

## **Effect of Mineral Filler and Additives Materials on the Adhesion Properties between Asphalt and Aggregate**

تأثير المواد المألثة والمضافات على خصائص التلاصق بين الاسفلت والركام

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### **Abstract:**

Numerous attempts to study adhesion properties between the coarse aggregate and asphalt and then study effect of additives materials and filler aggregate (fine) by using three types of pure asphalts (according to penetration grade 40/50, 60/70 and 85/100), which obtained from Durah refinery. Low density Polyethylene as additive material in three percent (0%, 2% and 4%) is used too, and limestone filler in two percent (0 % and 4%) are made. The surface tension stress theory between asphalt, aggregate and water is applied at assuming the aggregate which used are hydrophilic type (explain this type as glass plane plate).

From the results are obtained, the asphalt viscosity improved by using additives materials (increasing surface tension force between asphalt and aggregate (adhesion forces) greater than using mineral filler materials, then increase ability of mixture asphalt to resist stripping.

**Keyword: Adhesion Properties, Aggregate, Additives Materials, Filler Materials.**

### **الخلاصة :**

بذلت محاولات عديدة لدراسة خصائص قوى التلاصق بين حبيبات الركام الخشن والاسفلت ومن ثم دراسة تأثير المواد المضافة و الركام الناعم ( المواد المألثة) وذلك باستخدام الاسفلت من مصفى الدورة وبتلات تدريجات هي ( ٥٠/٤٠ و ٧٠/٦٠ و ١٠٠/٨٥ ) واستخدام البولولي اثلين الواطئ الكثافة كمادة مضافة وبتلات نسب هي ( ٠% و ٢% و ٤% ) واستخدام النورة المطفنة كمادة مألثة وبنسبتين هما ( ٠% و ٤% ) وتطبيق نظرية اجهادات الشد السطحي بين الاسفلت والركام والماء وعلى فرض امكانية تمثيل الركام بقطعة زجاجية مستوية. من خلال النتائج التي توصل اليها البحث نلاحظ بان لزوجة الاسفلت تحسنت باستخدام المضافات وبزيادة نسبة المادة المضافة وبالتالي زيادة قوة الشد السطحي بين الاسفلت والركام ( زيادة قوى التلاصق بينهما) بصورة اكثر من استخدام المواد المألثة وبالتالي زيادة قابلية الخلطة الاسفلتية لمقاومة الانفصال و التفكك بين مكوناتها.

### **Introduction :**

The durability of asphalt paving mixtures is mainly affected by securing and maintaining the adhesion between the asphalt and the aggregates in the presences of water. The loss of adhesion in the mixture (stripping) induces instability and promotes failure conditions in the asphalts pavement. This condition can be observed in asphalt layers using hydrophilic aggregates. *Petersen et. al.*(1994); in some locations only hydrophilic aggregates are available. A certain modification to ensure a durable adhesion must be made. Such modifications are usually made in two ways:

- 1) Modification of the adhesion properties of the asphalt by tensioactive adhesions.
- 2) Modification of the surface properties of the aggregates by a treatment with a cement-water solution or a hydrated lime-water solution.

Of the two, the first modification is more useful in asphalt paving technology. However, high quality additives are quite expensive for the mass production of asphalt mixtures. A solution to this problem can be obtained by considering the influence of natural mixture ingredients, such as filler, on the adhesion between the aggregate and the asphalt in the presence of water.

This paper describes a mechanism for creating adhesion between the aggregate and the asphalt , using certain types of fillers. Quantitative methods are used for evaluating the adhesion and for investigating the influence of the filler

### **Adhesion Phenomenon in Aggregate – Asphalt Systems.**

The adhesion in the interface between aggregate and asphalt can be defined as the property of the asphalt to adhere to the aggregate surface, and to maintain this condition in the presence of water. The adhesion phenomenon and water effects are complex. Many theories have been expressed regarding the water –resistance of asphalt –coated aggregates.

**Rice** (1977); classifies the theory of adhesion into three concepts.

#### **1) The Chemical Reaction Concept.**

When aggregates are wetted by the asphalt, selective adsorption occurs the interface, followed by a chemical reaction between the adsorbed material and the constituents of the solid phase. Under this condition the acidic components of the bituminous material chemical react with the basic aggregate mineral to form water-insoluble compounds.

