HARDENING OF ASPHALT CEMENTS DURING SERVICE IN PAVEMENTS
1963-66 PROGRAM

by

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Study 2-8-59-9
Research Area 8

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PROGRAM

I. OBJECTIVES OF STUDY NO. 2-8-59-9, RESEARCH AREA 8

Objectives of the Study are to:

(1) Investigate the asphalt cements used by the Texas Highway Department,

(2) Establish specifications to assure the use of superior asphalts by the Department, and

(3) Determine how the durability of asphalt cements in service can be improved.

This report describes an investigation which had as its objective the determination of changes in hardening and composition that occur in an asphalt cement while the hot mixture is being prepared and laid and during service in the pavement. This study applies specifically to objectives (1) and (3) above.

II. HISTORY

Study No. 2-8-59-9, Research Area 8 was started February 1, 1959, as Research Project No. 15. The eleven reports issued to date are listed in Table A-6 of the Appendix.

The portion of the study discussed in the present report was started in May 1963 and by August 1964 hot-mix surfacing projects were located at 17 sites in 14 of the 25 Districts within the Texas Highway Department. The asphalt cements used were obtained from ten different suppliers. Progress Report No. 8 gave the data obtained through one year of service at 13 sites in as many Districts. Information was also supplied concerning the aggregates used at the various locations. Progress Report No. 9 gave viscosity values at 77°F on the original asphalts used at the four additional sites established during 1964. Viscosity data at 50° and 60°F were reported for all seventeen original asphalts and shear susceptibilities (complex flow "c") were calculated for each asphalt of the two temperatures. Microductilities at 60° and 77°F were determined on the original and available extracted asphalts from the seventeen sites. A relationship was established between ductility and viscosity at 77°F.

III. CONCLUSIONS

1. Relative Viscosity, which is obtained by dividing the viscosity of an asphalt hardened by environmental and service conditions by its original viscosity, is useful for comparing the hardening of an asphalt or different asphalts under different environmental conditions.

2. Careful preparation, handling and laying of the hot mixtures discussed in this report resulted in 1.55 to 2.80 fold increase in viscosity of the recovered asphalt at 77°F. After three years of service in pavements, the Relative Viscosities of the recovered asphalts ranged from 13.5 to 41.5. The Relative Viscosities of the asphalts exposed to the laboratory hardening test ranged from 2.55 to 6.35 which is comparable to the 2.2 to 7.6 fold increase shown after two weeks of service.

3. The average Relative Viscosities at 77°F of the asphalts extracted after two weeks to three years in service give a straight line when shown against time (days) on a log-log plot.

4. The laboratory hardening test described in this report is the most satisfactory method now available for quickly evaluating the susceptibility of an asphalt to hardening in service.

5. Season of the year in which the pavement was laid, the Hveem stability and cohesion tests appear to have little effect on the condition of the highway surface after two to three years service. Limited data may be responsible for this conclusion.

6. High Average Daily Traffic count appears to be associated with a pavement surfacing showing excellent to good condition after two to three years in service.

7. The mixtures, which showed good service on the highway, flowed from the hot-mix plant at an average temperature of 300°F. Pavements showing poor condition after two or three years were surfaced with mixtures which left the hot-mix plant at an average temperature of 280°F. The higher temperature of the mix could have been responsible for better compaction on the highway.

8. Data accumulated during this investigation indicate that the nature of the aggregate used in the pavement may affect, to some extent, the hardening of the asphalt during service.

IV. OBSERVATIONS

1. After one, two or three years of service the asphalts recovered from the pavement, in some instances, have viscosities lower than the preceding sample. This appears to be caused by oil leakage from vehicles traveling on the road.

2. Asphaltene contents (n-pentane insolubles) of the asphalts extracted from the pavements were greater after one year than for the original asphalts by 1.4 to 8.3 per cent. However, after two years of service, three of the samples had decreased by 1 to 1.4 per cent. Asphaltene contents of the other eight samples had increased by modest amounts.

3. Viscosities of the petrolenes (n-pentane solubles) obtained from the asphalts extracted after two years of service were 1.05 to 5.85 fold higher than for those from the original asphalts. Investigation of a pavement made with <u>porous</u> limestone aggregate showed petrolenes from the asphalt extracted after two years of service to be 20 fold greater in viscosity than from the original asphalts. Low viscosity oil from the asphalt had been drawn into the capillaries of the limestone.

