixes. The engineering properties, including Marshall stability and indirect tensile strength were determined. Research results indicate that modified binders showed higher softening point, keeping the values of ductility at minimum range of specification of (100cm), and caused a reduction in percent loss of weight due to heat and air (i.e. increase durability of original asphalt). The results indicated that the inclusion of pyrolysis polypropylene with asphaltic concrete mixtures gave a quite satisfactory result in terms of stability and other Marshall properties, and increase in indirect tensile strength).

Keywords: polypropylene, polymers, asphalt modifiers, Asphalt binders



Introduction

One of the basic and important requirements of asphalt concrete pavements is durability. Durability is the resistance of asphalt concrete to the action of temperature and temperature changes, and the action of air and water, the action of traffic.

Most roadways in the Iraq provinces lack of durability, because Iraq has characterized with a continental climate. This resulted in a wide temperature difference between summer and winter. Such climatic conditions have a heavy impact on paving asphalt mixtures as it should possess a high softening point to sustain hot Summer conditions, with enough ductility to resist low winter temperatures.

This study is an attempt to use pyrolysis polypropylene as a modifier in asphalt paving mixtures to satisfy the following objectives:

- 1.To develop new asphalt mixtures that possess a higher softening point, than the original material and keeping at the same time a minimum range of ductility specified for paving purposes.
- 2.Increasing the durability of asphalt concrete mixtures to resist the aging and weathering conditions to provide improved adhesion of asphalt to aggregate in the presence of water and water vapour to reduce stripping.
- 3.Decreasing the disposing of wastes that cause considerable damage to the environment since polypropylene material formed in (1991) as 13.2% by wt. from the total production of plastic materials.

These objectives were achieved by treating asphalt with different percentages of pyrolysis polypropylene through physical mixing with asphalt.

The modified binders were subject to some standard test such as rheological properties, durability test, as well as to some other tests related to the homogeneity of the system. Modified asphalt concrete mixtures were subject to Marshall test at 60 °C and to the indirect tensile strength test at two degree of temperature namely (25°C and 60 °C).

Background

Al-Dubabe and Al-Abdul Wahhab *et al* [1] tried to evaluate the effect of the polymer modified Arab asphalt. They collected asphalt binders from four refineries in the Gulf Countries. They found that the addition of polypropylene was increase the softening point of original asphalt by (5%) They added that polypropylene modification is effective in improving the rheological properties of neat Arab asphalt binders.

Al-Gannam K. [2] studied the physical properties of asphalt–polyethylene mixtures. He concluded that the softening point increases as the concentration of polyethylene increases, while the penetration and homogeneity decreases. The ductility remains (100⁺) up to (8%) polyethylene while sharply decreases after this certain point.

Al-Hadedy A.IJ. [3] Studied the rheological properties of asphalt–polyethylene mixes .He found that the softening point increases as the

concentration of polyethylene increases the ductility values remains (100^+ cm) up to (11%) polyethylene content, results indicated that polyethylene and asphalt are compatible to extent, while he concluded that the inclusion of polyethylene with asphalt concrete mixtures gave a quit satisfactory results (i.e. in terms of stability values, other Marshall properties and the adhesion between asphalt and aggregates ,were improved ,stripping phenomenon was reduced).

Al-Layla [4] studied the rheological properties of asphalt treated with reclaimed rubber tire (CRT). She concluded that the resultant binder is characterized by lower sensitivity to heat compared to unmodified asphalt, the improved binder exhibited better resistance to aging and weathering conditions,as well She added that some of the prepared samples are characterized by a relatively higher degree of penetration at low temperatures compared to those of unmodified asphalts.

Lobna A.S. [5] studied the rheological properties of low density polyethylene- asphalt binder at two mixing period (4&24hr.). She concluded that the blending time of low density polyethylene with asphalt results in an increase of homogeneity and enhancement of the rheological properties of the resultant binder, she stated that low density polyethylene exposure to thermal degradation leads to keep ductility values of (100^+) up to (8%)wt. low density polyethylene.

Ramzi and Galal [6] were tried to investigate the potential use of calcined and green (uncalcined) dusts as modifiers in hot mix asphalt paving mixtures. They found that:

- 1-Both coke dusts seem to improve asphalt cement performance (high stiffness) at high temperatures (58 and 64°C) with minimal adverse effects (increased stiffness)at low temperatures(-12 and -18°C).
- 2-At intermediate temperatures, penetration values at (25°C)will decrease and dynamic shear values at(19 and 22°C) generally will increase as a result of coke dust addition.
- 3-Both coke dusts will act as reinforcing agents for the asphalt cement binders .
- 4-There is a slight improvement in retention of asphalt coating when coke dust is used.

