

□ 論 文 □

道路鋪裝 構造物에서의 스트리핑 現象과 스트리핑 防止劑의 利用方案

Stripping of Asphalt Pavements and Antistripping Additives

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요 약

물 또는 濕氣에 의하여 道路鋪裝 構造物의 아스팔트와 자갈(aggregate)사이의 結合力이 損失되어 아스팔트가 자갈로부터 分離되는 現象 즉 스트리핑(stripping)에 의하여 많은 道路鋪裝이 破壞되고 있다. 이러한 스트리핑 現象을 防止하기 위하여 여러가지 方法이 開發 使用되고 있는데 흔히 스트리핑 防止劑(antistripping additive)를 아스팔트에 섞어서 使用하고 있다. 本 研究의 目的은 스트리핑 現象의 原因을 糾明하고 이것을 바탕으로 효율적인 스트리핑 防止劑의 條件과 使用方法을 提示하는 것이다.

道路鋪裝用 자갈의 物理化學的 性質을 觀察하여 스트리핑과의 關係를 檢討한 結果 스트리핑 現象은 아스팔트와 자갈 사이의 電氣的 反發力에 의해 일어날 수 있으며, 자갈 表面의 電氣化學的 性質이 스트리핑에 重要的 役割을 함을 알 수 있었다. 또한 現在 使用되고 있는 스트리핑 防止劑는 IR 分析結果 대부분이 아민(amine) 系統의 陽이온 界面活性劑 이었다. 이러한 스트리핑 防止劑는 0.5에서 1.0%정도 아스팔트에 섞어서 使用하면 스트리핑을 현저히 줄일 수 있으나, 장시간 (약 24 시간 이상) 뜨거운 상태에서 放置하면 그 性能이 감소되는 것을 알 수 있었다.

I . INTRODUCTION

Due to its superior engineering qualities and ready availability, asphalt has been extensively

used as cementing material in road construction. Over 70% of all asphalt produced in the U.S. is used for road pavements; more than 90% of all road pavements is asphalt

road and less than 10% concrete road. Most of those asphalt pavements have been designed to resist variety of traffic loads and environmental stresses. However, in recent years, an increasing number of pavement failures associated with moisture damage, or "stripping", have been observed.

Stripping in asphalt pavement is defined as the displacement of asphalt films from aggregate surfaces due to the loss of the adhesive bond between aggregate and asphalt. The cause of stripping is attributed to the presence of water or moisture in the asphalt-aggregate matrix. Stripping results in a loss of integrity of asphaltic concrete and subsequent failures, requiring early and costly maintenance.

Stripping action is usually initiated at the bottom of the pavement, and once initiated, it progresses rapidly. Therefore, the damage caused by stripping is usually not apparent until after the pavement has failed over a large area.

1. Stripping Mechanisms

Over the years, great deal of basic and applied research has been conducted to determine the nature of the adhesion and the stripping phenomena(1-5). A number of studies have been concerned with laboratory test methods for predicting the stripping sensitivity for asphalt-aggregate mixtures(6-8) and the development of methods to prevent or minimize the occurrence of such damage when stripping-susceptible materials are used(9-12).

Stripping is complex phenomena and has been related to many factors including the physical and chemical properties of both aggregate and asphalt. Mixture design, construction, traffic loading, and climate also can

affect stripping. Several mechanisms by which stripping may occur have been proposed by previous investigators(2,3). The most widely accepted mechanisms are detachment, displacement, spontaneous emulsification, and pore pressure mechanisms(5).

The rationale for explaining these stripping mechanisms is provided by the mechanical, thermodynamic or interfacial energy, and/or chemical concepts of adhesion and loss of adhesion. The mechanical concept for explaining stripping mechanism suggests that the bond strengths between asphalt and aggregate surface is dependent upon mechanical interlock developed by the penetration of the asphalt into pores and cracks of the surface of the aggregate particles. The thermodynamic or surface energy concept involves the wetting behavior of asphalt at the asphalt-aggregate-water-air interface. The degree to which stripping occurs depends on the interfacial free energy relationships at the aggregate-asphalt-water-air interface. The chemical concept involves adsorption of asphalt on aggregate surfaces and chemical reactions between the adsorbed asphalt compounds and the constituents of the aggregate surface. Herein, water solubility of the asphalt-aggregate bond is the main factor affecting stripping(1-5).

