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# Absorption of Asphalt Into Porous Aggregates

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# Absorption of Asphalt into Porous Aggregates

## Introduction

Aggregates used in building asphalt pavements generally have some porosity which tends to absorb asphalt into the pore structure. This fact is becoming more important as the sources for higher quality aggregates are used up and lower quality higher porosity aggregates must be used in their place. In addition, geographic proximity often predicated the utilization of marginal, more absorptive aggregates. Absorption is a very complex subject. Although some absorption may lead to improved strength in compacted mixtures due to mechanical interlocking, the portion of the asphalt which is absorbed is not available for binding the aggregate particles together in the asphalt paving mix. In addition the time dependence of absorption can lead to incorrect estimation of air voids, an important parameter in asphalt mix design. Furthermore, if selective absorption occurs, then the nonabsorbed asphalt may have different physical, rheological, and chemical properties than the original asphalt to such an extent that the nonabsorbed asphalt would behave much differently than the original asphalt as a binder. Little is known about selective absorption. In terms of mixture performance, the nonabsorbed bulk asphalt (effective asphalt) may be the weak link because selective absorption may promote extraction of certain asphalt fractions such as the polar compounds, aromatics, or saturates from the maltenes, leaving the bulk phase rich in such fractions as asphaltenes. This component segregation may lead to less temperature susceptible bulk phases which may in turn be more susceptible to distresses such as, low temperature cracking, fatigue and moisture damage.

It is thought that asphalt absorption, with its consequent effects upon the remaining effective asphalt film, may contribute to premature pavement failure through various mechanisms such as water stripping, accelerated aging, and cracking. The amount and extent of asphalt absorption will depend on such asphalt properties as viscosity,

composition as measured by separation methods (yielding asphaltenes, resins, saturates, etc.), molecular size distribution (GPC), wetting properties such as, contact angle, surface tension, and time. Of course many of these factors are interrelated and depend upon other variables such as temperature. Aggregate properties thought to be important in the absorption process are porosity or void fraction, pore size distribution (internal and/or external), surface areas, surface roughness, presence of fines, aggregate size distribution (gradation), and surface chemical composition of the aggregate. Other factors such as the presence or absence of water also influence the degree of absorption.

Because of the above factors it has been generally recognized that absorption of bitumen by aggregates plays an important role in the design of bituminous mixtures for highway construction (16,23,33,34,36,38,39,43,47,55,73).

The results of aggregate absorption may be any combination of the following (all may lead to premature failure of the paving mixture):

1. Incorrectly calculated percent air voids, voids filled with bitumen or voids in the compacted mineral aggregate (V.M.A.). Since one or more of these void parameters is being used as criteria for designing asphalt paving mixtures, incorrect calculation may result in mixtures lacking durability or stability.
2. A thin film, more susceptible to stress and weathering, resulting in ravelling and cracking.
3. Not enough binder, more susceptible to water action.
4. Changes in physical and chemical properties of the asphalt film possibly causing low temperature cracking of asphalt paving and premature age hardening.
5. Construction problems such as tender mixes and segregation.

One serious problem associated with the absorptive characteristics of aggregates is the fact that a wide variety of methods of evaluating or describing, quantitatively, the absorption of aggregate with respect to asphalt are currently available. Consequently no uniform specific requirements can be imposed, even though most highway engineers do realize that absorptive aggregates often cause trouble. Also absorption can occur over long time periods, e.g. months, after the pavement is placed. It is difficult to predict this long term behavior using current laboratory test methods. A better understanding of the fundamentals of absorption behavior is required to predict absorption in the field from

laboratory measurements.

The most common method of attempting to deal with absorptive aggregates is to increase the asphalt content of the mix; this solves the problem sometimes, but not always. If the absorption is fast enough and is essentially completed during the mix design procedure, this solution is adequate. If, on the other hand, the absorption is slow, the need for additional asphalt content will not be detected in the mix design, resulting in mixes which eventually have too little asphalt, and fail. Adding extra asphalt to the slow absorbing aggregate also causes problems because the mix is over-asphalted when placed and will be tender and difficult to compact. The mix may also flush initially in the pavement.

Finally, perhaps even more important, is the problem of aggregate supplies and efficient use of available aggregates. The aggregate supplies in many parts of the country are limited, yet the highway program is constantly expanding -- it may not be long before local supplies of high quality aggregates are exhausted. The use of poor quality absorptive aggregates has been unavoidable at times in the past and certainly will be in the future. However, few techniques or methods have been developed that will provide comprehensive information on the use of absorptive aggregates in bituminous mixtures.

## **Objective/Purpose**

This literature review was conducted to determine the current state of knowledge with respect to:

1. How aggregate properties influence absorption and how absorption can best be determined for practical use.
2. How asphalt properties influence absorption.
3. Methods for recovery of nonabsorbed asphalt to ascertain the extent and importance of selective absorption.
4. Methods which can effectively be utilized to minimize the adverse effects of absorption on pavement performance.

## Scope

This review is divided into five sections which cover the following topics:

1. Effect of aggregate properties on absorption.
2. Effect of asphalt properties on absorption.
3. Methods for determining asphalt absorption.
4. Methods for characterizing selective absorption.
5. Methods of treatment to minimize asphalt absorption.

### 1. Effect of Aggregate Properties on Absorption

The most important aggregate properties that affect asphalt absorption into aggregates are pore size distribution, chemical and mineral composition of the aggregate, surface roughness, and wetting properties (15,16,23,25,26). The pore characteristics of aggregates can be determined using a variety of methods such as mercury porosimetry (27,28, 30,31), water absorption (4), ASTM C-127 and C-128 for specific gravity (and absorption of coarse and fine aggregates, BET analysis (3), and optical methods (49). The effective porosity has been found to have fair correlation with absorption and to indicate the general trend, although the exact pore size distribution should be considered for a more accurate evaluation of absorption. That is, the rate and extent of asphalt absorption will depend not only on the total void fraction but also on the actual pore sizes in the aggregate, due to capillary pressure and viscosity effects. Absorption effects are also co-mingled with surface adsorption in many cases, because much of the surface area in porous aggregates lies within the pore space, rather than on the outer geometrical area. Generally, immersion methods using water and other liquids are found to be only rough indicators of the maximum amount of absorption. In this regard, widely varying "rules of thumb" have been developed by engineers in mix design when allowing for absorption, being roughly equal to 50% to 75% the water absorption value; however many investigators do not subscribe to such rules and believe there is no general correlation between water absorption and asphalt absorption (23,25,41).

The importance of porosity and pore size distribution has been realized and studied extensively in the case of concrete aggregates (4,27,28). However, there has been little published data on the study of the pore-size characteristics of asphalt aggregates and their relationship to absorption other than by Lee and his co-workers (15,16,23), and the more recent work of Hoque and Cabrera (68).



Probably the most common method used in casual investigation of aggregate porosity is to measure the water absorption and assume the volume of water absorbed equals the pore volume, that is, to assume complete saturation. The ASTM test for specific gravity and absorption of coarse aggregate (C-127) is the method for absorption of coarse aggregates and is straightforward except for the question of surface drying. The surface moisture must be dried from the wet aggregate without removing any water from the pores, either by evaporating or by capillary attraction into the cloth used. The result may be termed water permeable or 24 hour soaked porosity. Dinkle (4) obtained vacuum saturated porosity, and mercury intrusion porosity as well. One main point with respect to the liquid absorption method as a measurement of porosity is that one usually will not get complete saturation unless boiling or vacuum is used because some gas (air) remains trapped in the pores (2,64,65).