Uttpal Dutta [7] made an attempts to examine the feasibility of automotive shredder residue (ASR) as road material additive. He studied the compatibility and rheological properties of(ASR) modified asphalt and he found that:

- 1-The glass temperature and photomicrographs of (ASR) modified asphalt indicated that (ASR) and asphalt are compatible to some extent.
- 2-Viscosity ratio at(60°c) indicated that the oxidation process of asphalt can be reduced by the addition of (ASR).

Wladysi Milkowski [8] used polyethylene as an additive to achieve asphalt concrete of much higher stability and lower thermal susceptibility. He found that the addition of polyethylene in small percents, reduce penetration, increased the softening point and increased the shear strength of asphalt joints.

Materials and Laboratory Testing Materials

Polypropylene

The polypropylene (PP) were obtained from one private factory for bags production in Mosul city. The results of the physical and chemical properties of polypropylene before and after pyrolysis processes shown in table (1).

Asphalt cement

The asphalt cement used was (40-50) penetration grade taken from Baiji refinery (200 Km. North Baghdad the capital). This asphalt is usually considered the type, which have been used widely in the highway construction projects in Nineva Government. The results of the physical properties of the asphalt used are shown in table (2). The results indicated that this asphalt complies with ASTM [9] and SCRB [10] specifications for penetration graded asphalt cement.

Aggregates

Al-Kazer aggregate was utilized in the preparation of asphaltic concrete specimens. Table (3) show the results of the physical properties of this aggregate, while table (4) show the gradation limits of the aggregate used in the mix design according to the ASTM specification (D 3515) [9] for dense graded mix, and the job mix formula used.

Filler

The filler used was Portland cement and it was brought from senjar cement factory. The results of the physical properties are show in table (3).

Polypropylene-asphalt formulation

At first (PP) was subjected to thermal degradation (pyrolysis) process, mentioned by Lobna A.S [5], after grinding to powder by means of mechanical grinding, then the asphalt was mixed with different percentage of (PP) (1,2,3,4,6,& 8) %wt. for 7min at temperature of $150\pm 5^{\circ}\text{C}$. Six modified binders were thus performed from this asphalt.

Marshall mix design

All specimens were prepared according to the Marshall method of mix design, using 75 blow [10] of the automatic Marshall compactor on each side. Table (5) shows the results of Marshall mix design for the control mix (having 0% PP a additive). The optimum asphalt content was determined as the numerical average of the values of asphalt content corresponding to the maximum stability, maximum density, and 4% air voids, without violating the minimum void content requirement.

Table (1) Physical Properties of Polypropylene

property	Result	Unit
Before pyrolysis process		
Density	0.82	gm/cm ³
Chemical unit	-CH ₂ -CH-CH ₃ - [15]	-
Thermal degradation (pyrolysis) Temp.	308	°c
Color	White	
After pyrolysis Process		
Density	0.6	gm/cm ³
Melting point	156-161	°c
Decomposed	139	°c
Color	Brown	

Table (2) Physical And Chemical Properties of Asphalt Cement

Property	ASTM Designation No.	Test Condition & units	result	SCRB limits	ASTM limits
Penetration	D- 5	(25 °C, 100gm, 5 sec., 0.1mm)	42	40-50	40-50
Softening point	D-36	Ring & ball	54	51-62	50-58
Ductility	D-113	(25°C, 5 cm/min.)	150+	>100	>100
Specific gravity	D-70	(25°C/ 25°C)	1.053	-	1.01-1.06
Absolute viscosity	-	Poise	5*10 ⁶	-	-
Flash point	D-92	Cleveland open cup, °C	263	-	>240
Loss on heat	D-1754	5hrs, 163°C, %	0.25	-	0.2 Max
Asphaltene	D-2006	%	32.65	-	-

Table (3) Results Quality Tests on Aggregates

Property	ASTM Designation No.	Coarse agg.	Fine agg.	ASTM limits
L.A. abrasion	D-131	17.76	-	40 max
Bulk Sp. gr.	D-127	2.634	2.561	-
Apparent Sp-gr.	D-128	2.669	2.584	-
Apparent Sp-gr. Filler	D-128	-	3.15	-
% water absorption	-	0.503	2.354	4.0 max

