

EFFECT OF pH OF WATER ON MOISTURE SUCEPTIBILITY OF ASPHALTIC MIXTURES

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الخلاصة

ان وجود الماء بين الرابط الاسفلتي و سطح الركام يسبب مشاكل انشائية لانه يفصل بين الرابط والركام مما يفقد استقرار المزيج الاسفلتي. وجود الماء ضمن هذه المنطقة يغير ون قيمة pH حيث تصل الى 10 وهي قيمة عالية بما يكفي لكسر الربط بين الركام والاسفلت. يمكن للماء ان يدخل الى هذه المنطقة من مصادر خارجية مثلا وجود فشل في انظمة تصريف المياه او مجاري المياه الثقيلة الطافحة او المكسورة او ارتفاع منسوب الماء الارضي. اضافة لما تقدم فيوجد مصدر داخلي للماء وهو الماء الموجود ضمن مسامات الركام. في هذا البحث تم تصنيف الركام العراقي بناءا على قابليته لمقاومة التأثير التخريري للماء وحسب التكوين الحبيبي له والشكل الخارجي. اشارت النتائج الى ان الركام المكسر اظهر مقاومة اقل لتاثير الماء من الركام غير المكسر كما ان المزيج الاسفلتي الذي لم يستخدم له اي مضافات لتحسين مقاومة تاثير الماء فشلت باختبار الماء المغلي حيث كانت نسبة الركام الذي حافظ على ارتباطه بالرابط الاسفلتي بوجود الماء (pH تقريبا 10) لا يتعدى 45%. تم استخدام الجير كمضاف لتحسين مقاومة الماء وتم اضافته بثلاثة طرق: الطريقة الجافة , الركام الرطب و slurry lime . الطريقة الثالثة اظهرت نتائج افضل من الطريقتين الاوليتين حيث بلغت نسبة الرابط المحيطة بالركام في فحص الماء المغلي 85% للركام غير المكسر و 78% للركام المكسر. الكوارتزيت يولف 80% من الركام المفحوص و الذي يعكس خصائص حامضية لسطح الركام حيث وبوجود الماء ترتفع قيمة pH الى 11. كخلاصة لما تقدم فان وجود الماء يسبب مشاكل للمزيج الاسفلتي تتطلب استخدام معالجات كطريقة slurry lime.

ABSTRACT

Moisture damage of asphaltic mixture is a term used to refer to the distress due to the effect of water presence between aggregate surface and asphalt binder. Water at this interlayer alter the pH to high value reaching 10 leads to splitting or breaking bonds between aggregate and asphalt which is known as stripping. Water can enter the pavement from external sources like poor drainage systems, broken or floating sewer conduits or high water table of the ground. Also, water can be enter the pavement through using wet aggregate in the production of asphaltic mixture. In this research, Iraqi aggregate were classified according to the resistance to water damage effect taking into account the mineral composition and texture. The results obtained refers that crushed aggregate had less resistance than uncrushed aggregate nevertheless, asphaltic mixture without additive for improving resistance to water damage effect, had been failed in boiling water test, where, in the presence of water (pH about 10) the retained asphalt coated aggregate were 45%. Lime was used as additive in three ways: dry method, wet aggregate and slurry lime method. Slurry lime method was improved water damage resistance where coated aggregate after conducting boil test were 85% for uncrushed aggregate and 78% for crushed aggregate. Quartzite compose 80% of aggregate which reflect acidic surface properties, where with the presence of water, high pH values were recorded (about 11). The main conclusions were moisture damage is a serious problem and there is a need to use treatment for elimination water damage through using slurry lime.

INTRODUCTION

The word ‘adhesion’ comes from the Latin word *adherere*, which means ‘to stick to’. A definition of ‘adhesion’ in its scientific context is given by ASTM D907 as “the state in which two surfaces are held together by valence forces or interlocking forces, or both”. ‘Fundamental’ adhesion refers to forces between atoms at the interface, also called ‘true’ adhesion. ‘Practical’ adhesion, on the other hand, is a term that can be used to describe the results of destructive adhesion tests, such as a tensile test. The parameter recorded reflects the fundamental adhesion at the interface as well as the mechanical response of the adhesive glue, substrate, and interfacial region. The mode of failure in tests of this nature can be adhesive (rupture of bonds between molecules of different phases), cohesive (rupture of bonds between molecules within the same phase), or mixed (Arno Hefer and Dallas Little 2005).