A common and effective method to determine the total porosity is to determine the bulk density and true density and to calculate the porosity by the ratio of the two. Bulk density can be determined by dimensional measurements if the samples are regular geometric shapes like cored cylinders or by the buoyancy method as per procedure given ASTM C-127, again involving uncertainties of surface drying. This can also be determined by displacement of some non-penetrating liquid, such as mercury. Caution must be used here since some aggregate pores are large enough to absorb mercury even at atmospheric pressure or below. The sample may also be coated with paraffin to prevent absorption of water and necessary corrections for the coating applied. This paraffin coating method could only be applied to a single rock; it would not be practical on a loose aggregate sample. The basic principles involved would be the same as when paraffin coating is used to determine the bulk specific gravity of a compacted mix. The true (solid) density of an aggregate can be determined with a volume change apparatus after powdering the sample and using a gas as the pycnometric fluid. It is assumed that no impermeable pores are left after the sample is powdered (54). Washburn and Bunting (54) employed the gas expansion method based on Boyle's law to determine porosity. Isolated voids will be counted as solids and thus the method measures effective porosity if the sample is not powdered. Beeson (1) modified the apparatus and most of the commercially available equipment is based on this principle. Most of these methods are reviewed by Collins (64).

Porosity can be determined visually on a polished or thin section of aggregate by measuring void area (pore space) microscopically by various camera-lucida or photomicrographic methods. Sweet (49) has used such optical methods on aggregates.

Dolch (5) determined the effective porosity of limestone aggregates with the McLeod gauge porosimeter developed by Washburn and Bunting. The method gives a value for the effective void volume by causing the head to be lowered on a dry sample while it is

immersed in mercury. The air in voids expands and leaves the sample and is then measured volumetrically at atmospheric pressure. If the bulk volume of the sample is obtained by the use of shaped pieces, porosity can be calculated.

The method most frequently used is that of mercury injection into the pore system. Washburn (53) was the first to suggest the use of mercury in determining the pore-size distribution of porous solids assuming a model based on a system of circular capillaries. Ritter and Drake (46) put Washburn's conception to practical use and developed the apparatus measuring the penetration of mercury into pores down to 200 Å in diameter at 10,000 psi covering the so-called macropore range. The apparatus generally referred to as a mercury porosimeter has been described by Purcell (40). A modern mercury penetration porosimeter can graph ambient to 60,000 psi in as little as ten minutes, instead of several hours required by earlier porosimeters, and measure pore sizes as small as 18 Å. Graphic output of data for a number of functional relationships is available from each analysis, included are  $dV/dP$ , the first derivative (slope) of volume versus pressure;  $Dv(r)$ , change in volume per unit interval of pore radius;  $Ds(r)$ , change in surface area per unit interval of pore radius; and  $S$ , total surface area. A phase transition porosimeter was developed at Iowa State University in 1986 which is capable of measuring the radii of pores from 10 Å to 5,000 Å. This instrument is unique in that it measures the radii of pore bodies as well as the radii of pore necks.

Hoque and Cabrera (68) found a reasonable correlation between mercury porosimeter pore size distribution and asphalt absorption for pores larger than 0.05 micron in radius. They stated that the smaller pores did not appear to absorb bitumen, but are penetrated by water. This was probably attributable to the higher viscosity of the bitumen as compared to water. Thus the pore size distribution is one factor causing a difference in water and asphalt absorption.

Conclusions from previous studies on asphalt absorption as affected by aggregate properties can be summarized as follows:

- Most aggregates absorb asphalt to some degree. The absorption of asphalt by aggregates may affect proper mixture design and pavement failure due to delayed absorption.
- Asphalt absorption is directly related to the porosity of an aggregate. If there is no porosity, there will be no absorption. The total porosity is an indicator for maximum possible absorption, but the actual pore size (permeability) determines the rate of absorption and the maximum may never be reached in practice.

- Pore-size distribution of an aggregate as it relates to permeability has a direct effect on the nature and degree of asphalt absorption. Good correlation was found between percent porosity larger than 1 micron and asphalt absorption for crushed aggregates and between percent porosity larger than 0.1 microns and asphalt absorption for rock cores. In general, asphalt absorption increases as these porosities increase. The higher the aggregate permeability, the greater the rate of asphalt absorption.
- Aggregate particle size and shape has an effect on the rate of absorption, with smaller aggregates being filled at a faster rate, other factors being equal.
- The presence of air and/or moisture in the pores has an effect on the rate of absorption (71,72).
- Wetting properties of the aggregate, e.g. contact angle, have an effect on the absorption process.

## 2. Effect of Asphalt Properties on Absorption

The absorption of asphalt into the pores of aggregate is driven by capillary pressure, the same force which makes water rise in a capillary tube. Equations governing the rate of rise in a capillary tube and associated phenomena have been given by Bikerman (59). More recent treatments of capillary rise have been made by Batten (62) and Good and Zin (63). Absorption into a porous aggregate is complicated by the nonuniformity of the pore structure, as well as many other factors. Wade and co-workers (60,61) have studied the absorption of various hydrocarbons and water into evacuated porous glass (Vycor). These workers have shown that the rate of absorption on a volume basis into an evacuated porous sphere is given by

$$-1/3 A - 1/2 [(1 - A)^{2/3} - 1] = t_r \quad (1)$$

where A is the fraction of pore volume filled,  $0 \leq A \leq 1$ , and  $t_r$  is a reduced time given by:

$$t_r = \frac{Kpt}{R^2\mu} \quad (2)$$

k	=	Darcy's Law permeability
P	=	capillary pressure
t	=	time of absorption
R	=	radius of sphere
$\mu$	=	fluid viscosity

For a circular cylindrical pore of radius r

$$P = \frac{2\sigma \cos\theta}{r} \quad (3)$$

$$k = r^2/8 \quad (4)$$

Consideration of equations (1) to (4) shows that the absorption process is governed by the fluid surface tension ( $\sigma$ ), contact angle ( $\theta$ ), viscosity ( $\mu$ ), as well as the pore radius (r), and sphere size (R). Important aggregate properties are the particle size (R) and permeability (k). A completely systematic study of these various properties on asphalt absorption does not appear to have been made. It is difficult to measure contact angles and surface tension of asphalts against porous aggregates, although a few attempts have been made (67). Nevitt (36) showed that absorption was time dependent up to about 6 months and Kandhal and Koehler (66) measured the time dependence of absorption using the Rice method. Generally the amount of absorption increased with time. Several researchers (25, 29, 16) have generally found that the rate of absorption increases as the asphalt viscosity decreases. Ellis et al. (69) found that asphalt was absorbed into the pores during repeated temperature cycles from 50 to 135°F, and that the less viscous asphalt was absorbed to a greater extent. These results are in qualitative agreement with equations (1) - (4) presented above. The absorption of asphalt in the aggregate pores is further complicated by its non-Newtonian rheology at low temperatures and the fact that the pores initially contain air (or even some water). The absorption of asphalt, thus, must displace a second fluid, such as air. Problems of this type have been studied in the oil industry using concepts of capillary pressure and relative permeability (64, 65). Whether or not these concepts can be applied usefully to asphalt absorption remains to be seen. Such application does not appear to have been attempted up to this time.

