A REVIEW OF RESEARCH PERTAINING TO ASPHALT COMPOSITION AND ITS RELATION TO QUALITY

by

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(The opinions, findings, and conclusions expressed in this report are those of the author and not necessarily those of the sponsoring agencies.)

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ABSTRACT

This report reviews both early and recent research on asphalt quality with emphasis on compositional factors and how the findings of such research relate to present-day problems with asphalt cements and asphaltic paving mixtures.

It is shown that historically the quality of an asphalt as a paving material has been judged by its physical characteristics and its performance in a pavement. However, when properly used, asphalts of widely different physical characteristics can provide good service. Conversely, if asphalts that normally provide good performance are used in improperly designed mixtures, poor performance can result. Accordingly, since many factors enter into pavement performance and these factors often interact with each other, it is difficult to define the quality of an asphalt on the basis of its characteristics alone.

Because of the complexity of the chemical composition of asphalt, it is necessary that analyses of asphalts be based on determining types of compounds or groups of constituents having similar behaviors. Several procedures are available for such analyses, but there is no universal agreement among chemists and asphalt technologists as to which procedure is best.

Research has also shown that asphalt-aggregate interactions vary depending upon the characteristics of the aggregates as well as those of the asphalt. Thus, an asphalt exhibiting optimum behavior with one type of aggregate may exhibit significantly different behavior with another aggregate. Accordingly, although it is recognized that physical characteristics are established by chemical composition, materials with significantly different compositions can possess similar physical characteristics.

It is concluded that asphalt specifications based on chemical composition do not now, or in the near future, appear feasible. However, expanded research to better define the relationship of physical characteristics to chemical composition is needed and should provide knowledge that would permit upgrading poor or borderline asphalt cements through adjustment of composition or the use of additives.

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INTRODUCTION

The first asphalt pavements in the United States were made with natural lake asphalts from Trinidad or Bermuda. Petroleum asphalts came into general use shortly after 1900.(1) In the beginning they were considered inferior to the natural lake asphalts, but as knowledge of how best to use them increased their lower cost and greater availability soon displaced the use of the natural asphalts.

The quality of petroleum asphalts has from the beginning been difficult to define. Most early users depended on personal judgment and trial and error to judge the quality of a paving mixture, and often the ideas of early asphalt technologists as to the most desirable properties of the asphaltic binder differed widely.

In 1935, the Association of Asphalt Paving Technologists (AAPT) held its first symposium on specification requirements for asphalt cements.(2) E. F. Kelley, chief of the division of tests, Bureau of Public Roads (BPR), opened the symposium by quoting a letter from Clifford Richardson's book "The Modern Asphalt Pavement," which had been published in 1905. The letter, written in 1893, was in reply to the question:

> What is the real test or standard of quality which will give the value of an asphalt for paving purposes?

The chemist to whom this letter was addressed made the following reply:

> The real and final test of the quality of an asphalt for paving purposes is actual trial for a proper length of time. Proper chemical and physical tests of a new variety of asphalt may strongly indicate its probable value as a paving material, but these tests, though of great assistance in forming an opinion, really only show the advisability of submitting the asphalt to the final and infallible test of actual trial.

Kelly went on to say, in 1935:

> It would be gratifying if, after the lapse of more than 40 years, we could say that this answer is no longer true. It would be gratifying if we could say that such advances have been made in our knowledge of paving asphalts as to enable us to judge the value of a new and untried product solely by its compliance with rational test requirements. Unfortunately, this does not appear to be the case. Rather, it seems to be a well-established fact that the service characteristics of a paving asphalt cannot be accurately defined by any of the tests which are now in use. It is the opinion of many well-informed engineers and technicians that the quoted statement is practically as true today as it was in 1893.

Today, in 1985, ninety-two years later, there are many who would say that the situation remains about the same -- our tests and specifications for asphalt cements do not adequately relate to performance in the pavement. And this is a major reason for the extensive national research effort now under consideration. However, much has been learned about asphalt composition in the last 92 years and new research efforts can and must build on that knowledge.