Asphaltic mixture consist primarily of aggregate and asphalt and the main function of asphalt to bind aggregate granules together for maintaining mixture stability against loads. Bonding strength depend on strength of adhesion between asphalt and aggregate.

REVIEW OF LITERATURES

The bitumen-water interface and influence of pH on interfacial activity have been studied using electrophoresis. Zeta potential or electrokinetic potential is the electric potential that exists across the interface between a hydrated particle (fixed liquid) and the bulk solution (moving liquid), i.e. a measure of stability at the interface. In addition, this quantity is sensitive to changes in the pH of the system, which is significant, since the pH of water in contact with an aggregate surface changes (Arno Hefer and Dallas Little 2005)and (Kunnawee Kanitpong, et. al. 2003).

The electric potential at this distance is called the zeta potential and can be measured using electrokinetic techniques. The concentration of potential-determining ions (PDI) or pH (log of inverse of hydrogen ion concentration) at which the zeta potential is zero, is defined as the isoelectric point (IEP). The point of zero charge (PZC), on the other hand, is the pH at which the charge at the surface is zero. The study of charge particles in motion in an electric field is called electrophoreses.

Electrophoreses has been applied to bitumen-aggregate systems in the past to explain moisture damage, or stripping, in these systems. However, the existence of bulk water, hydrogen bonded to the aggregate surface in the “dry” state, should not be overlooked. Thelen 1958 conducted experiments on quartz to demonstrate how fresh aggregate adsorb a water layer several molecules thick. At conventional plant mix temperatures at least a molecular layer of water would remain, requiring about a 1000°C to drive it completely off the surface (Bjorn Birgisson 2003).

From the foregoing discussion, pH can be expected to play a role by influencing surface charge. Two aspects should be considered with regard to bitumen-aggregate systems. The first is diffusion of external water to the bitumen-aggregate interface (Fromm, 1974; Nguyen et al., 1992; Cheng, 2002). The pH of this water will differ depending on the environment. Secondly, researchers in the past found that pH of interface water are influenced by the aggregate surface (Yoon and Tarrer, 1988; Labib, 1992; Scott, 1978; Huang et al., 2000). They showed relationships for the changes of pH values when different aggregate powders are added to water. These relationships reveal that many aggregate surfaces tend to increase the pH of the contacting water. This is not restricted to basic rock types (e.g. limestones), but also occurs with acidic mineral composites (e.g. granites) (A.R. Tarrer 1991).

If two surfaces approach each other and electric double layers overlap, an electrostatic double-layer force arises (Butt et al., 2003). Labib (1992) developed interaction diagrams between several aggregates and bitumen types based electrokinetic measurements in the form of zeta potential versus pH curves (Hussain Bahia et. al. 2007).

Mechanism of Failure

Adhesive failure process had been explained by several theories, where most researchers, (Sunghwan Kim, Brian J. Coree 2005, Martin H. Sadd 2002), consider that moisture damage in HMA is due more to the adhesive mode of failure than to the cohesive mode and can be summarized as follows:

- 1) Mechanical adhesion theory.
- 2) Chemical reaction between the asphalt binder and the aggregate (Larry Santucci et. Al. 2002). The reaction of aggregate surface combined with mineral composition of aggregate are collected in Figure 1.
- 3) The differential degree of wetting of the aggregate by asphalt and water.
- 4) Molecular orientation theory.