#### **2) The Mechanical Concept.**

The surface texture of the aggregate surface is the main factor, which affects mechanical adhesion factors such as the size of individual crystal faces, aggregate porosity, adsorption, and surface coating.

#### **3) The Surface Energy Concept.**

**Petersen** (1994) ; adhesion results from the interfacial energy relationship at aggregate-bitumen water-air-interface. In This regard, the mechanisms of spreading, wetting, and stripping may be mentioned. Generally when a liquid and a solid are brought to gather, the liquid may

- Neither spread on nor wet the solid surface.
- Spread on the surface without wetting,
- Spread on and wet the surface

Among these three theories, the interfacial energy concept is the most widely accepted. It provides a physical basis for a quantitative expression and evaluation of the adhesion and the effect of water. This expression can be obtained by the equilibrium state of the interfacial forces at the mutual contact point of the aggregate, asphalt and water.

Figure (1) describes the Interfacial forces acting at the mutual contact point (M) of a asphalt drop touching an aggregate surface in the presence of water. The equilibrium conditions of M are expressed as follows:

$$\sigma_{wa} = \sigma_{ab} + \sigma_{bw} * \cos \theta \dots\dots\dots(1)$$

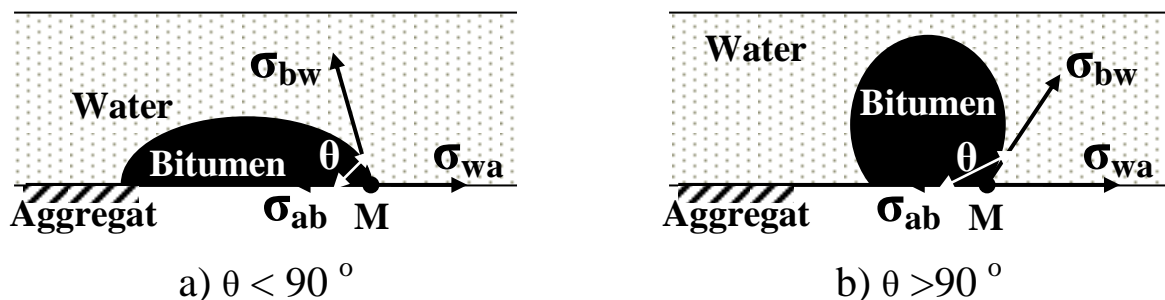


Figure (1): Interfacial Forces acting at the Mutual Contact Point of a Bitumen Drop. **Ishai**, 1977  
The adhesion potential can be expressed, using the angle of adhesion ( $\theta$ ) as follows:

$$\cos \theta = \frac{\sigma_{wa} - \sigma_{ab}}{\sigma_{bw}} \dots\dots\dots(2)$$

where:

- $\sigma_{wa}$  = Interfacial tension between water and aggregate.
- $\sigma_{ab}$  = Interfacial tension between aggregate and binder.
- $\sigma_{bw}$  = Interfacial tension between binder and water.
- $\theta$  = Angle of adhesion.

When

$\cos \theta > 0$  (It is the spreading stage as shown in Figure (1-a).

$\cos \theta < 0$  (It is the stripping stage as shown in Figure (1-b)).

$\cos \theta = 0$  (It is the border line between spreading and stripping).

### Practical Test Method for Evaluating Adhesion

Current practical test used to evaluate the effect of water in adhesion of aggregate bitumen systems can be divided into,

- 1) Visual inspection of coating conditions of the coated aggregate after a period of water immersion. (ASTM D1664 and ASTM D2727).
- 2) Measurement of the interfacial forces at the aggregate-bitumen-water interface. The measurements, which can be applied to each force component or to the combined resulting phenomenon, are directing quantitative, and expression of the basic physical essence of the phenomenon. Despite these advantages, these methods of evaluation are not so commonly used in practice since they require accurate measurements and controlled conditions.
- 3) Measurement of the strength of the bituminous mixture properties before and after water exposure. (ASTM D1075). These methods can be applied to any strength test by providing additional measurement after cold or hot water immersion.

It is not within the goal of this paper to recommend the best testing method. However, *Pertersen* (1994) explains the second and third methods were found to be the most suitable methods for evaluating the effect of the filler on the adhesion properties of asphalt-aggregate systems.

### Experimental Program

*Ishai et. al.* (1977) ; two methods of testing were used to evaluate the effect of the filler on the adhesion properties of asphalt-aggregate systems in the presence of water; they are

- 1) Measurement of the effect of interfacial forces at the aggregate –asphalt-water interface;
- 2) Measurement of the effect of variable environmental conditions on the mechanical behavior of sand asphalt mixtures.