Although good performance of an asphalt pavement is the major goal of highway engineers, the danger inherent in passing judgment on the quality of an asphalt on the basis of its service behavior in pavements has often been pointed out. Kelley stated in 1935:

> Asphalt is only one of the constituents of an asphalt pavement and it, alone, does not control the performance of the finished work. The character and amount of filler material, the grade of the asphalt and the amount used, the adequacy of the foundation and, finally, the construction practices followed, are all important variables which control performance. Defects in an asphalt pavement are not necessarily a reflection on the quality of the asphaltic binder. Nor is the fact that two paving asphalts cannot always be handled successfully in exactly the same manner necessarily an indication that one is inferior to the other.(2)

This statement is still valid today.

RESEARCH CONCERNING ASPHALT PROPERTIES

The 1935 paper by Kelley reflected to a large extent the policy and general research strategy followed by the BPR and its successor the Federal Highway Administration (FHWA) over the last 50 years.

The first comprehensive federal effort was to catalog the range of properties of asphalt cements used in the United States. This was reported on by Lewis and Welborn in January 1940.(3) The materials tested included 42 sets of petroleum asphalt cements from 30 producers and included all the then used grades of asphalt cements. These samples were collected in 1935 and were submitted to meet federal specifications. The discussion of the paper brought out that some materials meeting the specifications had in some cases not performed satisfactorily, but in other cases had given good service. The inference drawn by some was that materials meeting the federal specifications did not represent the "best quality" materials produced at that time.

In December of 1940, Lewis and Welborn reported continuation of the study of the same group of asphalts. In this report the "thin-film oven test" (1/8-in. film - 5 hours at 325°F.) (3.2 mm - 5 hours at 163°C) was introduced.(4) It was shown that this test correlated very well with the hardening of an asphalt that occurred in the Shattuck mixing test -- a laboratory mixing test that had been shown to closely produce the same hardening of the asphalt that occurred in a batch type mixing plant. Lewis and Welborn presented evidence to show that the thin-film oven test was a measure of asphalt quality, and they recommended its adoption in asphalt specifications. The amount of volatile matter, loss of penetration, and loss in ductility were all considered important parameters of asphalt durability. This test was vigorously opposed by a number of the producers of asphalt, but became a part of the AASHTO standards in 1947 and was later adopted by the ASTM.

A second study was made on asphalts gathered in 1954. These samples were collected through the division and regional offices of the BPR and were representative of products being used by the various states during the 1954 and 1955 construction seasons. This series included a total of 323 samples from 105 refineries and represented all grades. A number of studies have been based on these asphalts.(5,6,7) It is of interest to note that the range of properties of the asphalts collected in 1954, which are generally considered to be representative of materials produced in the 1950's and 1960's, is very similar to the range of the properties of the asphalts gathered in 1935, except that the asphalts that had shown extreme hardening or loss of ductility in 1935 were no longer on the market in 1955. This was partly due to the adoption of the thin-film oven test, but primarily was the result of the elimination of "cracked" asphalts by a change in refining technology.

The final major effort involving these asphalts was a study of the performance of a selected number of roads in which the asphalts had been used. Asphalts were recovered from the pavements and their properties were compared to those of the original materials. Efforts were also made to show relationships between performance and chemical parameters.(8) Generally, it can be stated that for these asphalts, differences in chemical constituents were overridden by differences in the physical characteristics of the asphalt pavement mixture (void content, asphalt percentage, etc.).

The next series of asphalts studied by the FHWA was the series of viscosity graded asphalts.(9) This series was based on the requirements of study specifications suggested by the Asphalt Institute in 1963. The emphasis in this study was to show the viscosity-temperature susceptibility over temperatures ranging from 40°F. to 275°F. The effects of shear susceptibility were also considered, as were the effects of changing from a system of penetration grading at 77°F. to viscosity grading at 140°F. Again, the discussion of the paper after presentation to the AAPT showed wide differences of opinion -- primarily on whether grading at 140°F. was appropriate.

