STUDY ON MODIFICATION OF RECLAIMED ASPHALT PAVEMENTS BY USING SOME ACRYLATE POLYMERS

A. M. Nasser ¹, H. Abd El-Wahab¹, M. Abd El-Fattah², Abdelzaher E. A. Mostafa³, and Ahmed G. Sakr⁴

¹Chemistry Department, Faculty of Science, Al-Azhar University, Cairo, Egypt

ABSTRACT

The use of Reclaimed Asphalt Pavement (RAP) in the new blend mix reduces the need of neat bitumen, making the recycling of RAP economically attractive. This study investigates the potential use of vinyl acetate-butyl acrylate copolymer and the RAP in Hot Mix Asphalt (HMA). In fact, the twofold purpose of this research is to improve the performance of asphalt pavement and mitigate the environmental impacts caused by waste tires and aged asphalt pavement. To achieve this aim, vinyl acetate-bu-acrylate copolymer was added at different loading levels 2, 4 and 6 wt. % to the virgin bitumen and was used as a control mix for comparison. The modified binder was tested for physical characteristics, however the prepared mixes were tested and evaluated using Marshall test methods. The obtained results revealed that, the hot mix asphalt (HMA) with the highest stability, was obtained with the addition of 4 wt. % vinyl acetate-bu-acrylate copolymer, compared to the control mix design. Which indicates that, the bitumen is compatible with the acrylate copolymer.

Keywords: Acrylate copolymer; Reclaimed Asphalt Pavement; Modified bitumen; Hot mix asphalt; Marshall test.

1. INTRODUCTION

Recycling hot mix asphalt (HMA) results in a reusable mixture of aggregates and aged asphalt binder known as Reclaimed Asphalt Pavement (RAP) [1, 2]. The RAP is the residue which is created when milling damaged pavements for maintenance and rehabilitation purposes. The use of RAP as a component of the new hot asphalt mix passes sustainable development polices, and is environmentally friendly and compatible technology. Using the old asphalt bitumen in the new blend mixtures reducing the required new bitumen content, makes the use of RAP in HMA mixtures economically attractive [3, 4]. It is considered that the most economical use of the RAP is in the intermediate and surface layers of flexible pavements, because the less expensive RAP binder can replace a portion of the more expensive virgin binder [5, 6]. A hundred percent of the reclaimed asphalt can be recycled [7] with different methods: hot recycling in asphalt plant, hot in-place recycling, cold in-place recycling and full depth reclamation are the most commonly applied techniques [8]. The performance of road surfaces can be improved by modifying bitumen [9]. One of the ways of maintaining the performance of asphalt pavements is by the use of polymers can significantly increase the resistance of asphalt mixture to permanent deformation, thermal fracture, fatigue cracking at low temperatures and they are also decease plastic flow and increase shear modulus at high temperatures [10]. Compatibility between polymer and bitumen should be high enough to avoid phase separation. Vinyl acetate-buacrylate is one of the polymers which is used to improve the properties of asphalt pavements. The present study is a part of a wider research on performances and durability of asphalt

²Production Department, Egyptian Petroleum Research Institute (EPRI), Cairo, Egypt

³Civil Engineer Department, Faculty of Engineering at Mataria, Helwan University, Cairo, Egypt

⁴Chemist, Arab Consulting Engineer office, Cairo, Egypt

mixtures made with RAP [11]. The research is divided into three stages: in the first stage, the bitumen extracted from RAP and the solid materials evaluated. The second stage, Vinyl acetate-bu-acrylate polymer added to the virgin bitumen for improving the bitumen characteristics [12]. Application of Marshall Stability forms the last stage of the experiment. Asphalt which conforms to Marshall properties are showing high resistance to stresses caused by high loads, high working temperatures and low temperatures due to weather conditions. HMA design using virgin asphalt as a control mix and hot mixes using the modified asphalt polymer to investigate Marshall characteristics [13]. The effects of modifying virgin asphalt of RAP with vinyl acetate-buacrylate polymer physical properties have been widely investigated. The objective of the research is to comprehensively characterize RAP mixtures in terms of performances of asphalt mixtures and bitumens by observing Marshall characteristics out of mix designs [14]. In this study Marshall stability and flow obtained and air voids, voids in mineral aggregate, unit weight and voids filled with bitumen calculated and shown in the curves to define the optimum asphalt content and the best percentage addition of polymer to the virgin asphalt [15]. HMA provides the best Marshall stability, due to better to proper mixing of asphalt binders and aggregates. This paper investigates how different ratios of vinyl acetate bu-acrylate polymer enhance the properties of RAP. Also, improve the mechanical and physical characteristics and hence improving the quality asphalt paving, increase asphalt-paving age and reduce the cost. The objectives of this research are set as follows:

- 1. Determined the effect of using 100% RAP instead of using virgin aggregates and asphalt.
- 2. Investigate the effect of thermoplastic elastomers polymer as asphalt modified.

3. Investigate the optimum modified asphalt content to improve the asphalt mix properties

2. EXPERIMENTAL

2.1. Materials

Vinyl acetate and butyl acrylate monomers, sodium lauryl ether sulfate (SLS), potassium per sulfate (KPS) and Sodium acetate were obtained from Sigma-Aldrich Company. Texapon P and nonyl phenol "NP30" were obtained from BASF. Cetyl alcohol was obtained from Dow Chemical Company. Ammonium hydroxide was produced by El-Nasr Pharmaceutical Chemical Company. The RAP used in this study, was obtained from a highway pavement in Cairo – Alexandria road, Egypt.

2.2 Methods and techniques

2.2.1. Preparation of vinyl acetate - butyl acrylate copolymer [16]

The polymerization was carried out in 500ml 3-necked flask fitted with a reflux condenser, thermometer and a mechanical stirrer. The temperatures of homogenization and polymerization were 25 & 80°C respectively, and nitrogen was purged during the polymerization step. Recipe for different ratios of vinyl acetate bu- acrylate copolymer are presented in **Table 1 and** the chemical structure of the prepared vinyl acetate butyl acrylate copolymer is represented in Scheme 1. During the process, the surfactant quantity was divided into two parts, namely, A & B with the ratio of 1:3 and the process included the following steps:

- 1. Part A containing vinyl acetate and butyl acrylate was emulsified in a little amount of de-ionized water, and homogenized for 15-20min at speed 3500 rpm in order to form pre-emulsion C.
- 2. 10% of C was seeded to the reactor, containing B; de-ionized water and pH regulator, during 15min with low speed mechanical agitator (80 rpm) and at 80°C. The allowed time for micelle formation

was an additional 15 min.

- **3.** Acrylic acid and acrylamide monomers were added to the remainder of part C (90%), under the homogenizer for 5-10 min.
- **4.** Afterward, this acidic emulsion was added to the reactor through dropping funnel in 150 min.
- **5.** In steps 2 & 4, a continuous dropping of the initiator solution was performed in the reactor.
- **6.** At the end of the addition of all ingredients, polymerization was allowed to continue for additional 2h then the reaction, the mixture was cooled to 50°C and subsequently neutralized with aqueous ammonium hydroxide to reach a pH value of 8.

Table 1: Recipe of vinyl acetate - butyl acrylate copolymer

Components	Wt.%
Vinyl acetate	25
Butyl acrylate	25
Sodium lauryl ether sulfate	0.4
Nonyl Phenol (NP ₃₀)	2.2
potassium per sulfate (KPS)	0.75
Sodium acetate (C ₂ H ₃ O ₂ Na)	0.6
Acrylamide (AA)	4
Dist. H ₂ O	42.05

2.2.2. Characteristics of vinyl acetate - butyl acrylate copolymer

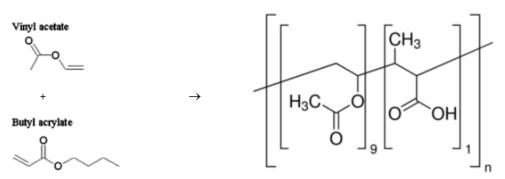
Fourier transform infrared (FTIR), Transmission electron microscopy (TEM), Thermo gravimetric analysis (TGA) and molecular weight (Mwt) were carried out in the international research center in Cairo, Egypt.