It is generally felt that the primary mechanism responsible for stripping involves the displacement by water of an asphalt film from the aggregate surface. The actual role of water in stripping is, however, still not entirely understood. Also, it has been difficult to relate quantitatively stripping potential to materials selection and mixture design parameters.

The purpose of this research work was to

study the nature of stripping in asphalt paving mixtures and to determine the importance of the physical and chemical properties of aggregate in the mix.

Table 1 gives a summary of the physico-chemical properties of asphalt, aggregate, and the asphalt-aggregate mixture that might influence stripping, based on different mechanistic stripping theories. Although all these properties are involved, this research was primarily concerned with aggregate properties such as surface area, porosity, and chemical and electro-chemical properties.

Table 1. Physico-Chemical Properties of Asphalt, Aggregate, and Mixture Influencing Stripping.

Materials	Physicochemical Properties
Asphalt	Viscosity Surface Tension Volatility Chemical Composition
Aggregate	Size and Shape Pore Volume and Size Surface Area Chemical Constituents at Surface Acidity and Alkalinity Adsorption Site Surface Density Surface Charge or Polarity
Mixture	Pore Space Fraction Filled with Asphalt Asphalt Adsorption Ratio Chemical Constituents of Adsorbed Asphalt

Several different types of aggregates were selected for this study and physical properties, such as surface area and pore volume, were measured. The change in pH of water after aggregate addition, and the surface charge of aggregates in water were also measured. These measurements gave some information

on the chemical properties of the aggregate surface.

2. Antistripping additives

Soon after stripping was recognized, various methods were developed and used to prevent or minimize the stripping damage. It is generally felt that stripping is primarily an aggregate problem, although the type of asphalt is also important. However, it is not always possible to choose the proper types of aggregates for obtaining durable adhesion properties in the mixtures. In many locations only water-susceptible aggregates are available. Thus, because, there are little alternatives available in aggregates, an alternative must be found in another area, namely that of improving the adhesion of asphalt to the aggregate by modifying the system. The following practices are commonly used when water-susceptible aggregates are used :

- (1) Addition of a chemical antistripping additive,
- (2) Addition of hydrated lime of portland cement,
- (3) Removal of surface coatings, and
- (4) Preheating of aggregate.

The selection of which method to use depends on a variety of efficiency and economic consideration. Because of its relatively low cost and ease of implementation, the addition of chemical antistripping agent is the most widely used method. Recent studies(11-13) have concentrated on searching for more effective and lower cost chemical antistripping additives. Today more than 100 approved chemical antistripping additives are used in the United States. The usage of antistripping additives in some European countries, particularly in Sweden, England, France, and

west Germany, is far ahead of usage in the United States.

Antistripping additives are substance added to asphalt to promot adhesion of the asphalt to the aggregate surface and thus to improve the resistance to stripping damage in asphalt pavements. Although these additives have been used in asphalt paving mixtures for many years and in recent years their use has increased significantly, there have been only a few studies of antistripping additives and their effectiveness. The lack of information concerning antistripping additives is mainly due to the fact that most antistripping additives used today are proprietary chemical compounds. In fact, the mechanisms controlling antistripping additive action in reducing stripping damage to the asphalt-aggregate mixture are not completely understood.

Fundamental information concerning antistripping additives and their effectiveness is needed in order to develop criteria for the selection and use of antistripping additives and to ensure that the use of antistripping additives improves the short-term and long-term performance of given asphalt and aggregate mixture if placed in pavements.

The main purpose of this work is to provide information on the selection and use of antistripping additives. To accomplish this objective, the following were investigated:

- (1) Characterization of antistripping additives,
- (2) Effectiveness of antistripping additives, and
- (3) Effect of heat on the additive performance.

II . STRIPPING TESTS

The most valuable means of studying strip-

ping and antistripping additive effectiveness are tests which duplicate field conditions. However, since stripping is related to many factors including aggregate and asphalt properties, mixtur design, construction procedures, and environmental conditions, it is extremely difficult to simulate conditions encountered by actual pavements. There has been no generally accepted, reliable procedures for testing stripping and additive effectiveness. However, various experimental procedures concerning stripping and antistripping additives have been developed and used. Each provides information that may be useful in stripping and additive effectiveness studies.

