# Effect of Mixing Process on Polypropylene Modified Bituminous Concrete Mix Properties

Noor Zainab Habib, Ibrahim Kamaruddin, Madzalan Napiah and Isa Mohd Tan

Abstract— This paper presents a research conducted to investigate the effect of mixing process on polypropylene (PP) modified bitumen mixed with well graded aggregate to form modified bituminous concrete mix. Two mode of mixing, namely dry and wet with different concentration of polymer polypropylene was used with 80/100 pen bitumen, to evaluate the bituminous concrete mix properties. Three percentages of polymer varying from 1-3% by the weight of bitumen was used in this study. Three mixes namely control mix, wet mix and dry mix were prepared. Optimum binder content was calculated considering Marshall Stability, flow, air voids and Marshall Quotient at different bitumen content varying from 4% - 6.5% for control, dry and wet mix. Engineering properties thus obtained at the calculated optimum bitumen content revealed that wet mixing process is advantageous in comparison to dry mixing as it increases the stiffness of the mixture with the increase in polymer content in bitumen. Stiffness value for wet mix increases with the increase in polymer content which is beneficial in terms of rutting. 1% PP dry mix also shows enhanced stiffness, with the air void content limited to 4%. The flow behaviour of dry mix doesn't indicate any major difference with the increase in polymer content revealing that polymer acting as an aggregate only without affecting the viscosity of the binder in the mix. Polypropylene (PP) when interacted with 80 pen base bitumen enhances its performance characteristics which were brought about by altered rheological properties of the modified bitumen. The decrease in flow with the increase in binder content reflects the increase in viscosity of binder which induces the plastic flow in the mix. Workability index indicates that wet mix were easy to compact up to desired void ratio in comparison to dry mix samples.

*Keywords*— Marshall Flow, Marshall Stability, Polymer modified bitumen, Polypropylene, Stiffness.

## I. INTRODUCTION

**B**ITUMINOUS binders are widely used by paving industry [1], where its worldwide use reaches to almost 100Mt [2]. Continuous increase of wheel loads, tyre pressure and change in climatic condition severely affect the performance of bituminous mix pavements. Modification of virgin bitumen with synthetic polymer binder is considered as solution to overcome the problems arises because of the increase in

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Isa Mohd Tan, Associate Professor, Chemical Engineering Dept., Universiti Teknologi PETRONAS(e-mail; isa tan@petronas.com.my) wheel load and changing climatic conditions [3]. Polymer modification is considered as one of the solution to improve fatigue life, reduce rutting and thermal cracking in the pavement [4].

Rutting is the most common pavement distress related to load and temperature associated problem in pavement engineering. In order to improve the problem of rutting, use of modified binders is taken as a solution. Polymer modification is usually used to improve the temperature susceptibility of bitumen by increasing binder stiffness at high service temperatures and reducing the stiffness at low service temperatures [4]. Most commonly used polymer globally include approximately 75% elastomeric modified binder, 15% plastomeric and remaining 10% belongs to either rubber or other modification [5].Polymer modification is also considered as one of the solution to improve fatigue life, reduce rutting and thermal cracking in the pavement[4].

Thermoplastics, when used as modifier alters mechanical properties of the mixture by enhancing its mechanical behavior in significant manner [6]. Polymer whether added directly into the mix (dry mixing) or blending of polymer with bitumen (wet blending) significantly alters the rheological behavior of binder.

Bitumen when blended with polymer forms a multiphase system, a phase rich in asphaltenes not absorbed by the polymer [7] which enhances the viscosity by the formation of more complex internal structure [8]. While mixing either using mechanical or chemical method the differences in molecular weight and polarity of base bitumen and polymer has critical affect on compatibility [9]. Thus thermoplastic when used as modifier gives rigidity to the binder and reduces the deformation under load [8]. The affect of this is more profound when the concentration of thermoplastic was kept below 1% by weight of the base bitumen [10]. It also gives better results in terms of Marshall Stability, resilient modulus, water susceptibility and fatigue life when the concentration of polymer was kept up to 2.5% [7]. Thermoplastic when mixed with bitumen even at ambient temperature increases the viscosity and thus stiffness at service temperature but unfortunately do not show any significant elastic behavior [9].

Thermoplastic morphology is strongly affected under stress and deformation as sliding of chains with respects to entanglements occurs at the nodes [10]. Addition of bitumen improves deformation resistance as the viscosity of blend is enhanced tremendously which is observed with increase in softening point and decrease in penetration values [7]. When a polymer and compatible base bitumen are mixed, the polymer strands absorbs part of the low molecular weight oil fraction of

the base bitumen and become swollen, the swollen strands connect together at nodes and form a three dimensional network which significantly affects the mechanical properties of the binders and ultimately the bituminous binder mixes [11].