Table (4) Gradation of Aggregates

Sieve size	% passing(ASTM)	Job mix formula	Tolerance
1 in	100	100	± 6%
3/4 in	90-100	95	± 6 %
3/8 in	56-80	68	± 5%
No. 4	35-65	50	± 5%
No. 8	23-49	36	± 4%
No. 50	5-19	12	± 3 %
No. 200	2-8	5	± 1.5%

Table (5) Marshall Results of Conventional Mixture (0% PP)

Asphalt content	Unit weight kg/m ³	Stability kg	Flow mm	Rigidity ratio kg/mm	% air voids	V.M.A	V.F.B
4.0	2320	886.9	2.8	316.75	7.2	14.9	51.7
4.5	2346	1024.5	2.94	348.47	5.6	14.4	61
5.0	2368	1095.8	3.3	353.48	4.0	14	70.7
5.5	2361	1030	3.6	286.2	3.7	14.6	74.7
6.0	2350	940.9	4.0	235.3	3.6	15.5	76.7
SCRB limits	-	700	2-4	-	3-6	14 min	60-80

Preparation of Marshall and indirect tensile strength specimens of modified concrete mixtures

An optimum asphalt content of 5% as found from Marshall conventional mix design (by wt. of total aggregate and filler) was used in preparing all other polypropylene modified mixes to maintain consistency through the study.

The following steps are performed for the formulation of compacted specimens:

1. The combined aggregate and filler were heated to (160 °C) in controlled electrical oven.
2. The modified binder was heated up to (150°C) in an electrical controlled oven.
3. The combination were mixed by mechanical mixing at temperature of (150± 5 °C) for 1.5 min.
4. The specimen formulated were then compacted using electrical Marshall apparatus specified by ASTM (D1559) [9].

Forty-five samples for all percentages of polypropylene were fabricated. For each percent of polypropylene, three specimens were used for Marshall stability test at 60°C for 35 min. and another three samples were tested for indirect tensile strength at 25°C for 2hrs.

In addition, after water immersion at 60°C for 24hrs followed by 2hrs immersion at 25°C Three samples were subjected to tensile strength test at 25°C, then the percent loss in tensile strength and tensile stiffness modulus were determined using the equation described by Lottman [11, 12].

Laboratory testing

A series of tests were carried out on modified binders according to ASTM [9] methods to characterize the mixes designed for different percentage of (PP) as additive. The tests that were conducted include the following:

- Rheological tests, such as penetration, ductility, softening point, ...etc.
- Temperature susceptibility;

- Short-term aging using the rolling thin film oven test (RTFOT), which simulates the aging effect of the asphalt mixture production and construction.
- Compatibility test.

While the tests were conducted on asphaltic concrete mixture modified with polypropylene includes:

- Marshall test (ASTM D1559) [9], and
- Indirect tensile strength test (ASTM D4124) [9].

Discussion of Results

Rheological tests

The rheological properties of (pp/ A) binders where evaluated and the results are presented in table (6). The result indicates that PP is effective in improving the rheological properties of asphalt cement.

Examining table (6), it can be seen that PP is keeping the ductility values of (100+) up to (4%), and raised the softening point of virgin asphalt by (20.4%) at (4%) wt. PP.

Table (6) The Main In Properties of (PP/A) mix.

%PP	Penetration	Ductility	Sp. gr.	Softening point °C	P.I	Viscosity 21°C poise	Binder stiffness Sb*10 ⁶ N/m ²	%loss wt. of heat
0	42	150+	1.0537	54	-0.644	5.10 ⁶	190.7	0.25
1	39	133	1.05	55.5	-0.478	5.78.10 ⁶	207.8	0.21
2	35.5	117.5	1.0473	57	-0.365	7×10 ⁶	236	0.18
3	31.5	115.0	1.046	59	-0.118	9.2×10 ⁶	268.5	0.15
4	29	101.5	1.0466	65	+0.71	11×10 ⁶	304.3	0.137
6	23.7	82	1.042	66	+0.475	17×10 ⁶	351.7	0.121
8	21.5	74	1.037	75	+1.67	21×10 ⁶	287.2	0.105
SCRB limits	40-50	>100	-	51-62	-	-	-	-
ASTM limits	40-50	>100	1.01-1.06	50-58	-	-	-	0.2 max

Temperature susceptibility

The penetration index relationship was used to investigate the influence of addition of PP on temperature susceptibility of asphalt cement using the following equation mentioned by Yang [13]:

$$penetration\ index(P.I) = \frac{20 - 500A}{1 + 50A}$$

$$A = \frac{\log(Pen.@T) - \log 800}{T - T_{R\&B}}$$

Where

T= Testing temperature &

T_{R&B}=Ring and Ball softening point

Fig (1) illustrates the relationship between the penetration index and PP contents. It can be seen that all modified binder are less susceptible to temperature changes than virgin asphalt cement.