Table 2 (9) shows various testing procedures, concerning stripping and antistripping additives used by state highway agencies in the United States for given purposes. Most testing is performed on complete mixtures. Complete mixture testing is more reasonable for determining the susceptibility to stripping of a given asphalt-aggregate mixture because stripping is affected by physical and chemical properties of both the asphalt and the aggregate. Test on compacted mixtures, however, can not be used to isolate particular stripping mechanism or mechanisms of additive function, because mechanical properties of compacted mixture, which are to be measured and related to stripping propensity of the mixture, depend on too many variables such as aggrrgate size and shape, asphalt viscosity and source, compaction method, and so on.

Among the coating evaluation tests, the boiling water test (ASTM D3625-77) is most widely used. The boiling water test in various form has been used for several decades to pre-

Table 2. Application of Stripping Test Procedures

Procedure	Number of Agencies Using Procedures to Determine Additive				
	Need	Dose	Presence	Heat Stability	Effectiveness
Additive Indicator Tests					
Bottle Test	—	1	1	3	2
Color Indicator	—	1	1	—	1
Tests on Compacted Mixture					
Immersion-Compression	16	9	2	—	5
Marshall Immersion	10	7	2	2	2
Tensile Splitting	8	4	—	—	3
Abrasion	3	1	—	—	—
Hveem Stability	1	1	—	—	—
Resilient Modulus	1	—	—	—	—
Swell and Absorption	1	—	—	—	—
Tests on Loose Coated Mixture					
Static Immersion	9	1	—	3	—
Dynamic Immersion	7	—	—	—	—
Tracer Salt	1	—	—	—	—
Boiling Water	5	2	3	7	5
Total	64	27	9	15	18

dict moisture damage in actual asphalt pavements. In this test, a specific amount of aggregate and asphalt are mixed at a specified temperature until complete coating is achieved. After cooling for a given period of time, the mixture is placed in boiling water for a specified period of time. The stripping potential is evaluated by visually observing the extent of asphalt loss from the aggregate surfaces following boiling.

The boiling water procedure is relatively simple and rapid, and also has reasonable correlations with field performance as reported by Kennedy(8). Many state highway

agencies are using this test, especially for predicting the presence, effectiveness, and heat stability of antistripping additives. In addition, due to its evaluation by visual observation of stripping phenomena, the boiling water test provides fundamental information (i.e., the contact angle) on stripping and adhesion of asphalt and aggregate in the presence of water. Hence, the boiling water test was chosen and used throughout the investigation. If the asphalt separated from the aggregate surface during this test, it was said to have stripped, and the asphalt-aggregate bond was concluded to have been weak and to have

a high stripping propensity.

III . STRIPPING MECHANISM

Five different aggregates were characterized by measuring their pore volumes, surface areas, pH values of contacting water, and surface charges in water(14). The experimental results were evaluated with respect to the effect of each of these aggregate characteristics—(1) pore volume and surface area, (2) pH value of contacting water, and (3) surface charge of aggregate—on the stripping propensity of the aggregate as measured by the boiling water test. The results are discussed below.

1 . Pore volume and surface area

Five aggregates (granite, limestone, dolomite, chert gravel, and quartz gravel) were tested for surface area and porosity, and their stripping propensity was determined by the boiling water test, as shown in Table 3. A low pore volume or surface area suggests a smooth, crystalline surface with low surface roughness. From purely mechanical considerations—that is, the requirement of large areas of interfacial contact and surface roughness to have good adhesion and interlock—the low pore volume and surface area of granite

should imply the existence of a low adhesive bond strength with the asphalt and a high moisture susceptibility.

A comparison of the effect of different pore sizes was provided by dolomite and limestone, as can be seen from Table 3. Dolomite and limestone were used for this comparison because of their similarity in chemical and electrochemical properties. Although dolomite had a higher surface area than did limestone, it had a higher stripping propensity because of its smaller pore size. It should be noted that, even though dolomite had nearly the same amount of pore volume as did the limestone, it had about seven times as much surface area, which meant that dolomite had a smaller pore size. In Table 3, the average pore size for each of these aggregates was estimated by assuming the pores to have a cylindrical shape.