2.2.3. Preparation of the modified asphalt

The calculated amount of virgin asphalt was heated above its softening point. Surfactant was added at 10 wt. % of the asphalt to improve the durability. Vinyl acetate buacrylate copolymer was added slowly at loading levels 2, 4 and 6wt. %, at 160°C – 180°C and mixed under high-speed of 500 rpm for 2h. The end point was determined visually. Virgin and polymer modified asphalt (PMA) were characterized by conventional asphalt as penetration (ASTM D5–06), softening point (ASTM D 36–06), specific gravity (ASTM D70–09), kinematic viscosity (ASTM D2170–10).

2.2.4. Characteristics of the solid materials

The solid materials were obtained after the extraction of bitumen from RAP and tested for sieve analysis (ASTMC136-14), resistance to Angeles abrasion using Los machine (ASTMC131- 14) and bulk specific gravity (ASTMC128-15 &127-15 respectively). Tables 2-3 shown the sieve analysis and physical properties of the fine and the coarse aggregates, the obtained result was found to comply with the standards requirements.



Vinyl Acetate - Butyl Acrylate co - polymer

Scheme 1: Chemical structure of the prepared vinyl acetate butyl acrylate copolymer

2.2.5. Preparation of HMA samples

Asphalt paving mixes were prepared using the Marshall test method (ASTM D 6927–15 & AASHTO T245 – 97 (2008)) [17-18]. For each compacted sample of the asphalt paving mix, the stability and flow are measured while the unit weight and air voids are calculated to define the optimum asphalt content. All the mixes were designed according to the Egyptian Specification limits for dense graded hot mix asphalt (Dense –Graded 4D) as the following;

Mix (1): "control mix" it consists of virgin

Mix (2): it consists of PMA, using 2 wt. % vinyl acetate-bu-acrylate copolymer added to RAP.

Mix (3): it consists of PMA, using 4wt. % vinyl acetate-bu-acrylate copolymer added to RAP.

Mix (4): it consists of PMA, using 6wt. % vinyl acetate-bu-acrylate copolymer added to RAP.

Table 2: The average gradation for (RAP), five samples lies within the Limits of the Egyptian Standard Specification (Dense –Graded -D)

Sieve Size	Gradation before extraction % Passing	Gradation after extraction % Passing	Limits of the binder mix, Egyptian Standard Specification (Dense –Graded 4D) 2008
37.5 mm (1½")	100	100	
25.0 mm (1")	100	100	80 – 100
19.1 mm (¾")	84.1	88.5	70 – 90
12.5 mm (½")	76.4	80.3	
9.5 mm (3/8")	68.4	72.1	55 – 75
4.75mm #4	54.3	59.7	45 – 62
2.36mm #8	39.2	43.8	35 – 50
0.6mm #30	19.5	24.6	19 – 30
0.3 mm #50	11.4	14.5	13 – 23
0.15mm #100	2.4	8.2	7 – 15
0.075mm #200	0.45	3.3	0 – 8
Bitumen content %	*3.8	8%	3.5 – 7.0

^{*}The asphalt percentage obtained is 3.88 %.

Table 3: The physical properties of the coarse and the fine aggregates after extraction process

Droporty	AASHT	Coarse		Fine	Mi	AASHT
Property	О	Ag	Ag	aggregat	neral	О
Los angeles abrasion (loss	T 96 –	19.	18.			Max. 40
Bulk specific gravity G _{sb}	Т 85 –	2.5	2.4	2.65	2.7	
Apparent specific gravity	T 85 –	2.6	2.6			
Specific gravity SSD	Т 85 —	2.6	2.6			
Water absorption (Wt. %)	T 85 –	1.1	1.1			Max. 5
Stripping	T 85 –	>9	>9			>95 %

Where AASHTO, is the American Association of State Highway and Transportation Officials (AASHTO)

asphalt sample AC added to RAP.

3. RESULTS AND DISCUSSION

3.1. Characterization of the prepared emulsion copolymer

3.1.1. FTIR characterization

FTIR spectra shown in **Fig. 1** illustrates the following groups: the carbonyl group C=O double bonds appear in the region of 1733.2 cm⁻¹, the band at 2971 cm⁻¹ is specific of aliphatic hydrocarbon of butyl acrylate and vinyl acetate, where the band at 3321 is due to – OH group and – NH₂ of acrylamide.