Consideration of the foregoing leads to the following conclusions regarding the effect of asphalt properties on absorption.

- Qualitatively speaking, the amount of absorption is directly proportional to the capillary pressure and time and inversely proportional to the viscosity.

- The amount of absorption was generally found to be less with increased asphalt viscosity (16,25,29). In this regard, the temperature of absorption is a critical variable.
- At this time, no unified, comprehensive quantitative theory is available for predicting or correlating the effect of asphalt and aggregate properties on absorption.
- Asphalt absorption is time dependent (23, 36, 66). Percent absorption increases with time at decreasing rate. In Iowa State studies, major adsorption occurred during the first 10-30 days and the absorption leveled off at about three months (23). The ratios between 100 days absorption and one day absorption varied from 1.15-3.15 for aggregates studied. A regression equation (significant at the 1% level) was established between absorption at one day and at 100 days.

### 3. Methods for Determining Asphalt Absorption

A number of investigators have attempted to evaluate aggregate absorption with kerosene and to correct for the amount of asphalt absorbed by aggregates in mixture design. In 1942, Hveem (14) devised his Centrifuge Kerosene Equivalent (CKE) test. The CKE, which is the quantity of kerosene absorbed by 100 g of aggregate under a specific set of soaking and centrifuging, has been shown to be a function of the surface area and absorptive capacity of the aggregate and has been used as a part of the design of the bituminous mixtures by Hveem's method. Lohn (32), using a method similar to that proposed by Hveem, was able to correlate bitumen absorption of an aggregate with kerosene absorption. He also studied the effects of saturation time, centrifuge time, and centrifuge force on kerosene retention and finally adopted a test procedure of ten minutes soaking and eight minutes centrifuging at a force of 400 times gravity. Donaldson et al. (6) proposed some modifications to the Hveem's CKE method, namely by increasing the soaking time to 30 minutes and by testing a nonabsorptive aggregate of the same gradation as the aggregate in question for purposes of comparison. It was believed that kerosene was a more representative absorptive agent than water in evaluating asphalt binders as it has wetting properties similar to the latter (20,21). Cechetini (58) has developed a modified CKE test procedure which can determine surface areas and absorption qualities of crushed aggregate mixes. By using the surface area determined and absorption data together with the data characterizing the asphalt to be used, the amount of asphalt needed for a particular aggregate can be determined by the use of a given nomograph. Surface areas and absorptions of the aggregate are determined from gains in weight following the kerosene immersion and centrifuging for

2 min at 400 times gravity.

Some investigators also evaluated the absorptive capacity of coarse aggregate by oils (19,29). Generally, as noted above, because of differences in viscosity and surface wetting properties these values do not agree with those obtained using asphalt cements and adjustments must be made for their usage.

ASTM Standards C-127 and C-128 outline methods for determining water absorption and bulk specific gravity of aggregates. These standards call for immersion of material in water for 24 hours, followed by drying until the saturated surface-dry state is attained. Coarse aggregates are rolled in an absorbent cloth until all visible water films are gone. Some operators judge this condition by observing the shine contributed by the water film while others judge by observing a slight color change in the aggregate.

Fine aggregates are spread on a pan and exposed to a gentle current of warm air until a free flowing condition is reached. The aggregate is then lightly tamped into a conical mold. If the cone stands when the mold is removed, the fine aggregate is assumed to carry moisture on its surface and it is dried further. When the cone just begins to slump upon removal of the mold, it is assumed to be in a saturated surface-dry state.

For natural, well graded fine aggregates, the saturated surface-dry condition is usually reproducible. However, the end point is more erratic for crushed fine aggregates because the angularity of the particles does not permit a definite slump condition as do the rounded surfaces of natural sands. Besides this, the higher percentage of material passing the No. 100 sieve also poses a problem in achieving slump condition.

One critical element in improving the accuracy of the bulk specific gravity of the aggregate,  $G_{sb}$ , is the reproducible determination of the saturated surface-dry condition. Various attempts have been made in the past to pinpoint the saturated surface-dry condition of the aggregates to improve the reproducibility of the bulk specific gravity test results. These include Howard's glass jar method (12,37), Martin's wet and dry bulb temperature method (33), Saxer's absorption time curve procedure (48) and Hughes' and Bahramian's saturated air drying method (13). However, the various modifications either offer little improvement or are too elaborate to be practical in the field or average laboratory.

A study was conducted at Iowa State University to develop new, simple, and more reproducible methods to determine the bulk specific gravity or the saturated surface-dry condition for granular materials. As a result, a new chemical indicator method was developed to determine the saturated surface-dry condition, and a glass mercury pycnometer was designed to determine bulk specific gravity of aggregates larger than the

No. 100 sieve size (16,23). A third method for determination of bulk specific gravity and water absorption was developed during absorption-time studies. The procedure involves the plotting of the weight-in-water versus time graph and extrapolating the weight of the aggregate sample in water at time zero ( $C_0$ ). The equations for specific gravity and absorption were:

$$\text{Apparent specific gravity (Gsa)} = A / (A - C)$$

$$\text{Bulk specific gravity (Gsb)} = A / (A - C_0)$$

$$\text{Absorption, \%} = 100 (C - C_0) / A$$

where

- A = oven-dry weight of sample in air, g.
- C = weight of sample in water after 24 hr immersion, g.
- $C_0$  = weight of sample in water at time zero, g. (from weight-in-water vs time)

Bulk specific gravity and absorption can be determined this way without trying to achieve the saturated surface-dry condition of the aggregate, which, for certain material, the reproducibility can be very poor. These methods offer promise, but are not yet ready for standard practice.

Reigel (41), in 1936, described a method to determine the relative absorption of coarse aggregate for water and for liquid bituminous material using the water displacement method. A sample of aggregate is first placed in water for 24 hours. It is then weighed in water, dried and weighed in air. Liquid bituminous material equal to 8% by weight of the sample is added and the mixture is heated to 140°F for up to 20 days. The excess liquid bituminous material is then removed from each piece of aggregate by wiping. The mixture of aggregate and liquid bituminous material is then weighed in air and also under water. The amount of liquid bituminous material absorbed is calculated by manipulating the four weight values. Liquid bituminous material absorption showed very small increase after 20 days; the amount absorbed at this time varied from 62% to 92% of water absorption at 24 hours. It was recommended that liquid bituminous material absorption be estimated as 75% of water absorption.

In 1942, Goshorn and Williams (9) developed an immersion method in which the coarse aggregate is immersed in a tared wire basket in asphalt at 275°F for three hours. The basket is removed from the asphalt and suspended in an oven for 10 min. to drain off excess asphalt. The basket with coated aggregate is cooled to room temperature and weighed in the air and in water at 77°F. The percent asphalt absorbed is calculated based on straightforward volume-weight relationships of aggregate, asphalt and effective

volume of coated aggregate.