One possible reason for the observed adverse effect of pore size on stripping propensity for the dolomite and limestone aggregates is that, when an asphalt coats over a rough surface having fine pores, air is trapped and the asphalt can hardly penetrate the fine pores. Consequently, only a fraction of the aggregate's apparent surface area might actually be in contact with the asphalt. In general, the depth of penetration of the asphalt

Table 3. Pore volumes and surface Areas of Aggregates and Their Stripping Propensities

Aggregate	Pore Volume 1000cc/g	Surface Area m ² /g	Percent Coating after Boiling
Granite	3.2	0.116	10
Dolomite	6.5	0.586	35
Chert Gravel	23.0	2.09	55
Quartz Gravel	5.4	0.052	65
Limestone	6.2	0.079	90

depends upon the size of the pore, as well as the asphalt viscosity and surface tension.

Overall, a correlation between aggregate physical properties and stripping propensity could not be established. The chemical properties of the aggregates considered varied significantly, and it was felt that this had been an overriding effect. As shown in Table 3, although limestone and crushed quartz gravel had nearly the same physical properties in terms of pore volume and surface area, limestone had a better stripping resistance. The chemical properties of the limestone were much different from those of the quartz gravel, as will be discussed.

2. pH of contacting water

In this study, some insight as to the effect of pH changes of contacting water was developed by considering the chemical and electrochemical properties of the aggregate surfaces. Figure 1 shows the changes in pH caused by the addition of several different aggregate powders to water. Apparently, lime-

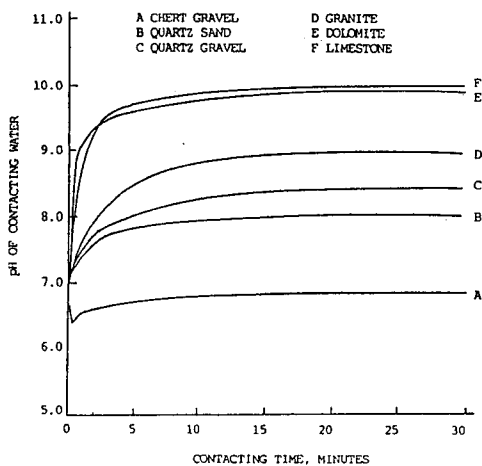
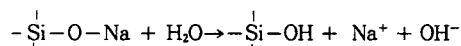


Figure 1. Change in pH of Water in Which Aggregates Were Immersed.

stone and dolomite, which are known to be basic aggregates, caused a rise in pH of the water to a relatively high value. Also, granite, which is known to be acidic, reacted with water leading to a gradual increase in the pH of the system. The silicate lattice of the granite surface reacted with water to impart excess hydroxyl ions as follows :



This chemical reaction illustrates a typical hydrolysis reaction of a salt of a weak acid.

While coating an aggregate with asphalt, the aggregate selectively adsorbs some components of the asphalt, such as the more polar species of the asphalt, forming hydrogen bonds or salt links. The types and quantities of adsorbed components are thought to play an important role in adhesion and stripping. (4) The presence of ketones and phenolics are thought, for example, to improve stripping resistance; whereas, carboxylic acids, anhydrides, and 2-quinolenes to increase stripping sensitivity, because of the high water susceptibility of their bonds with aggregate surfaces.

The water susceptibility of the hydrogen bonds (or some other dipole-dipole attractive bonds) and salt links (i.e., the ionic bonds) between the adsorbed asphalt components and the aggregate surface would increase as the pH of the water present at the aggregate surface was increased. For this reason, stripping damage might be expected to occur for an aggregate which causes an increase (to a relatively high value) in the pH of any water present at the asphalt/aggregate interface.

The data presented in Figure 2 appears to agree with the above suggestion. Granite, which imparted a high pH to contacting

water, had a higher stripping propensity than did either crushed chert gravel or quartz gravel, both of which imparted a lower pH to the contacting water. The pH values indicated in Figure 2 were obtained by measuring the pH of water used in the boiling water test after cooling for each aggregate.

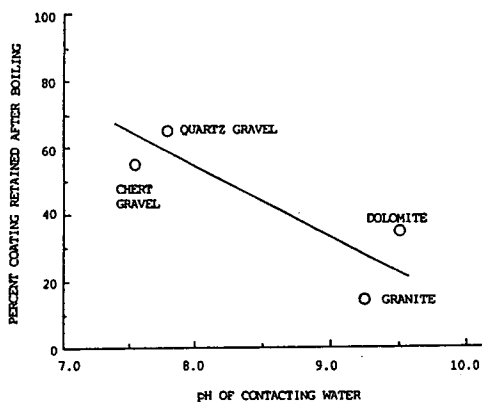


Figure 2. Comparison of pH of Contacting Water and Stripping Propensity as Determined by the Boiling Water Test.