3.1.2. Molecular weight (M.wt)

M.wt results for vinyl acetate - bu-acrylate

copolymer shown in **Fig. 2** Illustrates that, the relative time (RT) is 2.03 - 4.50, where M.wt average is 726 and mass over a charge number of ions is 50.00 - 805.37.

3.1.3. Thermo gravimetric analysis (TGA)

The thermo gravimetric analysis of the prepared copolymer is shown in **Fig. 3** and the data are represented in **Table 4**. It's clearly seen from Fig. 3 that, the weight of the sample continuously decreased as the temperature increased. The residue obtained in the degradation amount (% weight loss at maximum temperature) is 97.9 % where the decomposition temperature range is in between $(385-418 \, ^{\circ}\text{C})$.

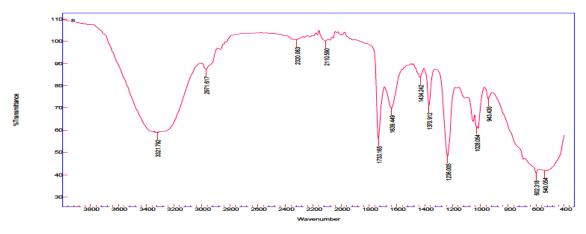


Figure 1: FTIR spectra of the prepared vinyl acetate – bu-acrylate copolymer

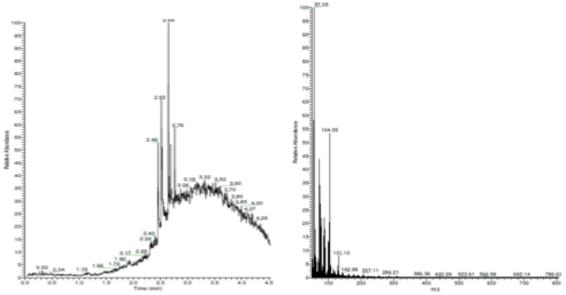


Figure 2: Mwt of vinyl acetate – bu- acrylate copolymer

3.1.4. Transmission electron microscopy (TEM)

Fig. 4 shows the TEM of the vinyl acetate – Bu – acrylate copolymer. It's clear from the figure that, the diameters of the observed particles of the polymer range between $0.5~\mu m$ and 200~nm. The implication here is that the progressive emulsion of the polymer improved the structure and robustness of the polymer. All particles are spherical and consist of a core from vinyl acetate – bu – acrylate copolymer. It's clear from the image that the particles are spherical in shape without any deformation with narrow distribution.

3.2. Modification of the asphalt

Table 5 illustrates the physical characteristics of virgin asphalt and the modified asphalt with 2,4 and 6 wt. % vinyl acetate – bu- acrylate copolymer (2, 4 and

6%). The obtained results showed that, the modification of asphalt with vinyl acetate-bu- acrylate copolymer produces a binder more hardener than the virgin sample, as it has lower penetration and higher kinematic viscosity and softening point. This may be attributed to this type of polymer produces a fine dispersion of the polymer in molten (solvating) phase with no disturbance of the bitumen structure, as it is a thermoplastic and a flexible polyolefin which does not contain any double bonds. Generally, the polymer creates a network to the asphalt molecule.

The obtained data also showed that, the penetration, softening point, kinematic viscosity of the modified asphalt improved by incorporation of different wt. % of the prepared emulsion copolymer, as the

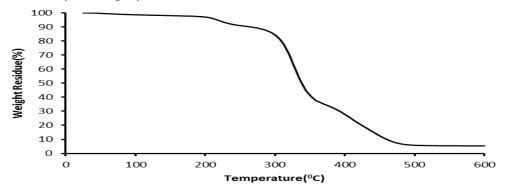


Figure 3: TGA of the prepared vinyl acetate – bu- acrylate copolymer

Table 4: TGA data of Vinyl Acetate – Bu- Acrylate co- polymer

% weig	ght loss at	various ter	mperature (⁰ C)	Decomposition temp.				
200	300	400	Max °C	range (°C)	T_{15}	T_{50}	T_{90}	T_{max}
5.67	23.6	72.7	93.7	300 – 460	247.8	331.6	463.5	473

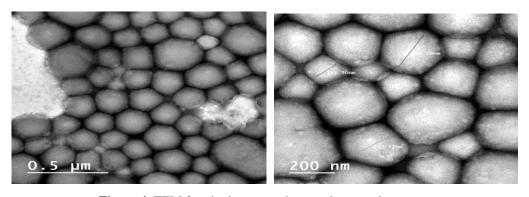


Figure 4: TEM for vinyl acetate – bu- acrylate copolymer

obtained results are within the specified criteria, where the specific gravity not has specific requirements.