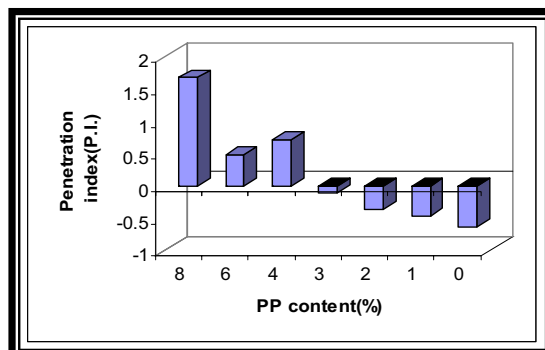


Fig. (1) Relationship between Polypropylene content and Penetration

Aging susceptibility of (PP) binders

Fig. (2) and (3) show the ductility and softening point properties of aged polypropylene –asphalt binders.

From these figures, and from table (7) it can be seen that the ductility decreases, while softening point increases, moreover the values of ductility are remain (100⁺) up to (4%)PP content.

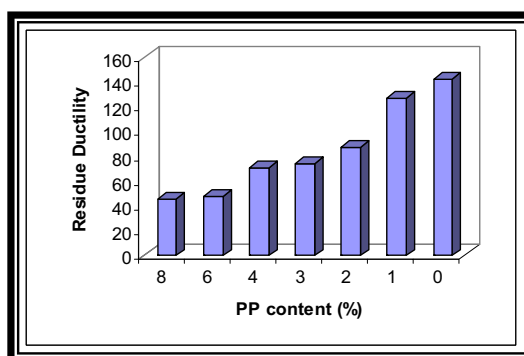


Fig. (2) Relationship between Polypropylene content and Residue

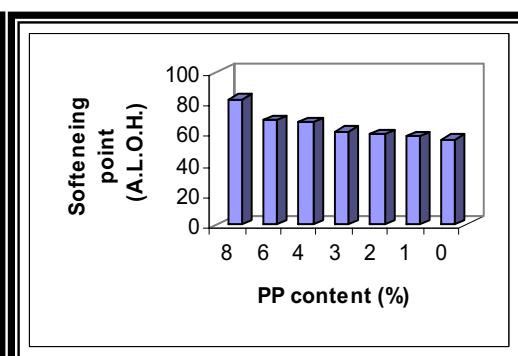


Fig. (3) Relationship between Polypropylene content and Aged Softening

Table (7) Test Results of Aged (PP/A) mixes

%PP	% loss of wt.	Residue Penetration (25°C, 100gm, 5sec.)	Residue Ductility (25°C, 5cm/sec.)	Softening point °C	Aging index
0	0.25	37	142	56	0.88
1	0.21	35	126.75	58	0.897
2	0.18	33.2	87	59.5	0.935
3	0.15	29.5	74	61	0.936
4	0.137	26	70.5	67	0.896
6	0.121	21.1	47.5	68.5	0.89
8	0.105	17.5	45.5	82	0.813

Fig (4) shows that the percent loss of (PP) binders decreases as the PP content increases, this is related to that the polyethylene was occupied a space of total mix, and cause reduction in asphalt volume, which leads to decrease in loss by dehydrogenation and oxidation of asphalt in the mix (i.e., durability increased slightly with the addition of PP content in the mix).

The hardening of modified binders was determined by the penetration of residue after exposure to heat and air as shown in fig (5). Aging was measured by aging index using the following equation:

Aging Index= (Residue penetration after aging at 25°C/Original penetration at 25°C). It can be seen from fig.(6) that aging index increases with increase in PP content, due to the increasing in bonds between PP and asphalt ,resulting in prevention of the brittleness of the resultant binder.