More recent studies evaluating overall ranges of characteristics were conducted by the Asphalt Institute and by Anderson and Dukatz for the U. S. Department of Transportation.(10,11) The Asphalt Institute study reported at the 1979 meeting of the AAPT was prompted by the concern that the characteristics of asphalt had changed as a result of the OPEC embargo in 1972 and 1973.(10) This study demonstrated considerable differences in asphalts of the same grade resulting primarily from differences in the sources of the crude petroleum from which they were obtained. However, it was concluded that present differences were not substantially greater than those that had been present prior to the embargo. The study by Anderson and Dukatz involved 100 samples gathered in 1978 and 1979. Statistical analyses were used to compare test results on these samples with test results on a number of samples collected by the FHWA in the period 1950-1970.(11) It was concluded that there was a statistically significant difference in the chemical and physical properties of the asphalts sampled over the period 1950-1980. Differences of opinion have been expressed as to the engineering significance of the differences reported by Anderson and Dukatz.

A full discussion of the various viewpoints and findings from such earlier research is beyond the scope of this presentation; however, from this general review of work in the area it is possible to draw certain conclusions that should form the basis of guidelines for further studies relating to the properties of asphalt. These are:

1. The characteristics of asphalt cements vary significantly depending on the crude source, the manufacturing process, or both. However, materials of widely different characteristics can be successfully used as the binder in asphaltic pavements.

2. Because of conclusion 1, it is not now feasible to write specifications for asphalt cements so that a given grade of

asphalt will behave the same in all climates and traffic situations regardless of the source of the asphalt.

3. Specifications should, therefore, be written so that extreme or unusual properties that indicate poor quality asphalt are eliminated but the range of suitable materials should be accepted.

4. Consumers must be made aware of any nonuniformity in the products being provided them for the same job or during the same construction procedure, in order that they may evaluate the need for adjustment of the asphalt grade or mix design during construction.

Primarily because of occasional unexpected and unexplained problems with asphalt pavements, the Virginia Highway and Transportation Research Council, in cooperation with the FHWA, undertook a study in 1983-84 to determine if significant changes in the characteristics of asphalt cements being supplied to the Virginia Department of Highways and Transportation had occurred as a result of the 1972 embargo and subsequent marketing changes. A second objective of the study was to summarize published literature concerning the chemical composition of asphalts, with particular emphasis on how composition has affected performance and its relation to asphalt specifications.

Two reports are now available as a result of this study. One, which is probably of most interest to asphalt suppliers and users in Virginia, summarizes the properties of asphalts supplied to that state during the 1983 construction season.(12)

Although tests to evaluate the adhesive characteristics of the asphalts with various aggregates are not yet completed, the results of the tests made to date have not revealed any asphalts with unusual properties. In commenting upon their products, all of the suppliers indicated that the properties of their asphalt produced at different times could possibly differ because of differences in the sources of crude and variable percentages of different crudes blended as the feed stock. However, the products from any one supplier tested in this study were relatively uniform in physical characteristics. Differences in viscosity-temperature susceptibilities of products from different suppliers were also relatively small. None of the differences noted would be expected to significantly affect the engineering properties of asphalt mixtures during construction.

All the asphalts tested were AC-20's and all met the requirements of the specification. The minimum viscosity value was 1,656 poises at 140°F. and the maximum was 2,258 poises. The minimum penetration at 77°F. was 64 and the maximum was 100. Under the penetration grading system, 18 of the asphalts would be graded at 85-100 and 5 graded as 60-70. Nine asphalts had penetration values between these two grades. All these asphalts had satisfactory ductility-penetration relationships.

Despite the assurance indicated by this study of 1983 production, however, the Virginia Department of Highways and Transportation experienced difficulty with tender or non-setting mixes on several projects during the hottest part of the summer of 1984. The results of the asphalt tests made were normal and as yet no single cause of the problem has been pinpointed. Most likely, the problem relates to a combination of mix design characteristics and, possibly, interactions between the aggregate and asphalt. This is strong evidence that all the answers concerning the behavior of asphalt are not yet known. Accordingly, a cooperative research effort involving asphalt cements such as that proposed as a part of the strategic transportation research study (STRS) is clearly justified.