This study focuses on the use of thermoplastic polymer namely polypropylene (PP), as modifying agent for the 80/100 pen base bitumen. Concentration of polymer in the mix was kept till 3% by weight of the bitumen. Different mixing processes namely dry and wet were used in order to compare the obtained resultant engineering properties which in fact used as indicator of mechanical behavior of modified mix. Three mixes namely control mix, wet mix and dry mix were prepared at 170°C in two ways to know the behavior of mix. In wet mix, polymer was blended separately with bitumen and then added to aggregate. In dry mix, polymer was added with aggregate and allowed to mix for 15mins before adding bitumen to the mixer. The objective of dry mixing was to overcome the problem of storage stability, oxidative ageing and economy in terms of reducing the cost of blending of bitumen with polymer at blending facility. The different blending process hence provides an overview of the effect of mixing processes on the mechanical strength parameter obtained from different modified bituminous mix.

#### II. METHODOLOGY

#### A. Materials

Materials used in this study includes 80/100 penetration grade base bitumen obtained from PETRONAS refinery at Malacca, Malaysia. The polymer Polypropylene Resin PP used for modification was supplied by Polyethylene Malaysia, was in powder form. Crushed aggregates used in this study include crushed granite obtained from quarry located at outskirt of Ipoh, Malaysia, while river sand was used as fine aggregate. Ordinary Portland cement was used as filler. Physical properties of material used are shown in Table I.

## TABLE I PHYSICAL PROPERTIES OF MATERIAL USED

	Coarse Agg	Fine Agg	Polymer	Bitumen	Filler
Specific gravity	2.63	2.64	0.887	1.02	3.26

#### B. Sample Preparation

## 1) Polymer modified bituminous blend (PMB)

PMB blend was prepared by mixing about 400gm of bitumen with polymer in shear mixer at 120rpm, while the temperature was kept at 170°C as the melting temperature of polypropylene lies between 160-166°C. The concentration of PP was kept as1-3% by weight of bitumen. Mixing was continued for 1 hr to produce homogenous mixtures. The modified bitumen was then sealed in containers covered with

aluminum foil and stored for further testing and Marshall Sample preparation. Empirical test such as penetration, softening point and viscosity were then conducted on the prepared samples. For dry mixing polymer was added directly with aggregate in mixer and was allowed to mix for 15 minutes before addition of bitumen.

#### 2) Marshall Samples

All Marshall Samples were prepared according to ASTM D1559. Standard size 101.6mm (4 in) diameter and 60 -65 mm (2½ in) high samples were prepared by using Gyratory compactor. Gyratory compactor was adjusted at an angle of gyration of 1.25°, normal pressure of 600 kPa and number of gyration was kept at 200 according to ASSHTO 2001 specification. For Marshall Sample preparation, 1200gms of well graded aggregate according to JKR specification (Jabatan Kerja Raya Standard, Malaysia) for ACW-14(asphalt concrete wearing course, as shown in Table II) was blended with bitumen at varying bitumen percentage between 4-6.5%, in order to get the optimum bitumen content for a mix. Three different types of Marshall Samples were prepared namely control mix prepared with virgin bitumen, wet polymer modified Marshall Samples using polymer modified bitumen with varying polymer concentration between 1-3% by weight of bitumen, where blending of PMB was done before mixing with aggregates and dry blend modified Marshall Samples, prepared by adding polymer (1-3% by weight of bitumen) directly with the aggregates before adding bitumen. Engineering properties calculated at optimum binder content for all three types of mixes, were then compared with JKR standard values as mentioned in Table VI.

#### C. Laboratory testing

Test on the prepared wet blend and virgin bitumen were conducted according to ASTM Standard method. The empirical test includes penetration at 25°C, softening point and viscosity test. The test results are presented in Table III and Table IV.

## 1) Penetration (ASTM D-5)

The standard 100g, 25°C and 5sec penetration test was performed both on base bitumen and PMB with the concentration of polymer varying between 1%-3% by weight of the bitumen. The results are shown in Table III and Table IV.

## 2) Softening Point (ASTM D-36)

Ring and ball standard test was used to determine the consistency of the bitumen, which represent the temperature at which a change of phase from solid to liquid occurs. The results are shown in Table III and Table IV.