### Measurement of the Effect of Interfacial Forces

The testing program was aimed at evaluating the effect of filler type and tension-active additives on the adhesion potential of asphalt-aggregate systems. The adhesion potential was expressed by the angle of adhesion ( $\theta$ ) as defined by equation (2).

Hot bitumen was dropped on smooth surface plates (glass plate). The plates were immersed in distilled water at a constant temperature (38 C° and 60 C°) for 10-min time. During the immersion the flat bitumen drop progressively changed its shape into a ball (the degree which it did so depending on the interface adhesion properties. The geometry of the drop was projected on screen and the contact angle ( $\theta$ ) measured. The measurement technique of ( $\theta$ ) is given in Figure (2).

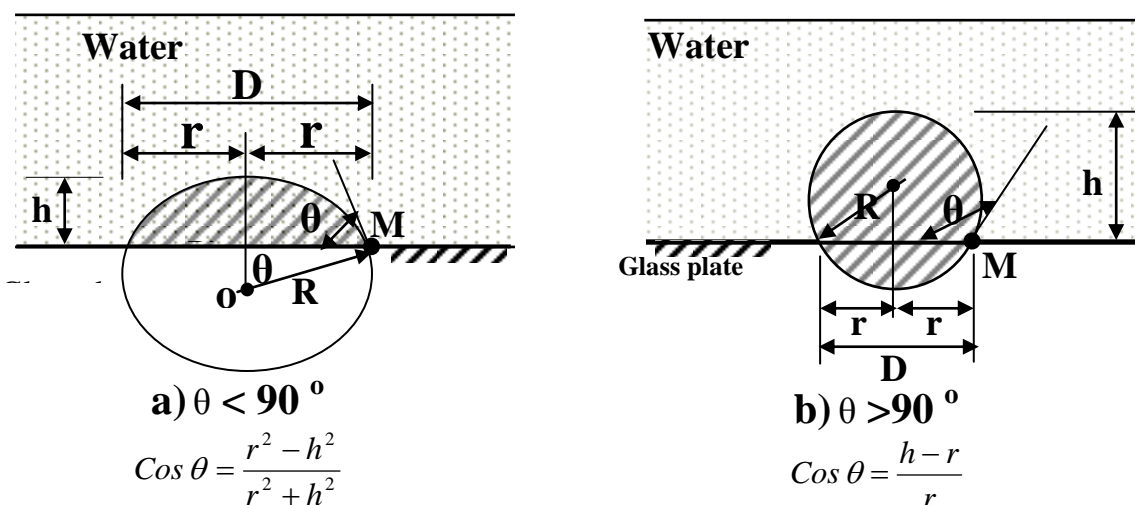


Figure (2): Measuring Technique of  $\cos \theta$ .

The testing equipments are described in plate (!) .The projected asphalt drop during shape variation is described in plate (2) and the measurement technique of is given in plate (3).

Testing conditions were as follow;

**1) Aggregate:**

The ideal surface and the most critical conditions for hydrophilic aggregates were simulated by using glass plate slides.

**2) Asphalts:**

Three types of asphalt cement were obtained from Daurah Refinery including (40-50), (60-70) and (85-100) penetration graded asphalt. The physical properties are shown in Table (1).

The consistency of asphalt cement changes with temperature and many methods can be used to measure temperature susceptibility, including:

**a) Penetration Ratio**

Is the percentage between penetration,  $P_1$ , at (46 °C, 50 gm, 5 sec) to the standard penetration,  $P_2$ , at (25 °C, 100 gm, 5 sec). *Alani* (1987).

$$P_R = \frac{P_1}{P_2} * 100 \dots\dots\dots (3)$$

The results are shown in Table (2). The lower values of penetration ratio are indicating greater temperature susceptibility.