Since the aggregate is in contact with an unlimited supply of asphalt at relatively low viscosity for extended periods of time, the absorption is much higher than would be expected in a bituminous mixture and is time dependent. However, the values can be taken as the absorptive potentials of aggregates used for bituminous mixtures. Accuracy of this test is limited by homogeneity of the aggregate and accuracy of the bulk specific gravity and water absorption.

Rice (43,44) proposed a procedure to determine the maximum specific gravity of the mixture by using volumetric flasks. The absorption of asphalt by aggregate in a mixture can be calculated by applying the concept of effective specific gravity of aggregates,  $G_{se}$ , if the theoretical maximum specific gravity of the mixture (after Rice's method), the asphalt content, and the bulk specific gravity of the aggregate used in the mixture are known (35). This method has been standardized as ASTM D-2041. One serious limitation of this method is that aggregate particles must be thoroughly coated; failure to do this may give erratic data because of possible water absorption during saturation. Also, the accuracy of this method depends on the accuracy of bulk specific gravity of aggregate. However, in this method, results can be obtained from actual mixtures. Time-absorption relationships can be determined by repeated determination of the theoretical maximum specific gravity of the same mixture over time and calculation of the effective specific gravity  $G_{se}$ . Warden (70) developed a large size vacuum pycnometer (volume = 13.5 liters) for performing the Rice method with 6000 g of mixture.

Effective specific gravity of aggregate, termed "indicated specific gravity", was determined by a high pressure test (5000 psi) by Larsen (22). Absorption of bitumen by aggregate was calculated as percent of the volume of the aggregate. The asphalt absorption, as percent of water absorption, ranged from 26% to 88% averaging 64%. No linear relationship existed between water absorption and asphalt absorption.

Asphalt absorption can be calculated from the effective specific gravity of the aggregate  $G_{se}$ , determined from the theoretical maximum specific gravity of bituminous mixtures,  $G_{mm}$ , by ASTM D-2041. This method is simple, easy to operate and can be performed by a relatively inexperienced technician, and the state of asphalt in the asphalt mixture is the most representative to that existing in the field. However, since the accuracy and reproducibilities of the bulk specific gravity of the aggregate,  $G_{sb}$ , and  $G_{mm}$ , limit the results, some refinement of ASTM C-127, C-128 and ASTM D-2041 may be advantageous. Using the acceptable ranges of two results by a single operator specified by ASTM of 0.011 for  $G_{mm}$  and 0.025 for  $G_{sb}$ , for a range of asphalt contents between 5% to 7%, the calculated asphalt absorption can vary up to 1.1% by weight and 2.9% by



volume of the aggregate.

The refinement of ASTM D-2041 is underway in ASTM Committee D04.21, Factors such as vacuum, vacuum gauge arrangement, time of evacuation, temperature, location of water vapor trap, etc. can possibly be varied to improve the method. Consideration should also be given to the calculation of asphalt absorption based on volume rather than weight as is now practiced because of its effect on the voids properties on the mix design and compaction control and because of the possibility of selective absorption of asphalt by aggregates. If the lighter constituents of the asphalts are selectively absorbed by the aggregate, as is commonly believed, absorption by volume can better reflect the extent of absorption than by weight. It may also be advantageous to determine G<sub>mm</sub> repeatedly at certain time intervals or after the mixes are artificially aged, e.g. in an oven at 140°F, to establish the absorption-time curve and the long-term or equilibrium absorption. One study based on this scheme was performed at Iowa State University for up to 1000 days. Calculated asphalt absorption showed increased absorption from 0.4% at one day at a decreased rate to one year at 3.7% and stabilized at 3.9% (23,26).

The U.S. Corps of Engineers (45,51) developed and used bulk-impregnated specific gravity in the design and control of bituminous paving mixtures. The bulk-impregnated specific gravity (G<sub>bi</sub>) can be defined as the ratio of the weight in air of a given volume of a permeable aggregate (including solids, impermeable pores, and pores normally permeable to water but which are variably permeable to bitumen) at a stated temperature (77°F) to the weight of an equal volume of water minus the weight of the volume of bitumen absorbed by pores permeable to it. The bulk-impregnated specific gravity, G<sub>bi</sub>, equals bulk specific gravity, G<sub>sb</sub>, if no asphalt is absorbed. On the other hand, if absorbed bitumen is equal to water absorption; its G<sub>bi</sub> is equal to the apparent specific gravity, G<sub>sa</sub>. Thus, bulk-impregnated specific gravity is a function of the ratio of bitumen to water absorption, which varies widely but follows a definite pattern with different types of aggregate. The percent absorption by weight of aggregate may be found from G<sub>bi</sub> using:

$$A = \left( \frac{1}{G_{sb}} - \frac{1}{G_{bi}} \right) \times g_a \times 100$$

where  $g_a$  is the specific gravity of the asphalt. This is suitable for general use with all aggregates used in bituminous mixtures and is particularly suited for use with porous aggregates. McLeod (34) has recommended the use of this method to determine the maximum, or upper limit, absorption for aggregate. Lee (26) compared the data with those obtained by other methods and concluded that these absorption values were by no means the absolute maximum absorption values for a particular aggregate, but can be looked upon as the "realistic" maximum absorption values aggregates will have in

bituminous mixtures.

The absorption capacity of aggregates might possibly be determined by colorimetric analysis with a photometer, using either asphalt solution or dye solution (e.g. methylene blue and safranin T) and calibration curves (26). The basic principle is that the amount of light absorbed by a given solution is proportional to the intensity of the incident light and to the concentration of the absorbing species in the path of the light beam. When a specific amount of asphalt solution of known concentration is absorbed by aggregates, the reduction in asphalt concentration due to absorption of the asphalt by the aggregate results in a change in the light transmittal of the solution. The method is simple and rapid and the equipment is standard. Limitations are: adsorbed moisture content of aggregate affects the repeatability or accuracy of this method and also only solutions of dilute concentrations can be used. Moreover the mechanisms of absorption of asphalt in solution and absorption of asphalt in bulk could be different. Finally, this method may be more indicative of adsorption, as opposed to absorption.

The Methylene Blue Test has been used in Europe in recent years to indirectly measure the absorption/adsorption, surface area, cationic exchange capacity, soundness and overall clay characteristics of aggregates and may hold potential in the study of aggregate absorption, especially fine aggregate. The Methylene Blue Test, first developed in France (50), uses methylene blue to quantify the adsorption/absorption of an aggregate. A sample of 1.0g finely ground aggregate is dispersed in 30cc of distilled water. The dispersion is continuously stirred throughout the titration with a methylene blue reagent, added stepwise in 0.5cc aliquots. After each addition one small drop of the dispersion is removed and placed on a filter paper. Initially a circle of dust is formed surrounded by an outer ring of clear water. The end point is reached when a permanent light blue coloration is observed in this ring of water. The blue value is calculated as the quantity of reagent used to reach the end point divided by the weight of the aggregate sample.

Cross-sectional measurements can be employed to evaluate asphalt absorption (26). The mixture is prepared and compacted in a kneading compactor. It is cut in halves by a diamond saw to expose the inner surface of the aggregates. The sample is then placed in an opaque projector and from its image projected on a screen, tracings are made of the external contours of the rock particles and lines of the deepest asphalt penetration are measured by a planimeter, and the percentage of asphalt absorption is calculated. This method is most suited for evaluation of selective absorption of asphalt by model aggregates of simple geometry such as flat plates or rock cylinders, especially in conjunction with a scanning electron microscope and automated image analyzer.