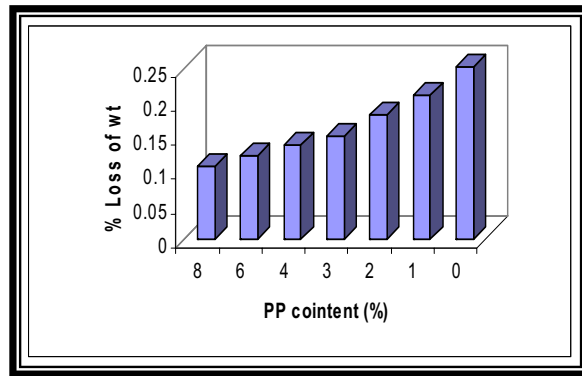


Fig. (4) Relationship between Polypropylene content and percent Loss of wt

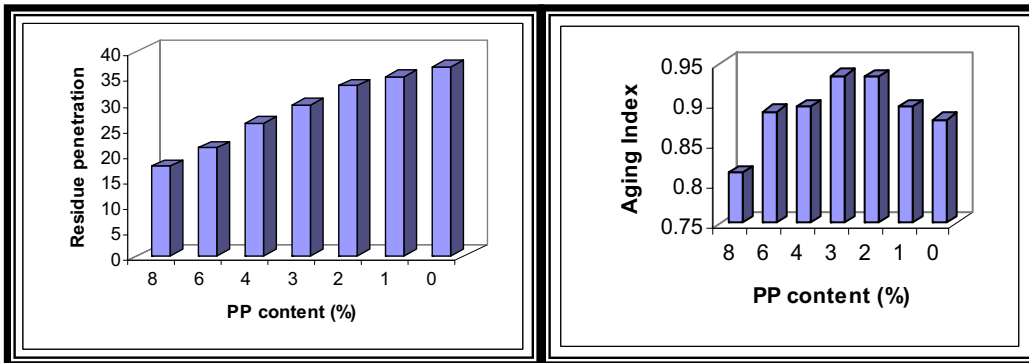


Fig. (5) Relationship between Polypropylene content and Residual Penetration

Fig. (6) Relationship between Polypropylene content and Aging Index

Polypropylene –asphalt compatibility

Light microscopy was used to evaluate the compatibility between PP and asphalt. The microstructures of asphalt and modified asphalt with PP were observed on a light microscope and shown in Fig. (7). Because of the identical photographs of four percent of PP which are (1, 2, 3 & 4%), are therefore one of them is listed and discussed herein.

The photomicrographs demonstrated that polypropylene is well dissolved in the asphalt matrix at temperature of (150°C) for all contents, which cause the reduction in asphaltene percentage from (32.65%) to (8.5%) at (4%) PP and as shown in Fig. (8) & table (8).