3. Surface Potential

The responses in stripping propensity to differences in the surface electrical charge of the aggregates are given in Figure 3. The interfacial activity occurring between charged surfaces of the mineral aggregates and asphalt can be of fundamental importance to the nature of stripping of asphalt from the aggregate(15). That is, the surface charge of the aggregate can be as important as are specific chemical interactions; in fact, mineral aggregates possess distinctive polarities or electrochemical properties.

The functional group types of an asphalt adsorbed on an aggregate surface consist mainly

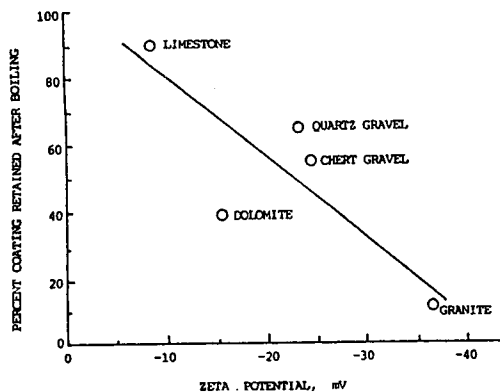


Figure 3. Comparison of Aggregate Surface Potential and Stripping Propensity Determined by the Boiling Water Water Test.

of the acidic fraction of the asphalt. For instance, one of the acid molecules is represented by carboxylic acids ($R-COOH$). In the presence of water, the acid molecules are separated into two ions; the carboxylate anion ($R-COO^-$) and the proton (H^+), causing the asphalt surface to have a negative polarity at the interface. The increase in pH of water present at the aggregate surface increases the extent of dissociation of the acid molecules.

The aggregates with water present are negatively charged to varying degree, as seen in Figure 3. As a result, a repulsive force develops between the negatively charged aggregate surface and the negatively charged asphalt surface at the interface, causing the separation of the asphalt from the aggregate surface (stripping). Solid surfaces in contact with water usually acquire charges through chemical reaction at the solid surface and adsorption of complex ions from the solution (16).

The intensity of the repulsion developed between the asphalt and the aggregate de-

depends on the surface charge of both asphalt and the aggregate. As shown in Figure 3, granite, which had a high stripping propensity, possessed a relatively high surface potential; whereas, limestone, which had a high stripping resistance, had a relatively low surface potential (as determined by zeta potential measurements). The general observed trend was that the aggregate which had a relatively high surface potential in water were more susceptible to stripping.

From the foregoing discussion, it appears that, although physical properties of the aggregate are important, chemical and electrochemical properties of the aggregate surfaces play an even more important role in stripping.

It should be noted that the values of pH and surface charge indicated in this study are only true for the sample tested since these values will be changed with variation of the mineral source and its history of aging.

IV. ANTISTRIPPING ADDITIVES

The theory of surface energy relations between liquid and solid indicated that the contact angle of a liquid drop on a solid surface is a measure of bond strength (17), as illustrated in Figure 4. The contact angle can then be used as a measure of the additive effectiveness. Hence, the contact angle of asphalt drops on aggregates immersed in water was observed for asphalts with and without antistripping additives to determine the fundamental characteristics of antistripping additive action.

The liqued asphalt drops were formed during the boiling of the asphalt-coated aggregates (3/8 inch—No. 4 mesh size granite) in water

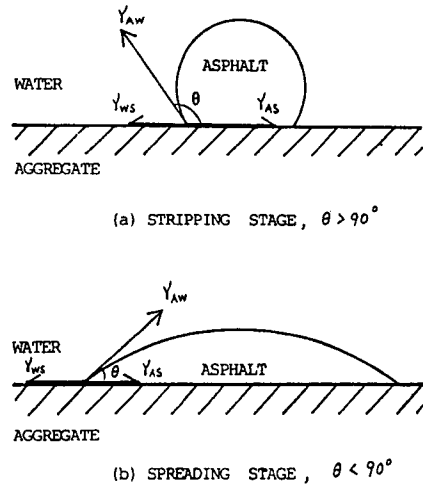


Figure 4. A Droplet of Liquid Asphalt on the Aggregate Immersed in Water.

under the influence of interfacial forces. The shape of the asphalt drop on the aggregate surface and thus the contact angle were observed visually and qualitatively after cooling. The shape was unchanged after cooling. The percentage of asphalt coating retained on the aggregate surface, the boiling water test result, was also visually estimated.