Conclusions from previous studies (14-17, 23-26) on asphalt absorption as affected by the

method of determination can be summarized as follows:

- No single standard method is currently utilized to evaluate, describe and specify the absorptive characteristic of an aggregate with respect to asphalt. There is a need to establish such a test.
- Indirect prediction of asphalt absorption by water, kerosene, or oil absorption is not always reliable.
- The bulk-impregnated specific gravity (Corps of Engineers) method can be used to determine the practical or realistic maximum asphalt absorption of an aggregate and has been recommended for evaluating and comparing potential aggregate sources; the Rice specific gravity method (ASTM D-2041) can be used to determine the practical or realistic minimum absorption of an aggregate and is recommended for mixture design.
- The accurate determination of asphalt absorption in both methods depends upon the realistic and accurate determination of bulk specific gravity of aggregate.
- The current ASTM methods for the determination of the bulk specific gravity of aggregate have some problems, especially the method for measuring this property on the fine material (-#4 material). The bulk specific gravity is used in pavement mix designs, and the absorption data calculated from the bulk specific gravity are of value for developing mix designs; however, the precision of the test procedures leave much to be desired.
- Three methods for determining bulk specific gravity of aggregates (chemical indicator method, mercury pycnometer or penetration method and time-weight in water curve method) were developed during Iowa State investigations (23). However, all three need more work and further refinement before possible formal adoption.
- A photo-colorimetric technique can be used to evaluate the asphalt absorption of aggregates of all sizes and gradings (23).

All potential methods for determining asphalt absorption such as immersion method, bulk impregnated method and modified Rice method will be evaluated using SHRP MRL aggregates and asphalts so that a realistic and accurate method can be recommended.

#### 4. Methods for Characterizing Selective Absorption

When asphalt is absorbed into the pores of an absorptive aggregate, there may be preferential compositional changes in that the absorbed asphalt inside the pores has a different composition than the nonabsorbed, or effective, asphalt film acting to bind the aggregate together. In turn, because of this selective absorption, the absorbed and nonabsorbed asphalt will have compositions different from the original bulk asphalt cement. In other words, due to possible selective absorption, the effective asphalt film may have different rheological, physical, chemical, and aging properties than the original bulk asphalt cement added during mixture preparation.

While mix design procedures used by some agencies make allowance for asphalt absorption based on the Rice test which determines the maximum specific gravity of the mixture, there is a notable absence of specifications and/or mix design methods for dealing with the possible selective nature of asphalt absorption. Selective asphalt absorption, that results in an effective asphalt film different in composition from the original bulk asphalt cement added to the mix, can be caused by several factors. In highly porous aggregates, a large fraction of the aggregate surface areas resides in the pore space, and selective adsorption of, for example, polar species, will lead to a state in which the absorbed asphalt has a different composition than the effective asphalt film. That is, the more polar, highly adsorbed species will be preferentially removed from the effective asphalt film and will reside in the absorbed asphalt inside the pores. Depending on the pore sizes in the aggregate, selective diffusion and partitioning effects based on relative molecule to pore sizes also can lead to segregation and compositional differences between the absorbed and nonabsorbed (effective) asphalt. For example, the larger molecular species, e.g. asphaltenes, may be preferentially left behind in the asphalt film while the smaller molecular sizes, e.g. maltenes, are preferentially absorbed into the pore volume. The net result of such selective absorption is that the actual effective asphalt film binding the aggregate together has a composition and properties, which are different from the original bulk asphalt cement. If selective absorption occurs very quickly, then it will be detected during the mix design, e.g. by higher than expected Marshall stability, etc., and this factor can possibly be compensated for by using a lower viscosity grade asphalt. However, if the selective absorption occurs over several days or months, then the mix design could provide misleading results, and premature pavement distress could occur at a later date.

Most previous studies concerning asphalt absorption have been concerned with only the overall amount of absorption and have not addressed its potentially selective nature. While the amount of asphalt absorption is certainly very important in that failure to properly account for this factor can lead to overly rich or lean mixtures, the selective aspects of absorption may have more significance in determining aging and weathering

characteristics of the effective asphalt film and their effect on long term pavement durability. Thus, while several workers have studied the phenomena of asphalt-aggregate absorption and recognized the potential deleterious effects of absorption on asphalt pavement performance, most of the studies have involved physical methods to study pore size, pore size distributions for selected aggregates, and asphalt absorption capacities of selected aggregates. Essentially, little information is available concerning the manner in which the bulk chemical composition of an asphalt is changed by aggregate absorption. Also, little information is available concerning the kinds of compound types that tend to be absorbed by various absorptive aggregates. Although little chemical information is available, this type of information is very important to understanding and solving the problems related to absorption.

In one study Petersen (37) has addressed the selective nature of absorption. He found that the lighter, less viscous, fraction of the asphalt tended to be absorbed preferentially into the pores while the heavier asphaltene rich fraction was left behind in the asphalt film. Such a segregative nature of the absorption process can produce a film more susceptible to premature aging related failure, upset structuring characteristics, and lead to early signs of pavement distress.

The problem of selective absorption may be further aggravated by porous aggregates in which most of the surface area is inside the pores, rather than the outer geometrical surface of the rock. In this case, adsorption may also play a role in contributing to compositional differences in the absorbed and nonabsorbed asphalt. Because of the changes in properties of the nonabsorbed asphalt, the mixture may not behave as expected and, in fact, may deteriorate prematurely and even fail much sooner than would be expected had selective absorption not occurred. Selective absorption of asphalt components with consequent changes in film properties can be a cause of accelerated aging, premature pavement distress due to ravelling, low temperature cracking and/or water damage.

Petersen and his co-workers (38) also found that certain compounds were selectively adsorbed at the asphalt-aggregate interface. It is not known whether these groups will be selectively absorbed in the pores of the aggregate. Thelen (56) also noted that an increase in asphaltene concentration in the film would produce a "drier" and harder film.

In his review of absorption, Kneller (18) found that Lettier et al. (29) described two problems possibly caused by selective absorption of asphalt by porous aggregates. Difficulties in mixing bitumen with the aggregates were noted where the stones appeared to be difficult to dry and the bitumen tended to "burn" (producing a dry brown, dull mix). Furthermore, the mix did not stick together as well. Secondly, he observed ravelling in a section of road, when only limited local traffic was permitted on the road.

Lettier attributed both problems to asphalt absorption, but he emphasized that the second problem was more related to selective absorption of the diluent by the porous aggregates.