The second report relating to asphalt composition was undertaken with the idea of "keeping it simple."(12) The aim was to present the fundamentals of asphalt composition and its relation to physical characteristics in such a way that an asphalt paving engineer could understand them, but that proved to be impossible -- not the understanding, but the simplicity. Asphalt composition is extremely complex and there is no simple approach that explains all the possible physical manifestations or interactions with aggregates and water, and changes with time, loading, and ambient conditions. However, there are some general relationships that should be helpful to the practicing asphalt paving engineer. These are discussed in a general way in the following section. Those interested in more detailed discussion should refer to the report listed as reference 13.

WHAT IS KNOWN

General Characteristics

Asphalts are principally carbon and hydrogen. Roughly, they are about 85% carbon and 10% hydrogen. The remaining 5% is nitrogen, sulfur, oxygen, and very small amounts of heavy metals such as vanadium or nickel. It has been estimated that there are more than a trillion possible ways these elements could be combined. To put this number in perspective, if one could begin today to examine and evaluate the effects of ten of these combinations each day he could complete the task in approximately 300 million years, provided he worked Saturdays, Sundays, and holidays. Because of these complexities, asphalt technologists have always approached the study of asphalt properties from a physical standpoint and, as previously indicated, have used trial and error methods. The early users of asphalt discovered that the black and sticky stuff held rock together pretty good and then, through experience, determined how hard it had to be. However, they soon found that "how hard it had to be" was dependent on temperature; that is, they found that different asphalts had different viscosity-temperature susceptibilities and that materials of the same consistency at one temperature could be of quite different consistencies at another. For the first 70 or 75 years, the penetration at 77°F. was the measure used for specifying consistency, but around 1970 viscosity grading specifications were adopted that specified viscosity at 140°F. as the means of designating different grades. It was reasoned that this would provide for more uniformity during mixing and paving for materials from different sources. While it is recognized that this could result in larger differences in consistency at low temperatures for materials of the same grade, the major concern is not with differences but only with whether or not thermal or load cracking would occur at the lowest temperature encountered, or if rutting would occur at the highest temperature. Mix design as well as asphalt properties enters into the picture with respect to these phenomena.

There are two other physical characteristics of the asphalt that are of concern. These are (1) its adhesion to stone and the degree to which water affects the adhesion or the strength of the mixture, and (2) durability -- that is, how well the asphalt retains its initial properties during storage and the construction process and in service in the road.

Compositional Factors and Methods of Analysis

All of these characteristics are controlled by the chemical composition of the asphalt. Not only do the types of compounds present affect the physical properties but the ratio of one type to another also makes a difference in performance. Since, as has been indicated, a study of individual components is out of the question, chemists have devised means of determining the percentages of various groups of components.

The most often used concept of an asphalt from the compositional viewpoint is that it is made up of asphaltenes, oils, and resins. All of these groups are in themselves very complex and vary in composition from one asphalt to another. The asphaltenes are composed of many different molecules that have a tendency to associate into large agglomerates. It is considered that these agglomerates are evenly dispersed in the oils by the action of the resins, which are the dispersing agent.

The procedures for analyzing an asphalt generally consist of first removing the asphaltenes. These are the constituents that are insoluble in nonpolar solvents such as pentane, hexane, or heptane. The constituents that are dissolved are called "maltenes." These are further analyzed by various methods.

Asphalt technologists don't agree on which method is best to use and a discussion of the fine points of each of them is beyond the scope of this presentation. Such methods are valuable as research tools, but none is considered applicable for adoption in asphalt specifications.

Another approach in determining the chemical properties of an asphalt is to identify and characterize the "polar" or "functional" groups within the asphalt that control how well the asphalt sticks to the stone and performs in the pavement. Under this concept molecules that have approximately the same type and number of "functional groups" will produce similar effects on physical properties even though the overall composition may be significantly different.

Still another way of examining asphalt composition is to measure the relative amounts of molecules of various sizes. This is done by a technique known as high performance liquid chromatography (HPLC). An arbitrary decision is made to characterize the asphalt molecules present as being large, medium, or small. The relative amounts of each of the sizes is then related to the performance of the asphalt in a pavement. Suitable ranges for the percentages of each size that constitute an acceptable asphalt from the durability viewpoint have been suggested but additional work needs to be done before the concept is generally accepted.