4. Nuclear activation analyses indicate that oxygen content of asphalts extracted after four months in service were in all cases higher than for the original asphalt. Then, in all but one case the oxygen dropped slightly after one year in service. After two years service, the oxygen content of eight samples increased or remained the same with respect to the one year sample, whereas five decreased.

V. RECOMMENDATIONS

It is recommended that the results given in this report be used in the design of any future investigations in order to avoid the oversights which occurred during planning and conducting the study discussed herein. In particular, consideration should be given to the following subjects:

1. For each source of asphalt used, at least two and preferably three sites at widely separated locations in the State should be established.

2. The sites used should be selected on the basis of different aggregate available as well as for the asphalt cement involved.

3. In addition to reporting the temperature of the hot-mix leaving the plant, temperatures of the mixture entering the paving machine and during the compaction of the surface should be recorded.

4. Ambient temperature during laying and compaction should be reported.

5. Average Daily Traffic and visual assessment of conditions should be obtained immediately after opening to traffic and on each anniversary of the pavement.

6. A longer test strip should be underlaid with aluminum foil so if an oil contaminated sample of pavement is obtained, another sample a few feet down the road can be removed immediately. It would also be advisable to prepare a longer test strip so that, if necessary, samples could be removed from the road after more than two or three years in service. Samples should be removed 1 day after the pavement is laid and then after 2 weeks, 6 weeks, 4 months, 8 months, 1 year, 2 years and as many anniversaries thereafter as are feasible.

7. At least a five-gallon sample of asphalt cement should be obtained from the discharge of the storage tank to the hot-mix plant and its temperature recorded.

8. All samples of mixtures taken at the plant or from the pavement should be packed immediately in dry-ice for delivery to the laboratory. On arrival the samples should be stored in a deep freeze cabinet until the extraction can be made. This procedure will retard hardening of the asphalt during the time which elapses between taking the sample and extraction of the asphalt.

9. Any available new methods of test on the asphalt and paving mixtures should be utilized.

VI. FUTURE WORK

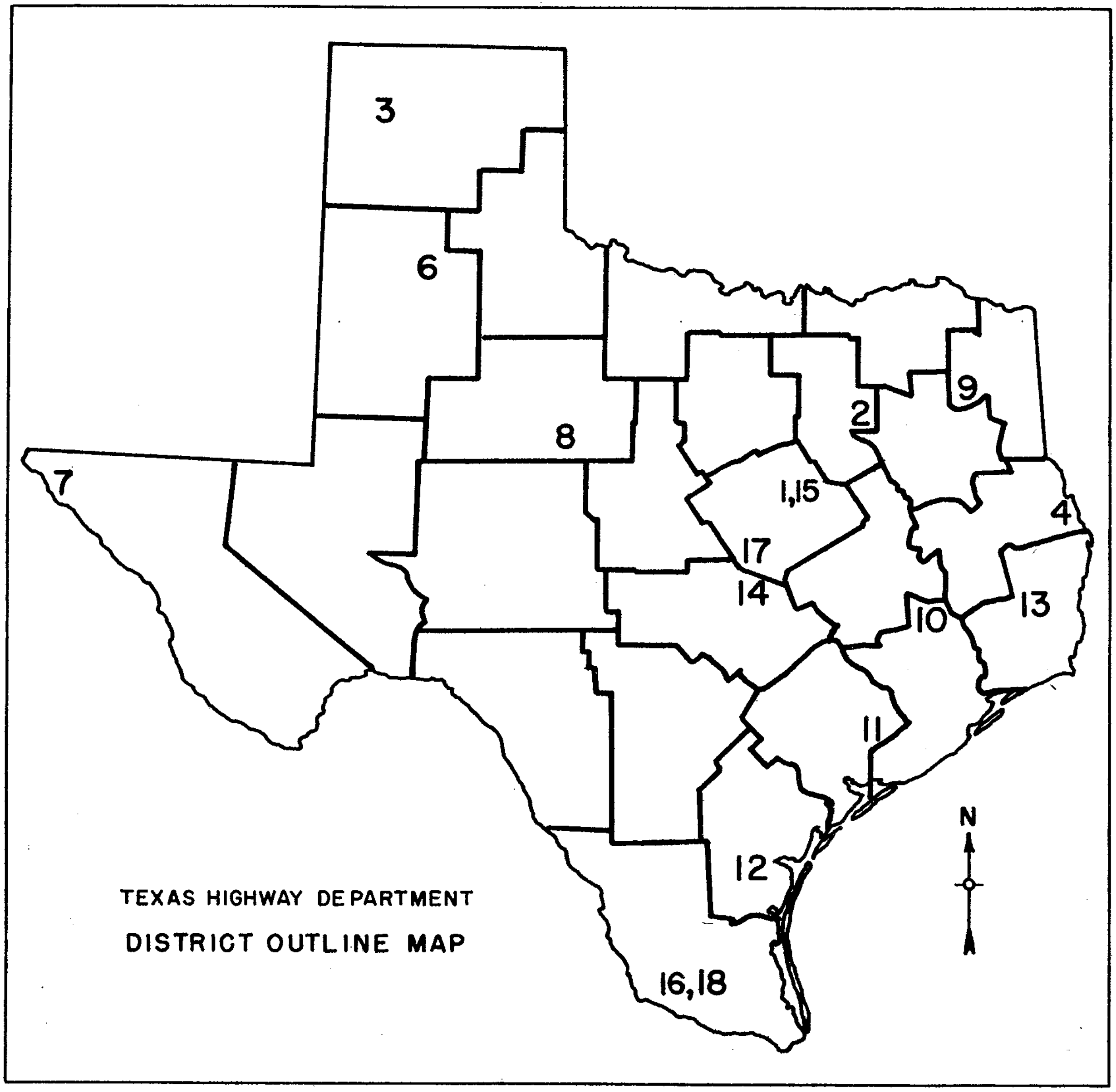
1. Polarographic studies will be made on the aggregate used in the field program discussed in this report. Such tests should yield information concerning the chemical activity of the aggregate surfaces.