Lee, in 1968, recognized the need to develop a method to recover the non-absorbed asphalt in order to study the changes in asphalt due to absorption (26). Thirteen mixtures were prepared using an absorptive aggregate and a nonabsorptive aggregate. Two attempts were made to recover separately the asphalt films that remained outside the aggregate pores and the asphalt inside the aggregate pores by cold extraction using a Rotarex centrifuge and analytical grade trichlorethylene as solvent. In two of these mixtures asphalts were extracted and recovered into three fractions. Loose mixtures were first soaked in solvent for three minutes and centrifuged, more solvent was added and soaked for additional five minutes, centrifuged, and finally more solvent was added and soaked for additional 45 minutes and centrifuged. Three fractions of solutions were collected and recovered separately by the Abson Method. The first fraction was arbitrarily considered to be from the asphalt films that remained outside the aggregate pores, the third fraction was considered to be from the asphalt that penetrated deeply into the aggregate pores, and the second fraction was from asphalt of moderate penetration. Asphalts were recovered in two fractions in six of the mixtures by first soaking in solvent for five minutes and recovering, and then soaking an additional 50 minutes and recovering. The first fraction was taken as nonabsorbed asphalt films, and the second fraction was the absorbed portion of asphalt. These recovery methods are somewhat approximate and may not always differentiate between strongly adsorbed asphalt components versus deeply absorbed components. Asphalts from the remaining five mixtures were recovered in one operation following the standard ASTM procedure. Changes in asphalts were measured by viscosity at 77°F and percent asphaltenes. No precision data are available for Dr. Lee's work on the successive extraction method. It is doubtful that the same soaking time will produce similar results with different mixtures.

With limited data and without field correlation, it was difficult to assess the effects of absorption in terms of asphalt durability in the field. Nevertheless, the following conclusions were made:

1. Selective absorption took place with both aggregates. This was evidenced by the lower asphaltene content and viscosity of the asphalt component inside pores compared with the films outside the aggregate pores. These differences appeared to be more pronounced for the absorptive aggregate.
2. Results can be misleading when extracting asphalt from highly absorptive

aggregate by the standard ASTM procedure in evaluating asphalt film characteristics. The actual films may be much harder or softer than the bulk asphalt recovered, depending upon the nature of absorption.

3. Extraction of asphalt into fractions by successive soaking can be used to identify the degree and nature of absorption. However, the method cannot be used routinely and is not precise because the asphalts do not really come off in layers.

Additional work is needed to identify the mechanisms, extent and implications of the effects of selective asphalt absorption with porous aggregates. As part of the SHRP research, several potential experimental techniques for recovery of nonabsorbed and possibly, absorbed asphalt fractions separately will be attempted. The methods to be explored include (1) physical (mechanical) separation, (2) cryogenic separation, (3) low temperature ashing, and (4) solvent extraction.

## **5. Methods to Minimize Adverse Effects of Absorption**

In the previous sections we have reviewed various aspects of the absorption effects present when asphalt is utilized with porous aggregates. We can summarize by saying that there is a general desire on the part of engineers to minimize the adverse effects of absorption including (a) loss of asphalt from the effective asphalt film binding the aggregate together and (b) change in properties of the effective asphalt film as compared to the original asphalt cement added to the mix. This latter factor is caused by selective absorption as discussed by Thelen (56) and Lee (26) in their investigations. The selective nature of absorption can result in effective asphalt films which are more susceptible to the action of water, and oxidative weathering, which can result in ravelling and cracking of the resulting pavement. Overcoming these detrimental factors due to absorptive aggregates would allow better utilization of more readily available, perhaps marginal quality, aggregates, which might be moved to the hot mix plant more quickly and economically than higher quality aggregates located further away. In certain regions of the U.S., high quality aggregates are in very limited supply as noted by Witzark et al. (57).

Because of the current oil glut, asphalt prices have moved lower in recent years and, therefore, the first concern (that is, the loss of asphalt into the pore spaces) is not as economically significant as it was ten years ago. However the change in effective asphalt film properties due to selective absorption and its consequences for highway performance remains a most critical phenomenon. In addition, the asphalt price will undoubtedly rise in future years as the current petroleum imbalance corrects itself, and

the asphalt lost due to absorption in the pore spaces will become of increasing economic importance.

Because of the aforementioned factors there is a need and desire to develop methodology for preventing or minimizing the adverse effects due to asphalt absorption into aggregate pores. Besides the work of Lee, et al. (7, 8, 23, 24), there have not been many investigations along these lines, although the very recent work by Kneller and Elias (18) is notable. These investigators showed that the particles of a marginal aggregate could be coated with an innocuous substance to seal the surface pores and decrease absorption values, as measured by the modified ASTM C-127 Absorption Test. Of seventeen potential sealants tested, nine were found to decrease the water absorption quite significantly. The sealants found to be most effective were silicones, siliconates, siloxanes, polysilicates, and colloidal silicas. SEM techniques were used to reveal the details of the surface coatings obtained by treatment of the porous aggregates by these sealants. One of the most effective sealants, sodium methyl silicate, gave a 47 percent reduction in absorption value as determined by ASTM C-127 which translated into an asphalt savings of \$1.20 per ton of mix based on asphalt at \$105/ton. These sealant effects still need to be verified using actual asphalt, rather than water, absorption tests. Also the costs of equipment/modifications for applying the sealant need to be evaluated.

In developing a coating or sealant for reducing adverse absorption effects it is important to require that the sealant not cause any adverse effects itself, such as increasing the propensity toward moisture induced damage (stripping) and that it is safe to use. Only processes which increase pavement durability are acceptable candidates for minimizing absorption. As we have noted earlier, asphalt absorption can be a two-edged sword, in that it has both positive and negative effects. In addition to the adverse effects as delineated above, the absorption of asphalt into aggregate pores can increase the physical interlocking of the binder and the aggregate resulting in a higher pavement strength. Porous aggregates can also have beneficial effects with regard to skid resistance as noted by Kneller and Elias (18). These beneficial effects with respect to absorptive aggregates must be considered when developing methods to minimize adverse absorption effects.

Several techniques have been suggested by Lee (23) to reduce both water and asphalt absorption of aggregates and to minimize the adverse effects of absorption. Some of these techniques are:

- (1) Modification of aggregates by chemicals to seal pores.
- (2) Treatment of aggregates to change the surface characteristics of these aggregates hereby the aggregates would become hydrophobic.



- (3) Treatment of aggregates by cut-back or "flux" to satisfy aggregate absorption.
- (4) Modification of asphalts.
- (5) "Two-shot" methods of mixing.
- (6) Modified mixing temperature and compaction procedure.

One approach that is sometimes used when dealing with absorptive aggregates is to precoat these aggregates with cutback asphalt or emulsion. The aggregates are first sprayed with a dilute solution of these liquid bituminous materials which increase in viscosity quite rapidly on application to aggregates. A relatively hard asphalt precipitates in the surface pores and further entry of asphalt is eliminated. Asphalt mixes are then made using the required amount of asphalt cement. This is called the "Two-Shot Method." However, it should be realized that the production costs will be higher compared to one-shot mixing. Some environmental aspects also need to be addressed if cutback asphalt is used.

Five classes of more than 30 agents were investigated by Lee and his co-workers (7,8,23,24): (a) industrial wastes, (b) polymers, (c) synthetic resins, (d) inorganic chemicals, and (e) bituminous materials, each of which had advantages and disadvantages shown later in terms of effectiveness in water absorption reduction, asphalt absorption reduction, and relative costs.

The materials used in these treatments achieved absorption reduction by: (a) changing the surface characteristics of the aggregate, (b) sealing the aggregate pores, (c) filling the aggregate pores, or (d) combining any of the above.