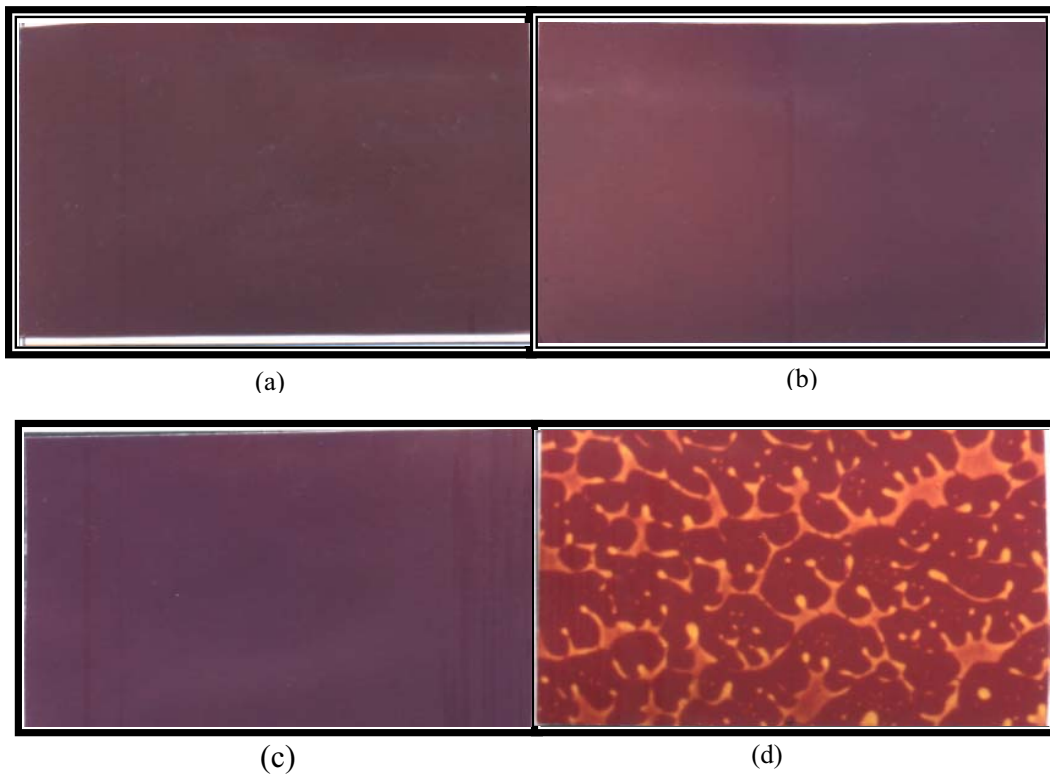


Fig. (7) The Photomicrographs of Polypropylene Modified Asphalt (a-0%, b-4%,c-6%,d-8%)

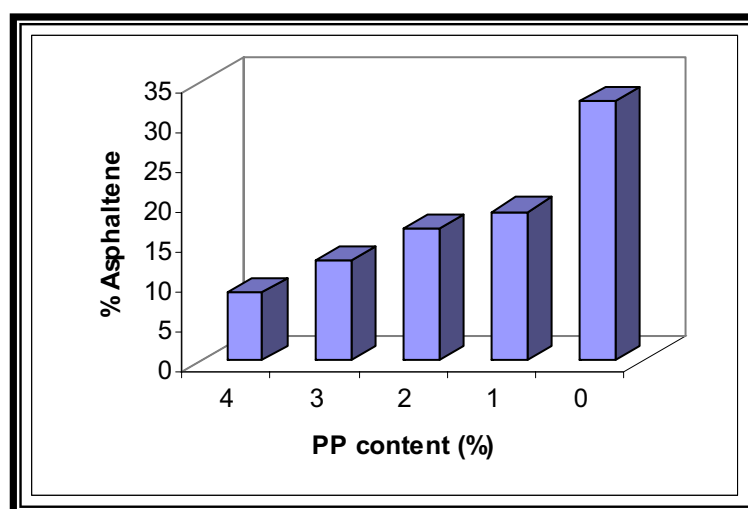


Fig. (8) Relationship between Polypropylene content and Asphaltene percent

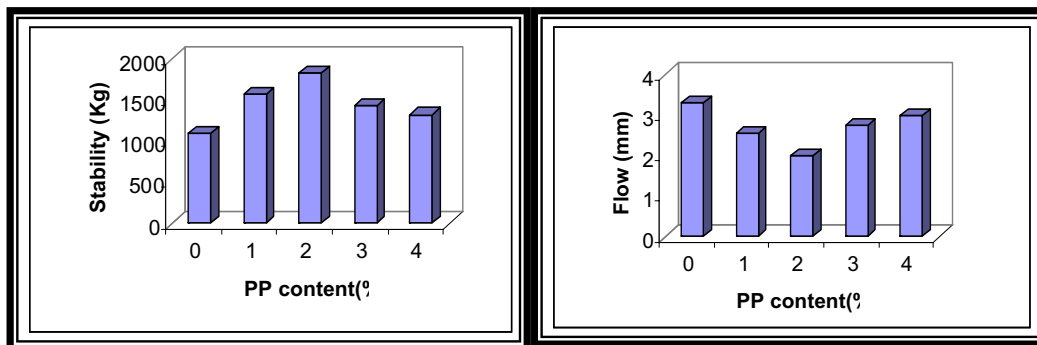
Table (8) Asphaltene Results of (PP/A) Mixes