3.3. Evaluation of the prepared HMAs

In accordance with the Egyptian standard, the optimum asphalt content of the AC mixture was determined using the design methodology. Marshall mix Traditional Marshall compaction tests of the HMA were carried out in the laboratory at 150 °C with 75 blows on each side of cylindrical samples. The results show that, the stability increases with the increasing asphalt content up to the optimum asphalt content and thereafter decreases. The increase in stability can be attributed to the improved adhesion between the aggregate and bitumen. It can be seen from the results that the flow decreases with increasing copolymer content. The slight decrease in flow value, may be due to the quantity of the copolymer used in the mixture. However, all mixtures met the Egyptian Specification for Road. Air void content, stability and other characteristics are illustrated in **Table 6** and **Figs. 5** - **12**, and comparing to the control mix (1) the following results are detected,(a) the optimum asphalt content increased compared to control mix (0.14 % for 2wt.% modified copolymer, 0.11 for 4wt.% modified copolymer and 0.077 for 6% modified copolymer); (b) the stability of the control mix increased from 1050 Kg to 1380 Kg for the modified mix with 4wt.%

Table 5: Characteristics of the used asphalt

Physical Characteristics	Virgin	Emulsion content		ESP	
	AC	(2%)	(4%)	(6%)	
Penetration (at 25°C, 100g, 5s) 0.1mm	63	63	64	66	60/70
Softening point (ring & ball) ° C	46.5	49	50	52	45/55
Specific gravity(at 25/25) °C using a pycnometer	1.02	1.044	1.092	1.111	Not specified
Kinematic viscosity (at 135°C) cSt	380	480	560	730	> 320

Table 6: Marshall characteristics of the virgin asphalt and modified asphalt mixes

	Control Mix (1)	Em	ulsion con	Egyptian standard	
Characteristics		2wt.%	4wt. %	6wt.%	Specification limits
	WIIX (1)	Mix (2)	Mix (3)	Mix (4)	For binder course
Optimum asphalt content before extracti	on (%)		3.88		
Optimum asphalt content (%)	5.503	5.65	5.62	5.58	Not specified
Stability of the mix (kg)	1050	1260	1380	1320	Min 700 Kg
Unit weight of the mix (gm/cm ³)	2.308	2.315	2.313	2.311	Not specified
Flow of the mix (0.01 inch)	12.2	12.0	11.8	11.5	8 – 16
Air voids of the mix (%) VA	4.8	4.6	4.4	4.3	3 – 8
Voids in mineral of (%) VMA	15.25	15.0	15.05	15.10	Min 15 %
Voids filled with asphalt (%) VFA	68.9	71.2	71.4	71.5	60 - 70
Marshall stiffness Kg/in	86.06	104.2	116.94	114.78	Not specified
Modified asphalt added to RAP (%)	1.62	1.77	1.74	1.7	Not specified
Polymer modified asphalt %	0.0	0.035	0.07	0.102	Not specified

copolymer; (c) Marshall stiffness of the prepared mixes increased from 86.06 for control mix to 104.2, 116.94 and 114.78 for copolymer modified mixes, this is due to the increase of stability and decrease in flow values of asphalt mix using different percentages of copolymer; (d) The air voids slightly decreased from 4.8 % to 4.3 % and (e) The indirect tensile strength increased from 9.2 Kg/cm² for the control mix to 9.5 Kg/cm² for 4wt.% copolymer.