Table 4 illustrates the schematic diagram of visual observation for contact angle and the boiling water test results. The contact angle was decreased and hence the forces of adhesion between asphalt and aggregate were increased markedly by the presence of an antistripping additive. Therefore, an effective antistripping additive can improve adhesion of asphalt to the aggregate surface and thus the stripping resistance of the asphalt-aggregate mixture if enough amount of the antistripping additive is present in the asphalt.

Several of the more common commercial antistripping additives were examined for their functional group types by observing their

Table 4. The Effectiveness of Antistripping Additives as Determined by Boiling Water Tests and Contact Angle Observation.

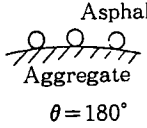
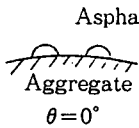
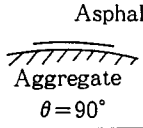
Antistripping Additive	Percent Coating Retained after Boiling	Schematic Diagram for Shape of Asphalt Retained after Boiling, and Approximate Contact Angle
Control AC-20-1	10	 <p style="text-align: center;">Asphalt Aggregate $\theta = 180^\circ$</p>
0.2% Additive No. 1	50	 <p style="text-align: center;">Asphalt Aggregate $\theta = 0^\circ$</p>
1.0% Additive No. 1	100	 <p style="text-align: center;">Asphalt Aggregate $\theta = 90^\circ$</p>

Table 5. Compositional Properties of Antistripping Additives

Additive No.	Major Functional Infrared Band, cm^{-1}				
	Deformation NH_2 (1)	Stretch CN (2)	Wag NH_2 (3)	Wag NH (4)	Other
1	1,650	1,050	870 (small)	720 (small)	3,270, 2nd amine, amide
2	1,650	1,070~1,120	820~840	730 (small)	3,280, 2nd amine
3	1,630	1,070~1,120 (small)	830 (trace)	720	3,300, amide
4	1,630	1,070~1,120	830 (small)	725	3,280, 2nd amine
5	1,630	1,070~1,120	820~840	730 (small)	3,280, 2nd amine
6	none	1,050~1,080	none	720	3,350, amide
7	—	—	—	—	—
8	1,630	1,050~1,120	830 (small)	720	3,300, 2nd amine

* Additive Nos. 1-7 contained salts of carboxylic acid at about $1,560 \text{ cm}^{-1}$.

- (1) Typical for primary amines
- (2) Typical for organic nitrogen compounds
- (3) Typical for primary amines
- (4) Typical for secondary amines

infrared spectra. Table 5 shows functionalities contained in each antistripping additives. The result showed that major functional infrared bands of all additives, except additive No. 7, which was iron naphthenate, were those typical for primary and secondary amines or for organic nitrogen compounds, suggesting amines or amine derivatives. Additive No. 2, No. 4, and No. 5 had relatively high primary amine components. In general, amines act as cationic type surfactants. It was, therefore, concluded that most antistripping additives used in the United States are cationic surfactants, which render an aggregate surface hydrophobic by being adsorbed on the negatively charged mineral aggregate surface and by reducing the surface tension of asphalt, and which thus make it far easier for asphalt to adhere to the aggregate.

In order to determine effectiveness and optimum dosage, the stripping test was carried out for several different antistripping additives with varying dosage. Figure 5 shows the

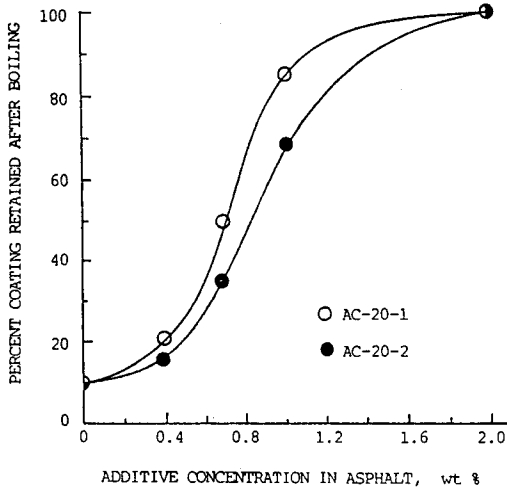


Figure 5. Effectiveness of an Antistripping Additive as a Function Concentration in Asphalt.