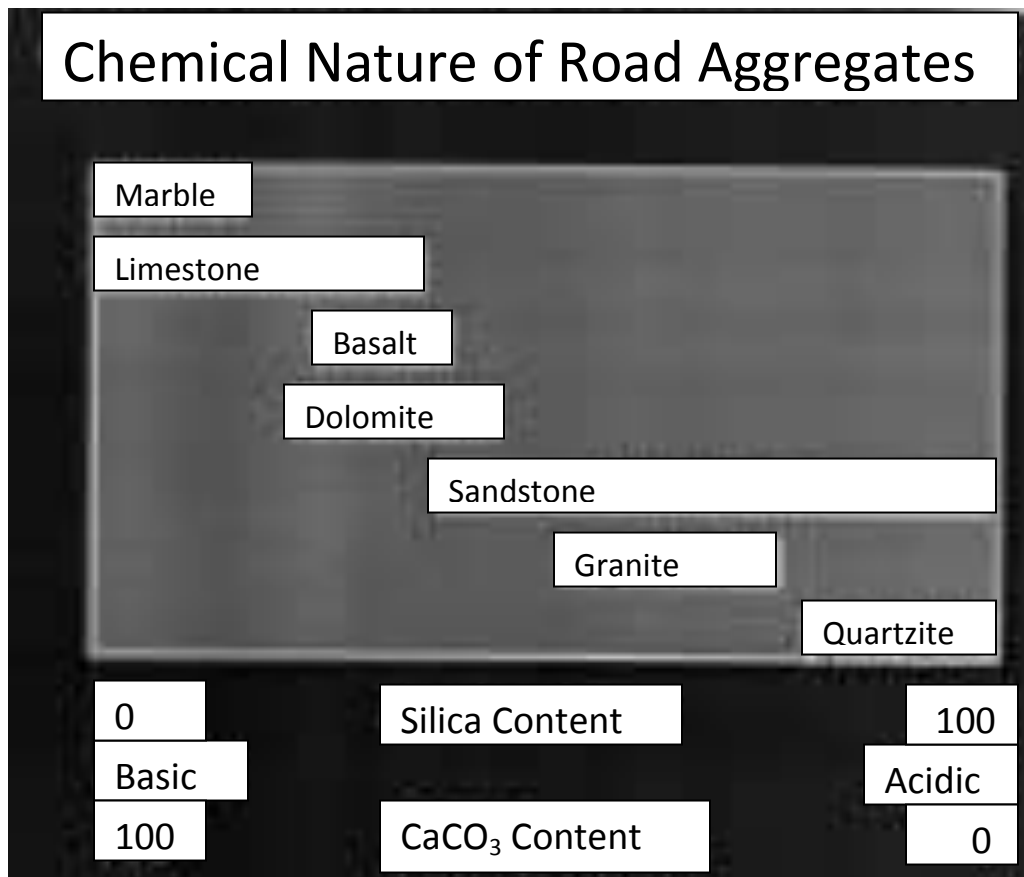


Figure 1. surface reaction of different types of aggregate (Larry Santucci et. Al. 2002).

Another mechanisms had been collected by (Harihar Shiwakoti 2002) as follows:

- 1) Detachment

- 2) Displacement.
- 3) Spontaneous emulsification (Curtis Berthelot 2005).
- 4) Pore pressure
- 5) Hydraulic scour.
- 6) pH instability (Audrey R. Copeland 2007).

As a conclusion (a) the bond between asphalt and aggregate depends on surface chemical activity, (b) water at the aggregate surface (in the field) has a high pH, (c) some liquids used as anti-stripping agents require a long curing period (in excess of about 3 hours) to achieve resistance to loss of bond at higher pH levels, and (d) it is possible to achieve a strong chemical bond between aggregate and asphalt cement that is resistant to pH shifts and a high pH environment. This strong chemical bond can be achieved by the formation of insoluble organic salts (such as calcium-based salts), which form rapidly and are not affected by high pH levels or pH shifts. pH values as high as 9 or 10 do not dislodge amines from the acidic surfaces of aggregates, nor do they affect hydrated lime. Values of pH greater than 10 are not normally developed in asphalt mixtures unless a caustic such as lime is added. However, pH values below approximately 4 can dislodge amines from an aggregate surface and can dissolve lime depending on the type of acid used; these low pH values are not found in hot-mix asphalt.