Treated aggregates were tested for water absorption, asphalt absorption up to 135 days by the bulk-impregnated specific gravity method and the Rice specific gravity method, and heat stability at 500°F. The time-absorption curves of treated and untreated aggregate were determined. Approximately 30 chemicals were investigated. Aniline-furfural, rapid curing cutbacks, crude tar, methyl methacrylate, PVA, two synthetic resins and sulfur were found to be effective in reducing asphalt absorption. As much as 90% reduction in asphalt absorption was obtained. The differences in asphalt absorption between treated and untreated aggregates were greater at 100 days than at one day, that is, there was evidence of a possibly significant advantage of reducing delayed absorption because of treatments. Results of asphalt paving mixes with treated and untreated aggregates showed considerable savings in asphalt requirement of paving mixtures could be obtained through chemical treatment. Based on Marshall immersion compression

tests, a considerable increase in the percent retained strength of the asphalt concrete mixtures made with the chemically treated aggregates was obtained.

The following conclusions were drawn regarding the use of coatings and sealants to minimize absorption.

1. Several promising sealants/coatings for minimizing absorption of asphalt have been examined by previous workers.
2. The actual total cost and economic feasibility of applying sealants/coatings has yet to be determined.
3. Little knowledge about the long-term negative effects, if any, of sealants/coatings on pavement performance is available.

### Overall Summary

From a fundamental (rational) basis, additional research is needed to understand the influence of asphalt and aggregate properties on absorption. Only general trends, e.g. the absorption increases with time and lower viscosity, are now evident from past research. Some empirical correlations have been made, however, which may prove useful for correlating field data. One objective of the current SHRP research will be to obtain a better understanding of the way asphalt and aggregate properties influence absorption.

There are several field type tests which have been proposed for determining the amount of asphalt absorbed by aggregates. One objective of current SHRP research is to evaluate these tests and then to propose a new and/or modified test procedure, if needed, to more accurately represent the absorption behavior obtained in actual hot mix plants.

Little is known of selective absorption; however, it seems reasonable, based on known scientific principles, e.g. hindered diffusion of large molecules, that such a phenomenon will occur. Current SHRP research efforts seek to elucidate the presence or absence of this phenomenon and to ascertain its importance for road performance.

Several methods for minimizing asphalt absorption by porous aggregates have been proposed, mostly utilizing coatings and sealants of some type. While these treatments have been shown to reduce the amount of absorbed asphalt in laboratory tests, the technical and economic feasibility of these treatments in actual practice remains an open question at this time and will probably vary depending upon local circumstances.

Current SHRP research is attempting to answer some of these questions as well as find new techniques for minimizing absorption.

# References

1. Beeson, C.M., *The Kobe Porosimeter*, Am. Inst. of Mining, Metallurgical and Petrol. Engrs. Vol. 189, pp. 313-315, 1950.
2. Blanks, R.G., *Modern Concepts Applied to Concrete Aggregates*, Proc. ASCE, 75, pp. 441-446, 1949.
3. Brunauer, S., Emmett, P.H. and Teller, E., *Adsorption of Gases in Multimolecular Layers*, J. Am. Chem. Soc., Vol. 60, pp. 309-311, 1938.
4. Dinkle, R.E., Freezing Resistance in Mortar Related Pore Properties of Rock Particles, University of Maryland M.S. Thesis, 1966.
5. Dolch, W.L., *Studies of Limestone Aggregates by Fluid Flow Methods*, Proc. ASTM, Vol. 59, pp. 1204-1215, 1959.
6. Donaldson, J.A., Loomis, R.J., and Krchma, L.C., *The Measurement of Aggregate Absorption*, Proc. AAPT, Vol. 18, pp. 278-300, 1949.
7. Dutt, R.N., Upgrading Absorptive Aggregates by Chemical Treatments, M.S. Thesis, Iowa State University, Ames, Iowa, 1970.
8. Dutt, R.N. and D.Y. Lee, *Upgrading Absorptive Aggregates by Chemical Treatments*, Highway Research Record, No. 353, Highway Research Board, National Research Council, pp. 43-46, 1971.
9. Goshhorn, J.H. and William, F.M., *Absorption of Bituminous Materials by Aggregates*, Proc. AAPT, Vol. 13, pp. 41-51, 1942.
10. Hills, J.F. and Pettifer, G.S., *The Clay Mineral Content of Various Rock Types Compared with Mythylene Value*, J. Chem. Tech. and Biotech., 35A (4), pp. 168-180, 1985.
11. Hiltrop, C.L.R., Relation of Pore Size Distribution to the Petrography of Some Carbonate Rocks, Unpublished MS Thesis, ISU, 1958.
12. Howard, E.L., Discussion, Proc. Am. Concrete Inst., Vol. 54, p. 1215, 1958.

13. Hughes, B.P. and Bahramian, B., An Accurate Laboratory Test for Determining the Absorption of Aggregates, Materials Research Standards, 7, pp. 18-23, 1967.
14. Hveem, F.N., Use of the Centrifuge Kerosene Equivalent as Applied to Determine the Required Oil Content for Dense Graded Bituminous Mixtures, Proceedings AAPT, Vol. 13, pp. 9-40, 1942.
15. Kandhal, P.S., Asphalt Absorption as Related to Pore Characteristics of Aggregates, M.S. Thesis, Iowa State University, Ames, Iowa, 1969.
16. Kandhal, P.S. and Lee, D.Y., Asphalt Absorption as Related to Pore Characteristics of Aggregates, Highway Research Record No. 404, pp. 97-111, 1972.
17. Kandhal, P.S. and Lee, D.Y., An Evaluation of the Bulk Specific Gravity for Granular Materials, Highway Research Board, Highway Research Record No. 307, 1970.
18. Kneller, W.A. and Elias, H.M.R., Development of an Innocuous Coating to Decrease Absorption in Aggregates Used in Bituminous Pavement, Final Report for France Stone Fdn., Toledo, OH, Grant No. 213-556-3658, Department of Geology, University of Toledo, Toledo, OH, 1987.
19. Kraemer, P., Die Optimale Bindemitteldosierung der Bituminösen Mineralmassen im Heißeinbau. Bitumen - Teere- Asphalte- Peche und Verwandte Stoffe 2, pp. 48-56, 1966.
20. Krchma, L.C., Donaldson, J.A. and Loomis, The Measurement of Aggregate Absorption, Proc. AAPT, Vol. 16,353, 1947.
21. Krchma, L.C. and H.G. Nevitt, Absorption of Liquid Bituminous Cement by Aggregates, Proc. AAPT, Vol. 13,52, 1942.
22. Larsen, O., Development and Use of High Pressure Apparatus for Determining Voids in Compacted Bituminous Concrete Mixtures, Proc. AAPT, Vol. 6, pp. 282-317, 1957.
23. Lee, D.Y., Absorptive Aggregates in Asphalt Paving Mixtures, HR-142 Final Report, Iowa State University, Ames, Iowa, 1971.
24. Lee, D.Y., Modification of Asphalt and Asphalt Paving Mixtures by Sulfur Additives, Ind. Eng. Chem., Prod. Res. Dev., Vol, 14, No. 3, pp. 171-177, 1975.
25. Lee, D.Y., The Relationship Between Physical and Chemical Properties of Aggregates and Their Asphalt Absorption, Proc. AAPT, Vol. 38, pp. 242-275, 1969.
26. Lee, D.Y., Study of Absorptive Aggregates in Asphalt Paving Mixtures, HR-127 Final Report, Iowa State University, Ames, Iowa, 1968.
27. Lemish, J. et al., Relationship of Physical Properties of Some Iowa Aggregates to Durability of Concrete, HRB Bulletin 196, pp. 1-16, 1958.
28. Lemish, J. and Hiltrop, C.L., The Relationship of Pore-Size Distribution and Other Rock Properties to the Serviceability of Some Carbonate Rocks, HRB Bulletin 239, pp. 1-23, 1960.
29. Lettier, L.A., Fink, D.F., Wilson, N.B. and Fraley, F.F., Mechanism of Absorption of Bituminous Materials by Aggregates, Proc. AAPT, Vol. 18, pp. 278-300, 1949.