The optimum asphalt content approximately have a very slight change it was between 5.503 and 5.58 %. The slight decreases in the flow value for the modified mixes, may be due to the increasing in the stability values. The air voids % of the control mix was 4.8 which decreased slightly to 4.6, 4.4 and 4.3 of the mixes contain 2, 4 and 6 wt.% copolymer, respectively. The air voids % in solid materials was also decreased slightly from 15.25 for the control mix to 15.10 and 15.05 for mixes contains 2 and 4 wt. copolymer, and increased to 15.10 for the contains 6 wt.% copolymer. The percent voids filled with bitumen increased for all the modified mixes comparing to control mix, it increased to 71.2, 71.4 and 71.5 for the modified mixes contain 2, 4 and 6 wt.% copolymer respectively, compared to 68.9 for the blank mix. The unit weight of the mix increased to be 2.315 g / cm³ for the mix contains 2 wt. % copolymer and slight decreased for to 2.313 and 2.311 for the mixes contain 4 and 6 wt.% copolymer.

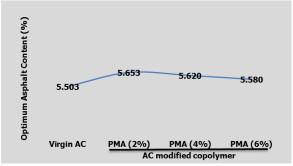


Figure 5: Optimum asphalt content for the mix designs

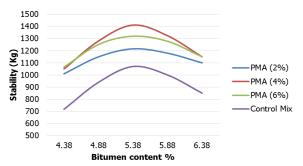


Figure 6: The stability graph for the mix designs

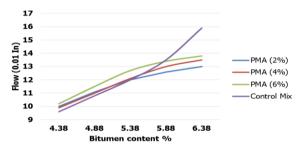


Figure 7: The flow graph for the mix designs

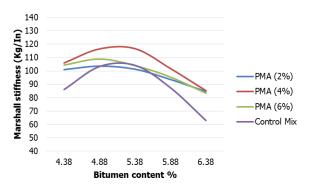


Figure 8: Marshall stiffness graph for the mix designs

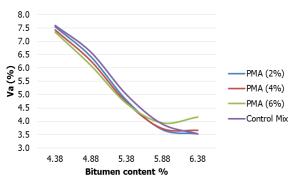


Figure 9: The air voids graph for the mix designs

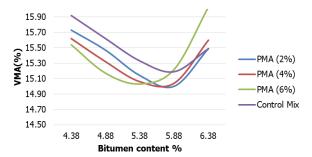


Figure 10: The voids in the mineral aggregate graph for the mix designs

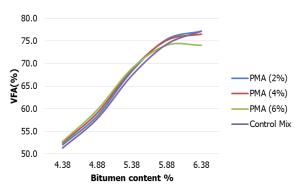


Figure 11: The voids filled with bitumen graph for the mix designs

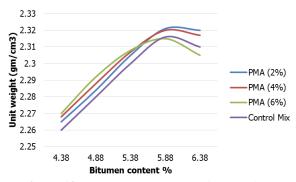


Figure 12: The unit weight graph for the mix designs

4. CONCLUSION

The possibility of reducing the cost of asphalt modified with copolymer without impairing the quality of the binder was proved. Asphalt composition modified by copolymer was found to comply with the requirements Egyptian of Standard Specification and has characteristics that are typical for using special copolymer modifier vinyl acetate - butyl acrylate copolymer. This study focuses evaluating the effects of polymer

modifying reclaimed asphalt pavement in order to improve the quality of paving and reduce the cost of asphalt. In this research, vinyl acetate – butyl acrylate copolymer was used as an asphalt modifier, it was mixed with virgin asphalt in three different loading levels 2, 4 and 6 wt. %. The best result was obtained at 4 wt.% loading level. The application of the Marshall design of the hot mix asphalt for control mix and the modified mixes showed that:

- The optimum asphalt content was decreased for mixes 2, 3 and 4 from 5.503 to 5.65, 5.62 and 5.58. This may due to the effect of emulsion copolymer, as a flexible copolymer causing the adhesive properties of asphalt to increase.
- The stability increased for mixes 2 and 3 from 1260 to 1380 Kg, then decreased for mix 4 to 1320 Kg compared to 1050 Kg for the control mix 1. This increase in stability for mixes 2 and 3, is due to the compatibility between emulsion and asphalt, while for mix 4, the decrease is due to the separation of polymer and asphalt.
- The percent of air voids in the mix for mixes 2, 3 and 4 decreased from 4.8 to 4.6, 4.4 and 4.3 % of 4.2, 8.3 and 10.4 respectively and this is due to the increase in stability of mixes in addition to the compatibility and flexibility of polymer samples.
- The percent air voids in solid materials decreased for mix 2 from 15.25 to 15.0 and increased for mix 3 to 15.5 and mix 4 to 15.1 respectively. All these results may due to the effect of compatibility of asphalt and the polymer which in turn affects an the adhesion power between the solid materials and the asphalt.
- Marshall stiffness of the prepared mixes increased from 86.06 for control mix1 to 104.2, 116.94 and 114.78 for

- mixes no. 2, 3 and 4 in percentages of 21.0, 35.8 and 33.4 respectively. This is due to the increase of stability and decrease in flow values of asphalt mix using PMAs.
- The addition of 2 wt. % vinyl acetate butyl acrylate copolymer produced HMA, has good Marshall stability as well as it gives preferable general characteristics of the modified asphalt.

5. COST INVESTIGATION

Cost is an important factor in terms of recyclability and reuse of material and can be an incentive to use such material. Aggregates and binders from old asphalt pavements are still valuable even though the damaged pavements have reached the end of their service lives. As shown in the **Table 7**, to calculate reuse of 1 cubic meter of RAP it needs 37.5 kg of virgin asphalt

and 230 ml of surfactant where using 4% modified asphalt, it needs 40.25 kg of virgin asphalt 1.61 Kg of vinyl acetate butyl acrylate and 230 ml of surfactant. **Table 7** also shown the total cost for using 1 m³, which is enough for paving a section with length 15.4 m, width 3m and thickness of 0.05 m. It was observed that, the cost when using 4% modified polymer is 32.45 of the total cost when using virgin HMA which make cost reduction of 67.55% of the total cost. The cost for producing 1 m³ virgin HMA is 1000 Egy pounds, where the cost for reuse RAP modified with the 4% polymer is 281.72 Egy pounds. According to test results and investigation studies of the using RAP with different percentages of modified copolymer, it is recommended to reuse RAP with 4% modified copolymer in order to save not less than 67% of the total cost.

Table 7: The cost reduction by using reclaimed asphalr pavements modified with the prepared copolymer

Characteristics	Control	Modifier content			
Characteristics	Mix (1)	2 wt. %	4 wt. %	6 wt. %	
		Mix (2)	Mix (3)	Mix (4)	
Optimum asphalt content (%)	5.503	5.473	5.45	5.44	
Optimum asphalt content extraction (%)		3.8	38		
Modified asphalt add to RAP (%)	1.62	1.77	1.74	1.70	
Polymer modified asphalt (%)	0.00	0.035	0.070	0.102	
Surfactant (%)		10	.0		
Required polymer, surfactant and aspl	asphalt calculation for reusing RAP				
Volume m ³ (1)	1	1	1	1	
Unit weight of the mix (gm/cm ³)(2)	2.308	2.315	2.313	2.311	
Ton(1)/(2)= (3)	2.308	2.315	2.313	2.311	
Modified asphalt add to RAP (Kg)	37.5	41.0	40.25	39.3	
Vinyl acetate – butyl acrylate (kg)	0.0	0.82	1.61	2.36	
Surfactant NP ₉ (L)	0.23	0.23	0.23	0.23	
Price of vinyl acetate – butyl acrylate (Kg) /Egy. P		2	4		
Price of surfactant (L) /Egy. Pound		1	8		
Price of virgin asphalt (Kg) /Egy. Pound		7	7		
Cost of va-buA for the mixture	0.0	19.67	38.64	56.57	
Cost of surfactant for the mixture	4.16	4.16	4.16	4.16	
Cost of virgin asphalt for the mixture	262.21	286.83	281.72	275.01	
Total cost for using RAP for 1 m³ per Egy Pound	266.37	310.66	324.52	335.74	
The cost for using HMA for 1 m³ per Egy Pound	1000				
Total percentage cost of RAP	26.64 %	31.07 %	32.45 %	33.57 %	
The saving percentage of using RAP	73.36 %	68.93 %	67.55 %	66.43 %	

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