Effect of Hydrated Lime

Hydrated lime ($\text{Ca}(\text{OH})_2$) was used as anti-strip additive, (Aniruddha Vilas Shidhore 2005) where three ways of adding lime has been evaluated, as follows:-

- 1) Addition of Dry Hydrated Lime to Dry Aggregates: The amount of hydrated lime used in this method is usually 0.9% by the weight of dry aggregates (Dallas N. Little and co. 2001).
- 2) Addition of Dry Hydrated Lime to Wet Aggregates: This marinating process has the following advantages: 1) moisture content is reduced over the period of stockpiling; and 2) due to stockpiling lime treatment can be carried out separately from the main HMA production providing some economic advantage. Disadvantages of marinating are: 1) additional effort required for handling aggregate load; 2) additional space required for stocking both lime-treated and untreated aggregates; 3) carbonation of aggregates could occur due to chemical reaction.
- 3) Addition of Hydrated Lime in the Form of Slurry: Advantages of using this method are as follows: 1) improved resistance of HMA to stripping; 2) as lime slurry is used, lime dispersion due to dusting and blowing is minimized; and 3) this method results in the best coverage of lime over aggregate. The disadvantages of using lime slurries are: 1) use of lime slurries can substantially increase the water content of aggregate resulting increased fuel consumption during drying process; and 2) use of this method requires specialized equipment that is costly to purchase and maintain.

Surface Energy

The fundamental law of fracture for viscoelastic materials was shown in detail by (Robert L. Lytton 2005, Corey James 2005) and is presented in Equation (1). This theory states that fracture damage caused by load-induced energy is balanced by the energy stored on newly created crack faces (Laith Tashman 2003).

$$W = E_R D(t_\alpha) J_R \quad \text{eq. (1)}$$

where, W is the work of adhesion (W_a) or cohesion (W_c) per unit of each crack surface area created (i.e., the minimum energy required to cause fracture).

Methods For Evaluating Moisture Susceptibility

The Static Immersion Test, Boiling Water Test, Texas Boiling Water Test, Texas Freeze-Thaw Pedestal Test, Tunncliff and Root Conditioning, Lottman Test, and the Modified Lottman Test are existing tests used to evaluate moisture susceptibility in HMA mixes. Descriptions of these tests are contained in the following sub-sections (Gary. Conner 2003) and (Elizabeth Rae Hunter 2001).

- 1) STATIC IMMERSION TEST (AASHTO T-182)
- 2) BOILING WATER TEST (ASTM D-3625) (Harold R. Paul 1995)
- 3) TEXAS BOILING WATER TEST
- 4) TEXAS FREEZE-THAW PEDESTAL TEST
- 5) TUNNICLIFF AND ROOT CONDITIONING
- 6) LOTTMAN TEST (NCHRP 246)
- 7) MODIFIED LOTTMAN TEST (AASHTO T-283)

Methods For Limiting Moisture Susceptibility

Moisture sensitive pavements can experience severely reduced service life when subjected to more than trace amounts of moisture. Additives have been developed to address the issues of poor pavement performance and high maintenance costs experienced by moisture susceptible pavements. These anti-stripping additives, whether solid or liquid, are used to promote adhesion of asphalt cement onto the aggregate surface. The effects of commercially available liquid anti-stripping agents on asphalt cement were evaluated by Anderson et al.. Their research showed that the addition of liquid anti-stripping additives can alter the physical characteristics and composition of an asphalt cement, typically increasing asphalt cement viscosity to the point of non-compliance with standard specifications. To date, there has been little guidance as to whether the undesirable effects of anti-stripping agents outweigh their positive moisture resistive effects.