TABLE II

Sieve size	Gradation limits	Gradation used
14mm	80 - 95	87
10mm	68 - 90	77
5 mm	52 - 72	60
3.35 mm	45 - 62	51
1.18 mm	30 - 45	34
425µm	17 -30	19
150µm	7 -16	12
75 μm	4 - 10	6

TABLE III

CONVENTIONAL PROPERTIES OF BASE BITUMEN				
Grade	80/100 pen			
Penetration (dmm)	84			
Softening point °C	53			
Penetration Index	0.5			
Viscosity (Pa s)	0.44			

TABLE IV

P	PROPERTIES OF PP MODIFIED BITUMEN				
Polymer cont	Pent at 25°C		Visc. at 135°C		
%	dmm	°C	Pa s		
1	34	54	0.78		
2	30	55	0.805		
3	28	55	0.83		

## 3) Viscosity (ASTM D-4402)

Viscosity test was conducted using BrookField viscometer on virgin and polymer modified bitumen with concentration varying between 1-3%. The dynamic viscosity measurement was made at 135°C with increasing shear rates (shear stresses). The viscosity test result is shown in Figure 1.

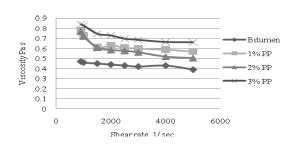


Fig. 1 Viscosity of Bitumen and PP modified bitumen at 135°C

#### 4) Marshall Test (ASTM D-1559)

The Marshall test was carried out according to ASTM D1559 on all the three different bituminous mix concrete samples namely control wet and dry mix to calculate optimum binder contents (OBC) and to select a mix proportion for further work. Marshall Properties used for determination of OBC include density, air void, stability and Marshall Quotient (stiffness). Engineering properties evaluated at OBC, from Marshall Test result was only used for comparative purpose without intention of taking as performance indicator of the mixes. Compaction effect of gyratory compactor was considered for evaluation of workability Index considering with the decrease in height of specimen for 200 gyration, which ranks the mixes on the basis of ease in compaction and laying at temperature of 135°C at site. The test results are presented in Table V.

TABLE V ENGINEERING PROPERTIES OF CONTROL, WET AND DRY MIX BITUMINOUS CONCRETE MIXTURES AT OBC

_	Control Mix	1% PP Dry mix	1% PP Wet mix	2%PP Dry mix	2%PP Wet mix	3%PP Dry mix	3%PP Wet mix
OBC %	5.0	5.2	5.0	4.7	5.2	4.5	5.2
Density Kg/cm <sup>3</sup>	2.38	2.33	2.35	2.29	2.26	2.14	2.39
Air Void%	3.49	3.9	4.61	7.0	3.42	9.2	3.79
Stability (kN)	12.64	12.45	15.38	10.6	17.03	9.1	18.82
Flow (mm)	2.3	2.22	3.01	2.3	3.2	2.0	2.86
Stiffness (kN/mm)	5.49	5.6	5.11	4.6	5.32	4.55	6.58
WI	9.41	8.0	7.47	6.5	7.77	6.0	8.28

TABLE VI SPECIFICATION OF JKR MALAYSIA FOR AC MIXTURES

Properties of Marshall Specimen		Specification limits
Stability	>	8kN
Stiffness	>	2kN/mm
Flow (mm)	>	2.0 - 4.0
Air Void %		3 - 5
Void filled with bitumen %		70 - 80

## III. ANALYSIS OF RESULTS

## A. Penetration Results

From the result shown in Table III and IV for virgin bitumen and PP modified bitumen, there is a sharp decrease in the penetration value of 84 dmm for virgin bitumen to 34dmm for 1% PP concentration shows that increase in the hardness of

the PMB was because of the use of the high molecular weight polymer PP with melt flow Index of 8g/10 min, enhances the hardness of PMB. It is obvious from the observation that thermoplastics influence more on the penetration with the increase in the viscosity of the bitumen [1] as can be observed by the decrease in the value of penetration with the increase in concentration of polymer. The melting temperature of PP is 165°C, thus it absorbs some oil from the bitumen and releases low molecular weight fraction into the bitumen which increases the viscosity of the PMB[12], thus by the end of mixing process and by the time it cools harden mixture was formed. The hardening of the bitumen can be beneficial as it increases the stiffness of the material, thus the load spreading capabilities of the structure but also can lead to fretting or cracking [1].