WHAT IS NOT KNOWN

The foregoing discussion shows that despite the complexities of composition, considerable progress has been made in unraveling how the composition of asphalt affects its physical properties but there is still a long way to go. Some of the problems that are of considerable importance to highway engineers are discussed below.

Long-Term Effects of Moisture

Asphalt-aggregate interactions are not fully understood, nor is the role of antistrip additives in promoting long-term resistance to moisture damage. In fact, there is evidence that in some situations the use of antistrip additives may detrimentally affect long-term behavior.

The popular basic concept of an additive is what is often termed the "tadpole" theory. The additive is made up of a polar end (the head of the tadpole) that has an affinity for the aggregate surface and can displace a water film on the aggregate. The tail of the tadpole is nonpolar and compatible with the asphalt.

In theory the polar molecules will migrate to the aggregate-asphalt interface and form a moisture resistant link between the asphalt and the aggregate. Petersen and his associates have shown, however, that not only do some of the tadpole molecules react with components in the asphalt, but that they also do not always reach the asphalt-aggregate interface. In addition, the reaction products may be of a type that when dry adhere well to the aggregate surface but are more easily displaced by water than were the initial asphalt molecules.

Slow Setting Asphalt Mixtures

It is not known why some asphalt-aggregate combinations give slow setting characteristics nor what test or tests should be included in specifications to measure any tendency of the asphalt cement to be slow setting.

None of the differences in asphalt relating to aggregate interactions and setting characteristics are measured by present specifications, and no test methods using equipment of the type normally found in highway testing laboratories are available.

Tailor Made Asphalts

In some discussions of asphalt research it has been suggested that high quality asphalts giving superior performance as a binder be manufactured by separating constituents and reblending them in optimum proportions or through the use of additives. However, at present the characteristics that should be possessed by such high quality asphalts cannot be fully defined. In fact, given the variations in asphalt-aggregate interactions based on differences in aggregate surface characteristics (electrochemical forces), it is doubtful that the same set of asphalt characteristics would be optimum for different type aggregates.

Finally, one of the greatest concerns is cost. Despite escalation in the last few years, the cost of asphalt remains relatively low compared to the cost of distillates from the crudes. To major oil refineries asphalt is still often an unwanted residue that must be disposed of. These companies are reluctant to spend money and research effort on the residues. If through cooperative research, such as the national program now being planned, a high quality "manufactured" asphalt should become available, costs are likely to be extremely high and may be difficult to justify if normally produced asphalts with an acceptable performance history were available at much lower cost.

THE ROLE OF ASPHALT PAVING ENGINEERS

In the interim until answers are obtained from new research efforts, a question to be considered is, What can the asphalt paving engineer do to improve his product? There is very little he can do with respect to the properties of the asphalt cement. State highway agencies establish the specifications and the grades to be used, and the asphalt supplier is generally responsible for compliance to the specifications. The asphalt cement is only one of the ingredients for the asphalt mix, and good business dictates that the materials available at the lowest cost be purchased. Thus, changes in characteristics of materials are not within the responsibility of the asphalt paving engineer. He does, however, have a responsibility to assure himself that there is compatibility between the aggregate he is using and the asphalt. He should also be sure that the mix is designed properly to provide for the proper amounts of asphalt and that, after compaction, the proper void content is present. Attention to these details would go far in eliminating problems often thought to be attributable to the asphalt cement. In fact, paving engineers might be surprised to find out how much stickier the asphalt cement will become when attention is given to known criteria for proper mix design and compaction on the road.

CONCLUSION

It can be concluded from this review that further studies of asphalt composition and the relation of composition to performance parameters are greatly needed and that such research is likely to provide benefits that far exceed its cost. However, the concept of a "tailor made" asphalt, as sometimes suggested, and a specification that would result in all asphalts of a given grade or designation having uniform construction and performance characteristics do not appear feasible. Differences in aggregates and subsequent asphalt-aggregate interactions are such that differences in performance are likely. If used properly in the proper mix design, asphalts of the same grade from different sources with significant differences in composition can provide equally satisfactory service. From the viewpoint of economics, if asphalts derived directly from the residues of petroleum distillation are available and are normally satisfactory, they should be used without extensive modification that would greatly increase their costs.

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