- 1) ANTI-STRIPPING
- 2) ADDITION OF LIME
- 3) LIQUID ANTI-



AGENTS

STRIPPING AGENTS

EXPERIMENTAL PLAN

The first step in this research

was to classify Iraqi aggregate

according to its ability to resist moisture damage effect depending on the previous researches as shown in the previous articles. The test conducted by measuring pH of aggregate powder submerged in water for 30 minutes and recording pH every 1 minute using microcomputer pH meter HI8424 (HANNA instruments) as well as Zeta potential mV. Before conducting the test the device had been calibrated using solutions of predefined pH. Also, readings of pH values at various temperatures were compared with table supplied with instrument for more precise results, the figure of instrument shown in Plate 1. The second step was to evaluate moisture resistance using boil test ASTM D – 3625 for no-additive mixtures, then lime was used as an additive in three ways of addition (dry lime, wet aggregate and slurry). In addition the boil test was conducted at various pH levels to simulate the real condition of road under the attack of water. In this research 0.9 % by weight of dry aggregate lime was used as additive, where selected from the previous literatures. For best estimation of retained coated aggregate, a picture had been taken and displayed on computer screen, then the picture of sample fragmented into squares to determine the coverage percentage in addition to visual estimation. Plate 2. a&b shows pictures taken for samples before and after boil test.

Plate 1. Microcomputer pH meter used in evaluation.



a. Before boil test



b. after boil test

Plate 2. samples before and after conducting boil test using slurry lime

ANALYSIS OF RESULTS

Three types of Iraqi aggregate were selected and tested to determine pH of aggregate powder. Chemical and mineralogical composition are shown in Table 1, (Haydar 1998) and the results are shown in Figure 2. It is obvious that pH of Quartzite increase with time to reach about 9 after 30 minutes. From Table 1, 80% of aggregate composition is Quartzite, which reflects weak resistance to moisture attack due to acidic reaction of aggregate surface when being in contact with water as shown in Figure 1, especially with poor drainage systems and/or float of sewer conduits.

Table 1. Chemical and mineralogical composition of aggregate, (haydar 1998)

Chemical Compound	%
Sio2	82.52
Mgo	5.37
So3	0.78
Fe2O3	2.7
Al2O3	0.69
L.O.I.	0.48
Total	6.55
Mineral Composition	99.09
Quartz	80.03
Calcite	10.92

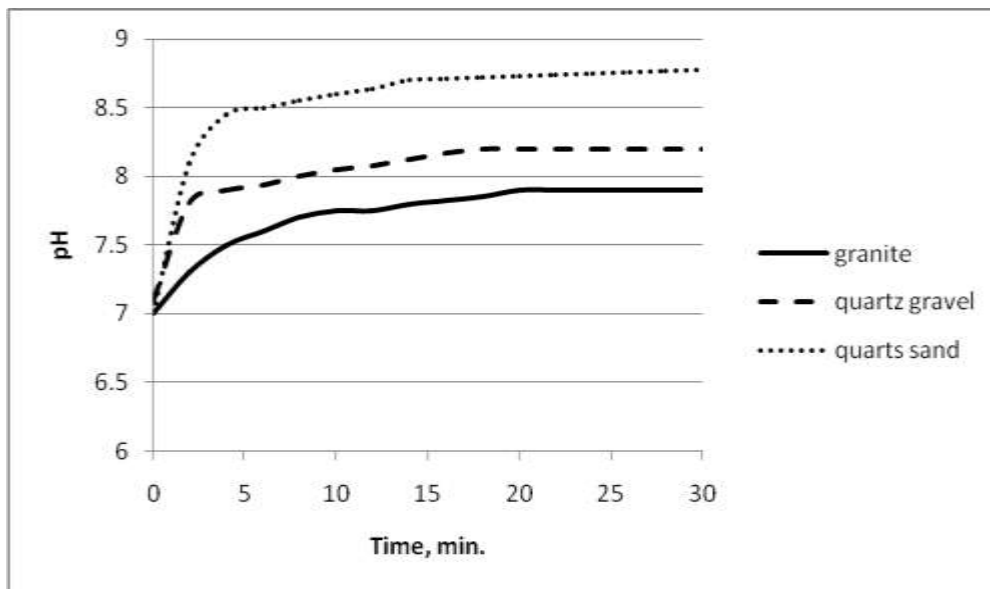


Figure 2. pH for different types of aggregate

Zeta potential with ph diagram for asphalt and limestone and granite aggregates as shown in Figure 3. If two surfaces approach each other and electric double layers overlap, an electrostatic double-layer force arises (Arno Hefer and Dallas Little 2005). As shown in the Figure 3, the isoelectric point for asphalt and quartzite aggregate is around four, while at pH around 7 (dry mixture), both of asphalt and quartzite are negatively charged. This indicates that the strength of adhesion is low and in wet