## B. Softening Point Results

The results obtained from the ring ball shown in Table III and Table IV shows that there is no great difference in the softening temperature for virgin bitumen and Polypropylene modified bitumen PMBs up to 3% concentration of polymer as thermoplastics modification does not significantly affect the softening point as compared to the penetration [1]. One of the cause was the internal structure formed by polymer seems to be thermodynamically stable which do not significantly affect the softening point of the PMB. This can be considered as positive sign as it may enhances the pavement performance characteristics in terms of rutting, fatigue and temperature susceptibility. Homogeneity which was achieved during blending of lower concentration of polymer PP with base bitumen would be good in terms of storage stability and it may offer better rutting resistance at higher temperature.

## C. Viscosity of Bitumen and PMB blend

The flow behavior of the material described in terms of viscosity, exhibits Newtonian and non- Newtonian characteristics depending on the composition and source of the crude. Temperature and loading time also affect the behavior describing the viscoelastic properties of the material. The internal structure of the base bitumen also play key role [13]. Bitumen with viscosity of 0.44 at 135°C shows increase in viscosity with the increase in polymer concentration. The non-Newtonian characteristic with the decrease in viscosity with the increase in shear rate was observed for all concentration of polymer in bitumen. These non-Newtonian phenomena are dependent on the shear rate and it influences the internal structure of the PMB [13]. From Figure1, it was observed that the non-Newtonian behavior with the decrease in viscosity with increase in shear rate was more prominent for 3% PP modified bitumen. The fluctuation of viscosity for 1% and 2% PP modified bitumen was also observed. This mixed behavior with the decrease in viscosity with increase in shear rate and again increase in viscosity do not truly represent non-Newtonian behavior but it represent thixotropic effect. The thixotropic behavior of modified bitumen may be due to reversible breakdown of structure which is commonly found in

multiphase system of polymer modified bitumen [14]. Thus it is very difficult to say that polymer modified bitumen purely exhibit shear thinning phenomenon with the decrease in viscosity with the increase in shear rate as shear thickening phenomenon was also observed. This pseudo plastic behavior of PMB may be attributed due to breakdown of structure, or polymer particles existing in equilibrium state being more aligned offering lesser resistance to flow but with the increase in shear rate, these offers higher resistance due to agglomeration, aggregation or flocculation of particle in multiphase system as the interparticle forces like Brownian, van der Walls forces become prominent [6]. This instability reflect the tendency of PMB to morphological phase separation which is due to Brownanian coalescence followed by gravitational flocculation, which ends up with creaming effect [3]. As this viscometer did not allow measurement of viscosity at both higher and lower shear rate, true statement about the shear thickening or shear thinning cannot be made. Thus expectation can be made for PP modified bitumen as rearrangement of molecules were observed more for it by linear decrease in viscosity with the increase in shear rate.

#### D. Marshall Test Results

The Marshall Test result of control mix when compared with the modified mixes revealed that wet blended mixes seems to be more promising in terms of all engineering properties in comparison to dry mix as presented when compared with control mix as shown in Table V.

## 1) Density of bituminous concrete samples

The density of the mix has strong influence on the in service life of the pavement [15]. From Figure 2 decrease in density was observed for both dry and wet modified mixes in comparison to control mix. Mixes with the higher internal resistance offers resistance to densification as observed for both wet and dry PP modified samples except for 3% PP wet mix which is almost equal to control mix. For dry mix sample the arrangement of aggregates and dry polymer resin doesn't allow further densification at same energy level of compaction. This trend of decrease in density for wet modified samples may happened because of increase in viscosity of binder causing hindrance in sliding of aggregate against rearrangement during densification caused by the application of constant vertical pressure of gyratory compactor. For dry mix modified samples the polymer resin acts as an additional aggregate material offering resistance to densification. Another cause of densification is bitumen when added to dry mix doesn't necessarily form multiple layer on the aggregate or polymer due to lack of adhesion or absorption of binder after the formation of first monolayer on the surface of the aggregate. This is also being confirmed by the decrease in flow values for dry mixed samples. For 1% PP dry mix sample results seems to be promising if the air void content doesn't exceed the limitation of 3-5 % as mentioned in JKR standard specification. For the rest of dry sample the increase in air

voids also reflects as one of the cause of decrease in density. In short values of density below or nearer to control mix density revealed that there is still a room for further densification and would be beneficial for the pavements which are subjected to unexpected excessive wheel load if the criteria for air void content at 4% can be maintained.