% PP	% asphaltene
0	32.65
1	18.75
2	16.7
3	12.5
4	8.5

Marshall and indirect tensile strength tests

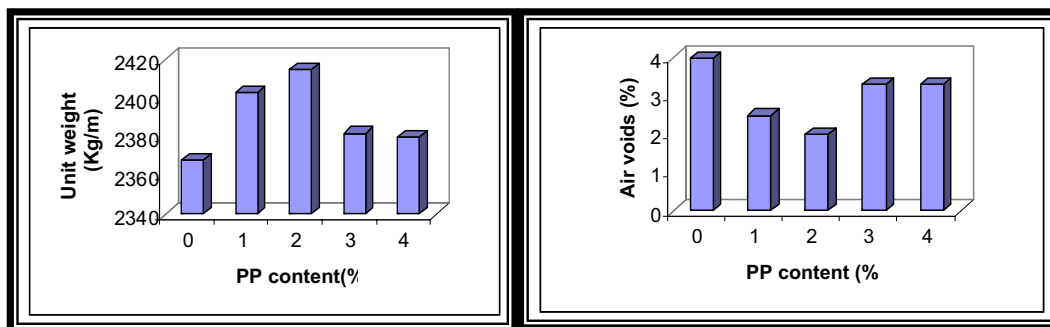
The relationships between Marshall properties and polypropylene content were plotted at optimum binder content (i.e. 5% wt of total agg. and filler) as shown in Fig (9).

Fig. (9 a&b) represents the effect of polypropylene content Marshall stability and flow. The Figure indicates that as (PP) content increase the stability increase and the flow decrease up to (2%PP). This attributed to the specific gravity of polypropylene which is less than that of pure asphalt, which serves to penetrates between particles and enhanced interlock of aggregates which increase stability and decrease flow. Beyond (2%)PP content the stability decrease and the flow increases.



(a)

(b)



(c)

(d)

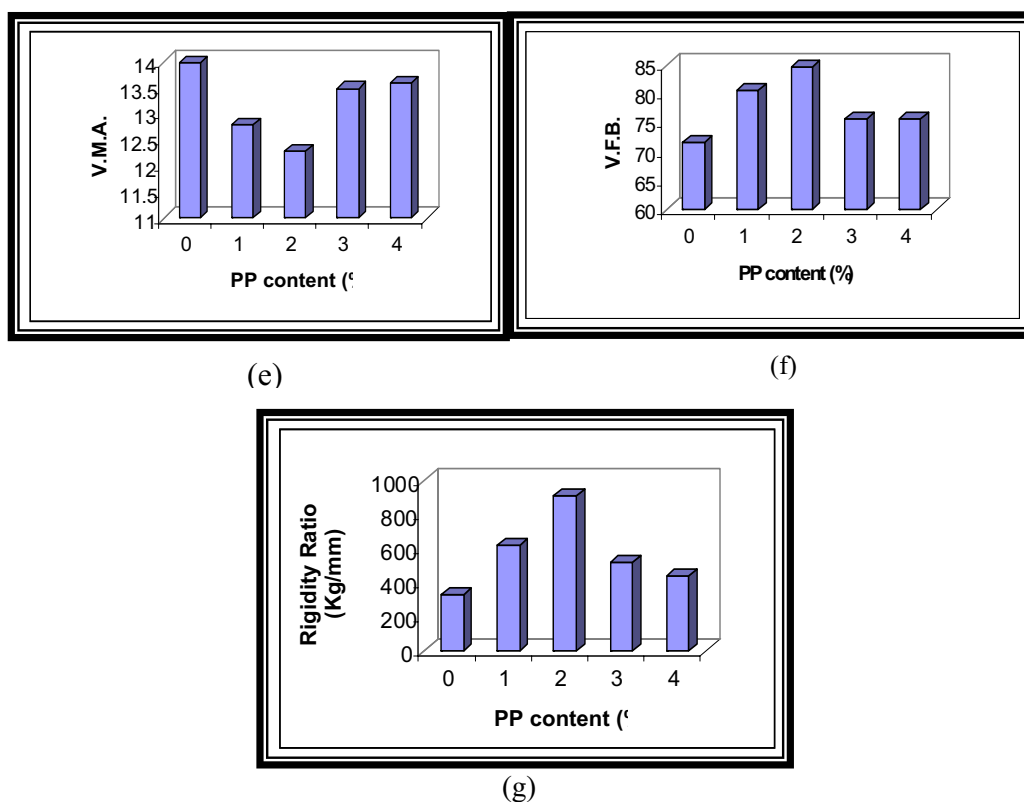


Fig.(9). Relationship between Polypropylene Content And Marshall Properties

Fig. (9 c&d) shows that unit weight increased and air voids decreased up to (2%) PP content due to the filling property attributed by polypropylene addition. After this percent the unit weight decreased and air voids increased.

Fig. (9e & f) shows the relationships between V.M.A. and V.F.B. versus polypropylene content, the V.M.A decrease with the increase in (PP) content and reaches a minimum value at 2% PP content. At higher polypropylene content the increase in polypropylene content cause an increase in V.M.A.