condition ($\text{pH} > 9$) both surfaces has high negative charges which may lead to repulsion. For mixture of limestone and asphalt at dry condition (pH around 7) both surfaces has opposite charges which lead adhesion. The strength of adhesion depends on the magnitude of difference between charges, while the strength of adhesion will decrease as water present ($\text{pH} > 8$) due to negative charges of both sides.

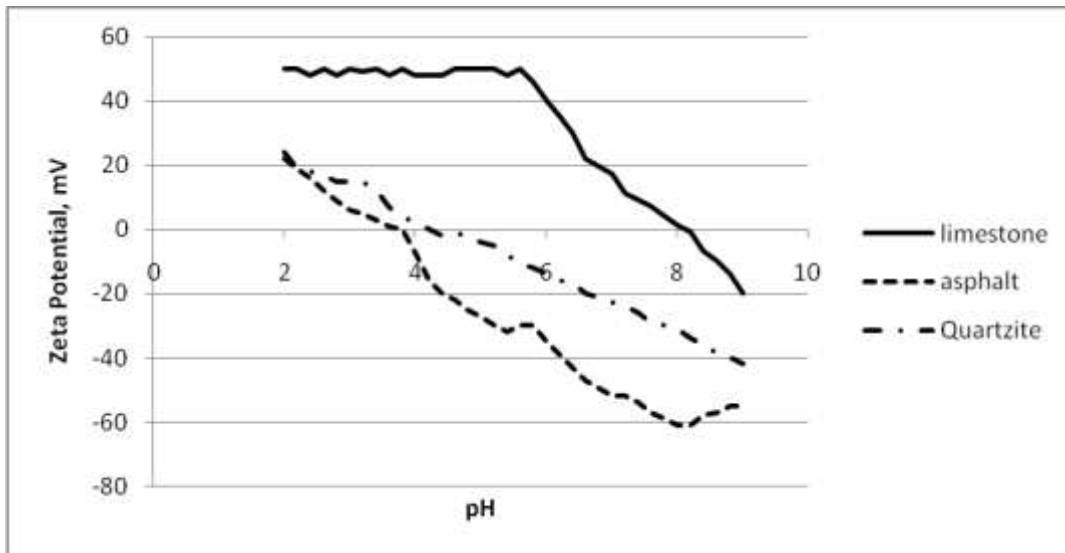


Figure 3. Effect of pH on zeta potential, mV on asphalt and two types of aggregate

Evaluation of moisture susceptibility

Boiling water test ASTM D – 3625 results are shown in Figure 4 using the three methods of adding lime (dry, wet, and slurry) as well as with no-lime at $\text{pH}=7$. From the results obtained, adding lime as slurry to the asphaltic mixture will increase the resistance of road to water attack. Also, it can be noticed that the resistance of uncrushed aggregate mixtures showed more resistance to water attack of about 5% as compared to crushed aggregate mixtures using the same method of adding additive. This increase may be attributed to the aggregate texture, where crushed aggregate has acute angles which may has less resistance.