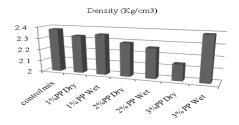


Fig. 2 Density Vs Binder content for Control, Wet and Dry PP Modified bitumen

## 2) Flow of Bituminous concrete samples

From Figure 3 it can be observed that flow values for wet blend samples are higher than control mix, while for dry mixed samples flow values up to 2% polymer content remain same. These results revealed that for wet blend samples binder acts as lubricating agent allowing sliding of aggregates with each other thus enhancing the elastic and plastic properties of the bituminous concrete mixture. For 3% PP wet mix samples decrease in flow was observed which may be due to agglomeration of polymer resisting the sliding past of particles as observed from viscosity test results (Fig .1). For dry mix decrease in flow was observed for 3% PP dry samples. Thus the better stiffer initial mix with improved flow properties is beneficial with better distribution of load with lesser permanent deformation [16].

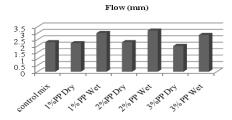


Fig. 3 Flow Vs Binder content for Control, Wet and Dry PP Modified bitumen

## 3)Stability of Bituminous concrete samples

From Figure 4 it is seen that stability values for control, wet and dry mix samples satisfy the requirement of JKR standard (Table VI). Significant increment in stability values in comparison to control mix was more pronounced for all wet mix samples. The enhanced viscosity of wet blend modified binder is one of the cause of the increases the stability of the mix. For dry mix samples decrease in stability was more pronounced for 2 and 3% PP modified samples. The decrease in density with increase in polymer content for dry mix is

because of the decrease in adhesion as being confirmed by the increase in air void and decrease in density for all dry mix samples.

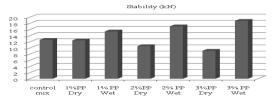


Fig. 4 Stability Vs Binder content for Control, Wet and Dry PP Modified bitumen

#### 4) Stiffness of Bituminous concrete samples

Stiffness or Marshall Quotient is a measure of the materials resistance to shear stresses, permanent deformation and thus rutting [1]. High stiffness values indicate a mix with a greater ability to spread the applied load and resistance to creep deformation[17]. From Figure 5 it was observed that in comparision to control mix, wet mix shows increase in stiffness values with the increase in polymer content. The enhanced viscosity of blended binder enhances the mechanical properties of the mix as being observed for wet mix samples. This characteristic would be beneficial in enhancing creep modulus of the mix. For dry mix samples only 1% PP dry mix sample shows a marginal increase in stiffness value where the strength obtained seems to be imparted by the binder itself with some strength obtained from dry polymer resin.

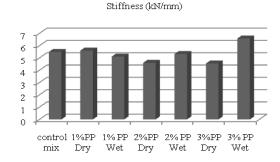


Fig. 5 Stiffness Vs Binder content for control, Wet and Dry PP Modified bitumrn

#### 5) Workability Index of Bituminous concrete samples

Workability Index is the parameter used to describe the ease with which bituminous mixture can be laid and compacted. Workability Index for both dry and wet mix samples decreases in comparison to control mix. For wet mix samples the enhanced viscosity of modified binder offers resistance to compaction but all the values for wet mix were well above the value of 6.0 as the mixture having workability index less than 6.0 offers difficulties during compaction in the field [18]. Only 1%PP dry blend samples has workability index well above the limit of 6.0, as small quantity of dry polymer in the mix behave only as filler without disturbing the mix properties. For 2 and 3% PP dry sample the added

polymer unable to form compatible network, offering resistance to densification and thus lower workability index for the mix.

#### IV. CONCLUSION

From this study following conclusions have been drawn

- It was found that mixing processes used for manufacturing of polymer modified bituminous concrete has profound effect on the engineering properties.
- Control and modified wet bituminous mixtures meets the requirement of JKR Standard for Malaysian highways.
- The rheology of binder used for wet mix as observed by penetration and viscosity tests revealed that increase in polymer content decreases the penetration and increases the viscosity of the modified binder.
- Dry mix sample doesn't show any significant increase in stiffness as polymer doesn't interact properly in short mixing time thus failed to impart any of its desired properties.
- Binder rheology significantly affects the stiffness, flow and density of the wet bituminous mix.
- Softening temperature are least affected by thermoplastic modification thus considered a positive point considering the application side.
- 3% PP modified wet bituminous mixture shows promising performance in terms density, stability and stiffness in comparison to 1 and 2% wet bituminous mixture.
- 1% PP dry bituminous mixture results are better than 2% and 3% PP dry bituminous mixture in terms of stability, flow, density and stiffness.
- Density of all wet and dry bituminous mixture is lower than control mix.
- Percent air voids for 2 and 3% PP dry bituminous samples exceeds JKR Standard specification as polymer offers resistance against densification.
- Workability index for control and PP wet modified bituminous mixture are well above the minimum level of 6.0, thus would offer ease in compaction and lay down at site.

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