30. Lewis, D.W. and Dolch, W.L., *Porosity and Absorption*, ASTM STP No. 169, pp. 303-313, 1955.
31. Lewis, D.W. et al., *Porosity Determination and the Significance of Pore Characteristics of Aggregates*, Proc. ASTM, 53, pp. 949, 1953.
32. Lohn, R.N., *A Method to Determine Aggregate Absorption and Control of Bituminous - Aggregate Properties*, Proceedings AAPT, Vol. 15, pp. 188-197, 1942.
33. Martin, J.R., *Two Years of Highway Research at Oklahoma A&M*, Proc. AAPT, Vol. 19, pp. 41-54, 1950.
34. McLeod, N.W., *Selecting the Aggregate Specific Gravity for Bituminous Paving Mixtures*, Proceedings HRB, 36, pp. 282-317, 1957.
35. Mix Design Methods for Asphalt Concrete, The Asphalt Institute. Manual Series No. 2 (MS-2), pp. 65-78, May 1984.
36. Nevitt, H.G. and Krchma, L.C., *Absorption of Liquid Asphalt Cements by Aggregates*, Proc. AAPT, Vol. 13, pp. 52-68, 1942.
37. Pearson, J.C., *Simple Titration Method for Determining Absorption of Fine Aggregate*, Rock Products, Vol. 32, No. 10, pp. 64-65, May 11, 1929.
38. Petersen, J.C., *Chemical Composition of Asphalt as Related to Asphalt Durability: State of the Art*, Transportation Research Board 999, pp. 13-30, 1984.
39. Plancher, H., Dorrence, S.M. and Petersen, J.C., *Identification of Chemical Types in Asphalt Strongly Absorbed at the Asphalt-Aggregate Interface and Their Relative Displacement by Water*, Proceedings, AAPT, Vol. 46, 1977.
40. Purcell, W.R., *Capillary Pressures - Their Measurement Using Mercury and Calculation of Permeability Therefrom*, Trans. AIME, 186, pp. 39-48, 1949.
41. Reagel, F.V., *Absorption of Liquid Bituminous Material by Coarse Aggregate*, Highway Research Abstracts No. 40, HRB, pp. 5-6, 1937.
42. Rhoades, R. and Mielenz, R.C., *Petrographic and Mineralogic Characteristics of Aggregates*, ASTM STP. No. 83, pp. 20-47, 1948.
43. Rice, J.M., *Maximum Specific Gravity of Bituminous Mixtures by Vacuum Saturation Procedure*, ASTM. Special Technical Publication, 191, pp. 43-61, 1956.
44. Rice, J.M., *New Test Method for Direct Measurement of Maximum Density of Bituminous Mixtures*, Crushed Stone Journal, pp. 10-17, Sept. 1953.
45. Ricketts, W.C., Sprague, J.C., Tabb, D.D. and McRae, J.L., *An Evaluation of the Specific Gravity of Aggregates for Use in Bituminous Mixtures*, Proc. ASTM, Vol. 54, pp. 1246-1257, 1954.
46. Ritter, H.L. and Drake, L.C., *Pore Size Distribution in Porous Materials*, Ind. Eng. Chem. Anal. Ed., Vol. 17, pp. 782-786, 1945.

47. Ruth, B.E. and Potts, C.F., *Changes in Asphalt Concrete Mixture Properties as Affected by Absorption, Hardening, and Temperature*, Transportation Research Record, No. 515, Transportation Research Board, Washington, D.C., pp. 55-66, 1974.
48. Saxer, E.L., *A Direct Method of Determining Absorption and Specific Gravity of Aggregates*, Rick Products, Vol. 59, No. 5, pp. 78-79, May 1956.
49. Sweet, H.S., *Research on Concrete Durability as Affected by Coarse Aggregates*, Proc. ASTM, Vol. 48, pp. 988-1019, 1948.
50. Tran, N.L., *The Methylene Blue Test*, Bulletin de Liaison des Ponts et Chaussees, 107, pp. 130-135, 1980.
51. U.S. Corps of Engineers, *Investigation of the Penetration of Asphalt into Porous Aggregates as Related to and Affecting the Specific Gravity of the Aggregate*, Waterways Experiment Station, Corps of Engineers, Misc. paper No. 4-88, 1954.
52. Walker, R.D., Larson, T.D. and Cady, P.D., *Research Needs Relating to Performance of Aggregates in Highway Construction*, NCHRP Report 160, 1970.
53. Washburn, E.W., *Note on a Method of Determining the Distribution of Pore Size in a Porous Material*, Nat. Acad. Sci. Proc. 7, pp. 115-116, 1921.
54. Washburn, E.W. and Burtling, E.N., *The Determination of Porosity by the Method of Gas Expansion*, Am. Ceramic Soc. J., Vol. 5, pp. 112-116, 1922.
55. Zube, E., *Cracking of Asphalt Concrete Pavement Associated with Absorptive Aggregates*, Proceedings AAPT, Vol. 35, pp. 270-290, 1966.
56. Thelen, E. *Surface Energy and Adhesion Properties in Asphalt Aggregate Systems*, Highway Research Bulletin No. 192, pp. 63-74, 1958.
57. Witczark, M.W., Lovell, C.W. and Yoder, E.J., 1971, *A Generalized Investigation of the Potential Availability of Aggregate by Regional Geomorphic Units Within the Coterminous 48 States*, Highway Research Record 353, pp. 31-42.
58. Cechetine, James A. *Modified CKE [centrifuge kerosine equivalent] test*. (California Div. Hwy., Sacramento, Calif., USA). Proc. Assoc. Asphalt Paving Technol., 40, pp. 509-26 1971.
59. Bikerman, J.J. Surface Chemistry for Industrial Research, Academic Press, Inc. New York, N.Y., pp. 19-22, 1977.
60. Wade, W.H. *Spontaneous Imbibition of Fluids into Porous Vycor*, Soc. of Petrol. Eng. J., April, pp. 139-144, 1974.
61. Wingrave, J.A., Wade, W.H. and Schechter, R.S. *Liquid Imbibition into Evacuated Mesoporous Media*, Chapter 12, in Wetting, Spreading and Adhesion, ed. by J.F. Padday, Academic Press, New York, pp. 261-288, 1978.