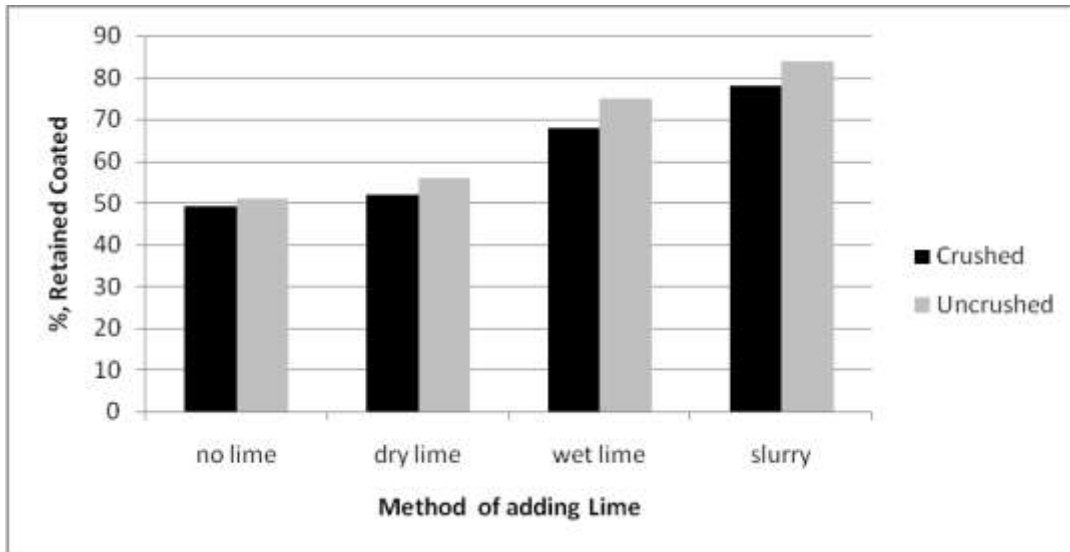


Figure 4. Effect of methods of addition of lime on moisture susceptibility

The evaluation of moisture susceptibility at different pH levels has been evaluated and presented in Figure 5, For uncrushed aggregate mixtures and Figure 6, for crushed aggregate mixtures where at dry condition (pH about 7) all mixtures showed more resistance to moisture damage as compared to high acidity or alkalinity levels. In addition, 85% of retained coated aggregate was obtained for uncrushed aggregate mixtures when adding lime as slurry and about 75% for wet aggregate method and less than 55% coated for dry lime method, while, the less result was obtained for mixture with no-lime where about 50% coated aggregate mixture where obtained. The increase of coated aggregate will be about 35% more than same mixtures with no additive, while, for crushed aggregate mixture the highest retained coated aggregate was slightly less than 80% using slurry lime, less than 70% for wet aggregate and more than 50% for dry lime addition.

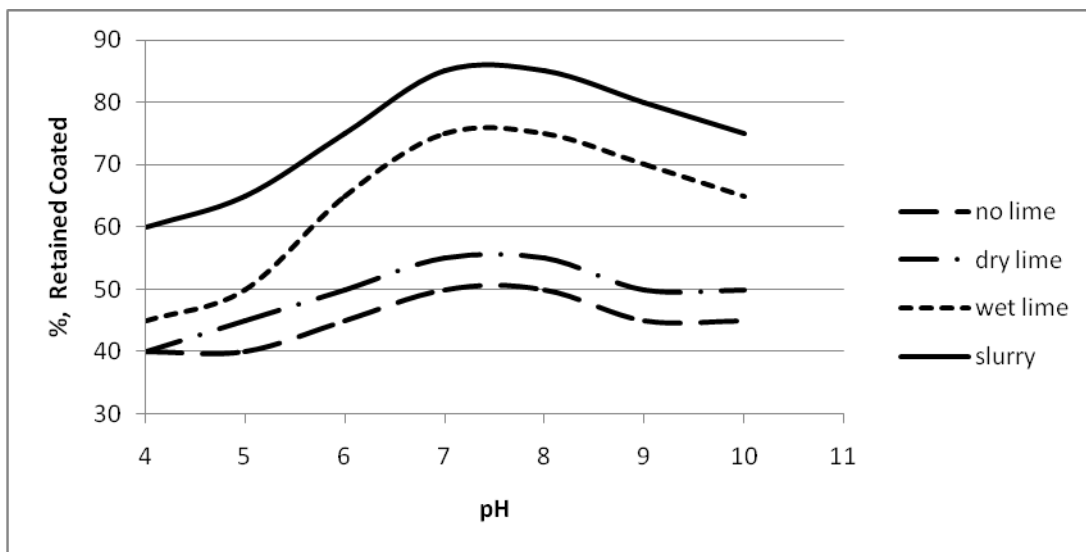


Figure 5. Effect of lime addition methods on moisture susceptibility at different pH levels (uncrushed)