Examining table (9) indicates that 4% PP content satisfies the specified limits of (3-6%) of Air voids, (60-80%) of V.F.B., and (2-4 mm) of flow while the V.M.A. at this PP content is complying with the minimum specified limit (13-15%) of V.M.A mentioned by Asphalt Institute MS-2 [14] for 19 mm(3/4in mix).

Table (9) Marshall Result of (PP/A) Mixtures.

% PP	Unit weight kg/m ³	Stability kg	Flow mm	Rigidity ratio kg/mm	% air voids	V.M.A	V.F.B
0	2368	1095.8	3.3	332	4.0	14	71.5
1	2403	1567	2.54	617	2.5	12.8	80.5
2	2415	1825.4	2.0	912.7	2.0	12.3	84.7
3	2382	1439.5	2.76	521.6	3.3	13.5	75.6
4	2380	1328	3.0	442.7	3.31	13.6	75.7
SCRB limits	-	700 min.	2-4	-	3-6	14 min	60-80

Indirect tensile strength test

From the sited results in table (10), it can be concluded that addition of PP increase the adhesion between aggregate and asphalt, which leads to decrease in stripping of asphalt concrete mixtures and decrease in the horizontal deformation, and increase the tensile stiffness modulus values.

Table (10) Indirect Tensile Strength Results of (PP/A) Mixtures

% PP	Test Temperature				
	25 °C		60 °C		
	Horizontal deformation mm	Tensile Stiffness modulus Mpa	Horizontal deformation mm	Tensile Stiffness modulus Mpa	% Loss in strength
0	1.143	74.3	1.397	64.8	19.8
1	0.558	90.7	0.762	70.4	14.8
2	0.508	99.7	0.635	71.9	10.3
3	0.431	107.3	0.533	78.0	9.8
4	0.355	110.7	0.431	88.9	6.71

Conclusions & Recommendations**Conclusions**

Based on the testing and analysis presented, the results of the study warrant the following conclusions:

A) A review of the standard binder testing and grading results indicated the following.

1. Penetration at 25 °C generally will decrease as polypropylene content in increased, which indicates an improved shear resistance in medium to high temperatures.
2. The addition of polypropylene kept the ductility values at minimum range of SCRB specification at (100⁺cm) up to 4% PP content.
3. Softening Point tend to increase with the addition of polypropylene, which indicates improvements in resistance to deformation.
4. Polypropylene seem to improve binder performance viscosity (stiffness) which tend to increase with the addition of polypropylene provided that rutting, shoving probably would be reduced in hot mix asphalt concrete mixtures.
5. The percent loss of air and heat decreases with the addition of polypropylene (i.e. the resistance of asphalt to the action of temperature and temperature changes, and the action of heat increases);
6. The photomicrographs indicated that polypropylene and asphalt are compatible to extent; and
7. Penetration index values indicated that polypropylene was reduced the temperature susceptibility of asphalt.

B) A review of the Marshall and indirect tensile strength mixes design result indicated the followings:

1. Marshall stability, Marshall stiffness, V.F.B. and density values will increase as a result of adding polypropylene while Marshall flow, Air voids and V.M.A tend to decrease.
2. There is a slight improvement in the retention of asphalt coating (i.e. reducing bleeding phenomenon) when PP/A binder is used due to the adhesiveness of polypropylene material;
3. The stiffness determined from the mechanical properties of Marshall and indirect tensile strength tests indicated that these mixtures may resist the pavement deformation forces, rutting and shoving, therefore it should be used on busy intersection or truck stops and parking lots where standing loads cause extended periods of such deformation; and
4. The percent loss in tensile strength due to immersion test will decrease as a result of adding polypropylene, which indicates that PP/A concrete mixtures are less susceptible to stripping phenomenon.

Recommendations

Based on the finds of this study, the following recommendation and comments may be offered.

1. For coarse or moderately coarse (close to the middle ASTM curve) aggregate gradation, the air voids content should be kept around 3%;
2. The short-term aging using the rolling thin film oven test (RTTOT) according to ASTM (D1754), indicated that high content of polypropylene (i.e. 3&4% PP) tend to separate from the base asphalt at temperature of 163 ° C for 5hrs. This places a handling storage, and mixing limitations on the polypropylene modified asphalt binder, therefore blends must always be prepared at temperature of (155±5 C), and mixing temperature of modified binder with other mix component should be kept around (150-160 C);
3. The modification binder with PP should be mixed thoroughly before mixing with aggregate and filler materials.

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