**Table (1): Physical Properties of Asphalt Cement.**

Asphalt Property	Test Conditions	Units	ASTM, AASHTO Designation	Asphalt Cement Grade		
				40-50	60-70	85-100
Penetration	25 °C , 100 gm ,5 sec	0.1 mm	D-5 T-49	47	69	90
	46.1 °C , 50 gm ,5 sec	0.1 mm	D-5 T-49	57.7	111.5	167
Ring and Ball Temp. (Softening Point)	Rate 5 °C per 1min	°C	D-36 T-53	49	47	46
Ductility	25 °C , 5 cm / min	cm	D-113 T-51	+120	+120	+120
Flash Point (Cleveland open cup)	-----	°C	D-92 T-48	270	285	290
Specific Gravity	25 °C	-----	D-70	1.03	1.04	1.01
Kinematic Viscosity	135 °C	Cst	D-2170	490	511.37	----
	100 °C	Cst	D-2170	3860	5019.98	----
Penetration after Thin-Film Oven Test	25 °C , 100 gm , 5 sec	0.1 mm	D-5 T-179	32	40	44
Solubility	Trichloroethylen e Disulfide	% wt	T-44	99.5 %	99 %	99 %
Loss in Heating	163 °C , 50 gm ,5 hr	% wt	D-1754	0.18	0.16	0.02

**Table (2): Penetration Ratio.**

Asphalt Grade	Penetration Ratio, P <sub>R</sub> %			
	Pure Asphalt	Effect of adding LDP		Effect of adding 4% Filler
		2 %	4%	
40-50	1.23	1.3	0.77	1.60
60-70	1.61	1.29	0.75	2.12
85-100	1.86	1.16	0.64	2.47

**b) Penetration Index (PI)**

It can be calculated from the following equations;

$$PI = \frac{20 - 500A}{1 + 50A} \dots\dots\dots(4)$$

$$A = \frac{\log 800 - \log (\text{Pen. at } T_{\text{measured}})}{T_{R\&B} - T_{\text{measured}}} \dots\dots\dots(5)$$

Where:

T<sub>measured</sub> : temperature in °F , at which the penetration is made.

T<sub>R&B</sub> : softening point temp. °F.

The results are shown in Table (3).

**Table (3): Penetration Index.**

Asphalt Grade	PI
40-50	+ 2.37
60-70	+ 2.80
85-100	+ 3.30

*Alani H. M.*, (1993), concluded that the Iraqi asphalt values of penetration index are ranged between value 0 to –2. Higher values of PI indicate lower temperature susceptibility.

**3) Mineral Fillers;**

One type of mineral filler is used in this work. This is the limestone dust from Kerbala factory. The physical properties are shown in Table (4).

**Table (4): Physical Properties of Mineral Filler.**

Property	Limestone Dust
Percent passing sieve No. 200	97 %
Specific gravity	2.96
Specific surface (m <sup>2</sup> /kg)	156

In order to study the effect of percent of filler on the behavior of the HMA, two percents (0 % and 4 %) has been selected.

**4) Additives Materials;**

In this study low density polyethylene (LDP) has been used as an asphalt modifier to control fatigue cracking failure .The physical properties are shown in Table (5).

**Table (5): Physical Properties of Low Density Polyethylene. Alani H. M., (1993).**

Property	Value	Unit
Density	920	Kg / m <sup>3</sup>
Tensile Strength	10	MN / m <sup>2</sup>
Flexure Modulus	0.2	GN / m <sup>2</sup>
Chemical Unit	(-CH <sub>2</sub> –CH <sub>2</sub> ) <sub>n</sub>	----
Thermal Degradation Temperature	404	°C

The effects of low-density polyethylene on the properties of asphalt binder are shown in Table (6).

**Table (6): Effect of Low-Density Polyethylene on the Penetration.**

	Percent of Low Density Polyethylene					
	2 %			4 %		
Asphalt Grade	40-50	60-70	85-100	40-50	60-70	85-100
Penetration (0.1 mm)						
at 25 °C ,100 gm,5 sec.	66	99	164	56	86	141
at 46 °C,50 gm, 5 sec.	78	128	191	43	64	90

The kinematic viscosity, Centistokes (Cst), for (60-70) asphalt with 4 % low-density polyethylene is equal to 5946.8 Cst at test temperature equal to 100 °C and 613.8 Cst at 135 °C respectively (this test making at Al-Daurah Refinery 3764 , 17/ March/2003).

### **Testing Results and Discussion for Interfacial Effect Study**

Generally, adhesion in aggregate-asphalt interface is influenced by the viscosity of the binder. This is a mechanical phenomenon. On the one hand, higher viscosity can reduce the coat ability and wetting of the asphalt in coating phase. On the other hand, when good initial coating and wetting are achieved, the resistance to stripping is increased with increasing viscosity of the binder.