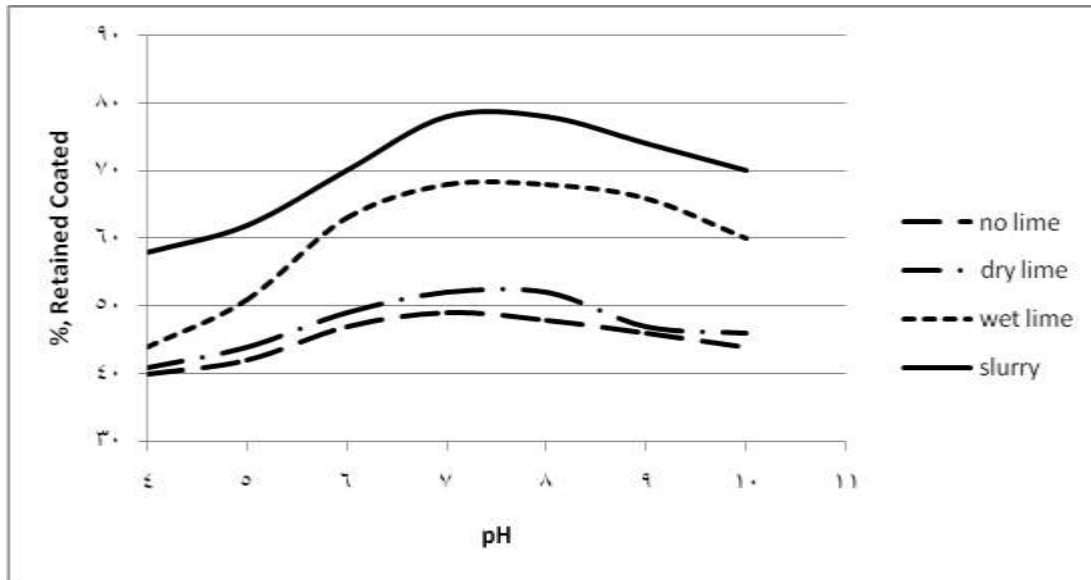


Figure 6. Effect of lime addition methods on moisture susceptibility at different pH levels (crushed)

CONCLUSIONS

According to the tests were conducted, materials used in this research and literatures cited, it can be concluded that:-

- 1) Increase of pH values of water surrounding aggregate had been recorded, reached high values of about 11 where at this value, the mixtures tested were heavily stripped according to boil test.
- 2) Surface properties of aggregate has an effect on adhesion between asphalt cement and aggregate with or without the presence of water, where similar charges of asphalt and aggregate surface were recorded (Figure 3) which lead to contraction between the two surfaces. The presence of water will increase the problem through raising pH value of interlayer between aggregate surface and asphalt.
- 3) Mineral composition of Iraqi aggregate consist of acidic surface properties (80% Quartzite), which will lead to early adhesion failure for untreated asphaltic mixtures. The treatments using lime in the form of slurry will increase the adhesion (high values of coated aggregate) between asphalt and aggregate surface for about 35% for uncrushed aggregate at normal condition (pH=7) and about 30% at high moisture rate (pH>9), while for crushed aggregate the increase was 27% at normal condition and 24% at sever condition (pH>9) .
- 4) Boiling water test according to ASTM D 3625 is reliable for using in assessing stripping resistance of asphaltic mixtures where it is easy to conduct and fast results could be obtained, also possible to perform in-situ. The recommended retained coated aggregate is 75%.
- 5) Aggregate texture is a factor that affecting strength of adhesion between asphalt and aggregate. This effect could be attributed to the break of sharp edges due to traffic loads, which give permission for water to penetrate interlayer of asphalt and aggregate. The using of slurry lime has reduced the effect of this phenomenon.

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