effectiveness of each antistripping additive as a function of its concentration in two different asphalts. The results varied considerably with the different asphalts and with the different antistripping additives. However, in general, the antistripping additives improved the adhesion or stripping resistance to some extent, depending on their concentration in the asphalts. The results indicate that the dosage of an antistripping additive is more important than the type of the additive. Although some additive were more effective than others at some dosage levels, the effectiveness of all antistripping additives tested was improved as the concentration of the additive in the asphalt increased.

In order to better use antistripping additives, the effect of important paving mixture manufacturing process conditions of the performance of antistripping additives was investigated. It was found that many antistripping additives were susceptible to heat (18). Therefore, storing the additive in hot asphalt can severely reduce the effectiveness of the additives. For this reason, the effect of additive holding in hot asphalt was investigated by carrying out stripping tests with heat treatment of antistripping additive and asphalt mixtures.

All antistripping additives tested in this investigation lost their effectiveness and failed to function to some extent when maintained for hours in a hot asphalt, as shown in Figure 6. Although, certain additives, such as Additive No. 3 did not deteriorate considerably until 48 hours of heating at 325 F, all these additives known to be efficient could not withstand these temperatures for prolonged periods; therefore, the original efficiency of the antistripping additives was lowered. This

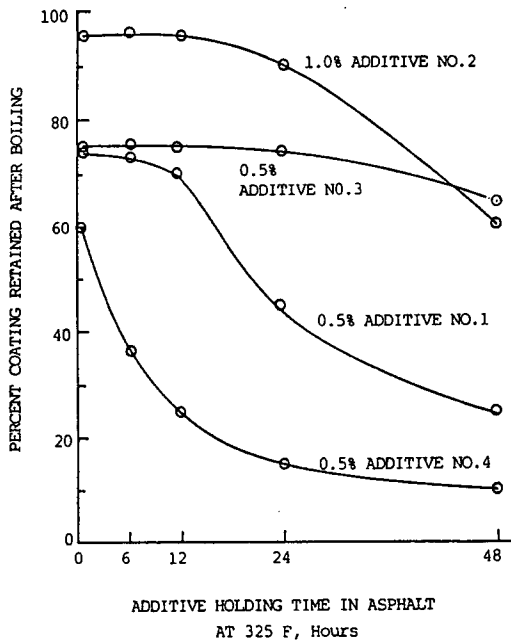


Figure 6. Hest Stability of Antistripping Additives as Determined by the Boiling Water Test.

finding suggests that for best results the antistripping additives must be added on-site in asphalt distributor or at least during the same day the asphalt is to be used. The more heat unstable of an antistripping additive, the higher dosage level may be required.

V. SUMMARY

Physico-chemical properties of asphalt, aggregate, and asphalt-aggregate mixture that might influence stripping were summarized in Table 1, based on the fundamental theories concerning stripping. It was found that although physical properties of aggregate affected stripping, there was no strong correlation between the physical properties of aggregate, such as pore volume and surface

area, and the stripping propensity of the aggregate. Chemical and electrochemical properties of aggregate surface in the presence of water were most important factors for stripping.

All mineral aggregates tested in this study imparted distinctive pH values to the contacting water and possessed distinctive electrochemical properties as measured by zeta potential. It was found that aggregates which had relatively higher surface potential in water and/or which imparted relatively higher pH to the contacting water were more susceptible to stripping.

The functionalities contained in antistripping additives tested were primary and secondary amines and those of organic nitrogen compounds. The functionalities were determined by examining their infrared spectra.

Based on the interfacial energy concept, the contact angle of an asphalt drop on an aggregate surface immersed in water related to the stripping propensity. The contact angle and stripping propensity were markedly reduced by the presence of an antistripping additive. In general, all the additives tested improved stripping resistance to some extent, depending on their concentration in the asphalts. The optimum dosage of an additive varied with different asphalts, as well as different aggregates.

All antistripping additives tested in this study lost their effectiveness and failed to function to some extent when maintained for hours in a hot asphalt.

VI. ACKNOWLEDGEMENT

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