The introduction of the filler into the binder to form asphalt – filler usually increases the viscosity of the original asphalt binder. The influence of the filler on the adhesion, therefore, is similar to the influence of increasing the viscosity of the original asphalt.

A summary of the test results, as expressed by the angle of adhesion ( $\theta$ ), is given in Table (7). As can be seen from this table, positive values of  $\cos \theta$  characterize the adhesion potential between the pure asphalt and the glass, in the presence of water. This indicates a good level of stripping.

A slight improvement in adhesion is achieved by adding limestone filler to the asphalt. The positive values of  $\cos \theta \cong 0.5$  indicate a weak adhesion potential. On the other hand, the positive values of  $\cos \theta \cong 0.8$  indicate a good adhesion potential. Intermediate values of  $\cos \theta \cong 0.7$  indicate a weak adhesion potential. Figure (3); shows effect of different parameters (test temperature, percent of additives and percent of mineral filler)

The values of  $\cos \theta$  (adhesion potential) increase with adding LDP.

### **Conclusions**

The following are the summary of the results, based on the theoretical study and experimental test:

- <sup>1</sup> It can be seen that all results of adhesion potential( $\cos \theta$ ) , are obtained from laboratory test are positive value ( $\cos \theta > 0$ ) or the values of adhesion angle ( $\theta$ ) less than 90 ° , that mean asphalt binder at spreading stage and this asphalt improved mixture and given good resist to stripping.
- 2.The additives and filler materials improved viscosity of the asphalt binder, and then increased ability of asphalt to coat with aggregate and increased ability of mixture asphalt to resist stripping and disintegration.
3. The higher temperature reduced the asphalt binder viscosity, then adhesion potential ( $\cos \theta$ ) properties and decreases ability of mixture to resist stripping.
- 4.The LDP additives materials increased asphalt viscosity, so the increased LDP content increased ability of asphalt binder to adhere with aggregates and then increasing mixture resistance to stripping.
- 5.The LDP additives change physical and mechanical properties and reduced penetration value (increasing ability of asphalt to resist hardening or consistency).
6. The asphalt viscosity increased with increasing percent of LDP

**Table (7): Average Values of Adhesion Potential (Cos  $\theta$ ) for asphalt –filler –Water Combinations.**

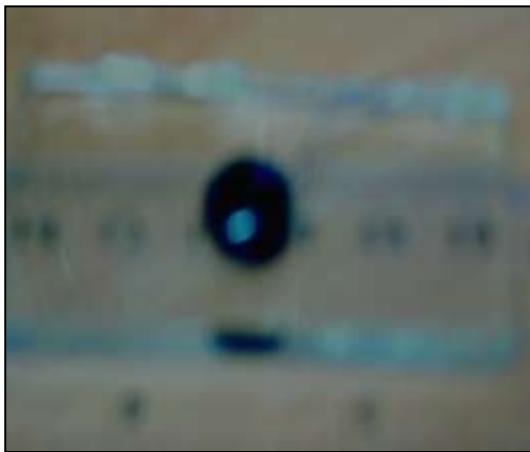
Test Temperature		38 C°			60 C°		
A) Original asphalt without additive and filler.							
	Asphalt type	40-50	60-70	85-100	40-50	60-70	85-100
	D mm	15	11.67	14.5	17	15	17
	H mm	2.5	3	3	1.8	2	2
	R=D/2 mm	7.5	5.84	7.25	8.5	7.5	8.5
	Cos θ	0.8	0.582	0.707	0.914	0.865	0.895
	θ°	36.87°	54.42°	44.96°	23.913°	29.86°	26.5°
	Contact area mm <sup>2</sup>	176.71	106.96	165.1	226.92	176.71	226.96
B) With Additives 2% LDP							
	D mm	20.00	16.25	17.2	23	22.67	23.00
	H mm	4.00	2.50	3.00	2.00	1.50	2.00
	R=D/2 mm	10.00	8.125	8.625	11.5	11.34	11.5
	Cos θ	0.824	0.827	0.784	0.94	0.96	0.96
	θ°	43.6	34.2	38.4	19.7	43.9	19.8
	Contact area mm <sup>2</sup>	314.16	207.4	233.7	415.45	406.6	415.5
	H/D ratio	20	15.4	17.6	17.4	13.2	17.4
C) With Additives 4% LDP							
	D mm	13.5	11.5	24	18.17	22.83	30.3
	H mm	1.5	3.5	3	1.125	1.86	0.8
	R mm	6.75	5.75	12	9.1	11.4	15.2
	Cos θ	0.906	0.895	0.88	0.96	0.94	0.99
	θ°	25	62.6	28.07	14.09	18.5	6.1
	Contact area mm <sup>2</sup>	143.1	153.9	452.4	259.3	409.36	722.5
	H/D ratio %	11.1	30	12.5	6.2	8.5	7.64
D) With add 4% of the filler.							
	D mm	12.25	11.75	11.75	12.00	12.50	12.50
	H mm	2.50	2.00	2.10	2.00	1.80	2.00
	R mm	6.125	5.87	5.90	6.00	6.25	6.25
	Cos θ	0.71	0.79	0.77	0.80	0.846	0.81
	θ°	44.00	37.6	39.40	36.90	32.10	35.50
	Contact area mm <sup>2</sup>	117.86	108.4	108.4	113.10	122.70	122.70
	H/D ratio %	20.40	17.00	17.90	16.70	14.40	16.00
Note:- Values in table are average of 3 drop samples.							



**Plate (1):-Testing Equipments.**



**Plate (2):-Asphalt drops.**

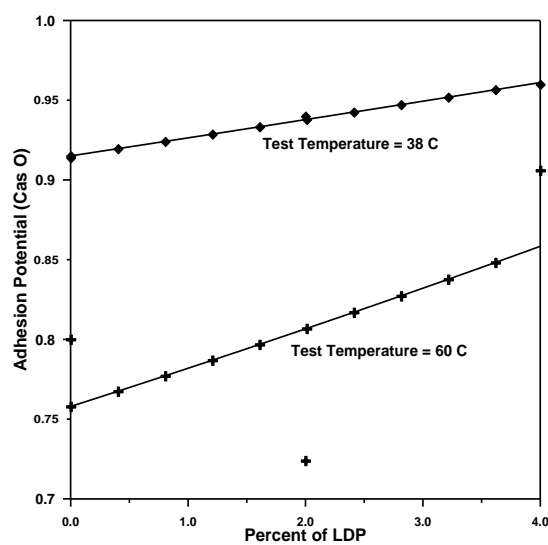


**a) Measured Diameter (mm).**



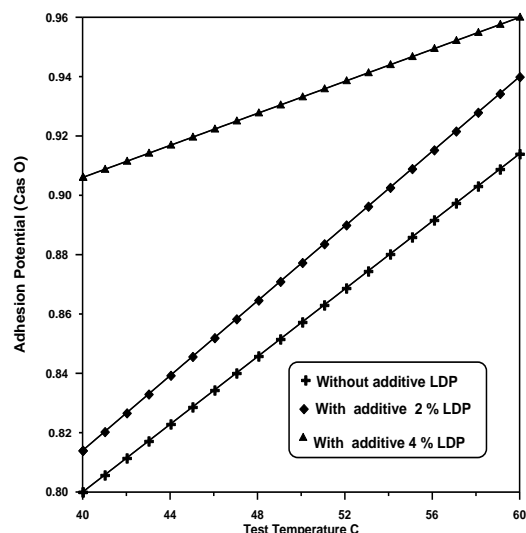
**b) measured height (mm)**

**Plate (4):-Measurement Technique.**

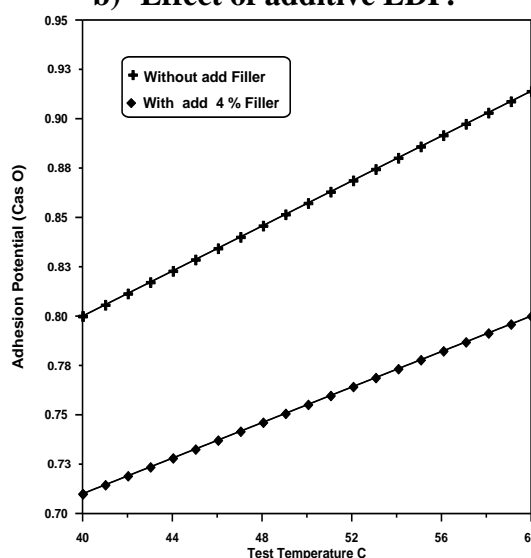


**a) Effect of Test Temperature.**





**b) Effect of additive LDP.**



**c) Effects of add Filler.**

**Figure (3): Effect Parameter on the Adhesion Angle.**

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