2. Molecular weight measurements by the osmometer method will be made on the original and recovered asphalts. It is hoped that the data obtained on the original and recovered asphalts from the field program may be helpful in establishing a better understanding of this fundamental property and how it changes during service.

3. Experiments will be made to determine the effect of actinic light (ultra violet and short wave length visible radiation) on asphalt in an asphalt-mineral mixture. The main reason for this experiment will be to determine whether different mineral surfaces in contact with the asphalt can influence the hardening induced by short wave length radiation.

4. Thermal properties of selected original and recovered asphalts will be measured over a wide range of temperature in a Differential Thermal Analyzer.

5. It is planned to use Nuclear Activation Analysis to further investigate the oxygen content of asphalts. One phase of the study will be to determine whether the solvents commonly used to extract the binder from paving mixtures significantly affect the oxygen content of the asphalt.



LOCATION OF EXPERIMENTAL SITES

FIGURE 1

VII. EXPERIMENTAL WORK

1. Field Program

Seventeen sites in 14 Districts of the Texas Highway Department were selected for investigation. The locations of the 17 sites are shown on Figure 1, facing, which is a Texas Highway Department District outline map. Locations were selected where THD, in its maintenance program, was placing 1 1/4 to 1 1/2 inch thick (about 125 lbs./sq. yd.) hot-mix surfacings. On each highway a particular spot (station) was selected by District personnel.

A schematic layout of sample locations is shown in Figure A-1 of the Appendix. From this diagram it can be seen that the samples were taken from between the wheel paths, and from the outside lane if the experiment was located on a multi-lane pavement.

Before the paving machine arrived, heavy aluminum foil was fastened to the base by roofing nails. This was done to facilitate removing slabs of the surfacing material and to prevent contamination of the asphalt cement by primer applied to the base. Slabs taken from the test sections by District personnel were shipped to the Transportation Institute in special wooden boxes to prevent breakage during transport.

Pertinent information obtained at each site included District No., County, highway designation, project number, station, asphalt manufacturer (Code No.), date pavement was laid and temperature of fresh mix. These data are shown in Table A-1 of the Appendix. Identification and descriptions of the aggregates used at the first 13 sites are given in Table A-1 of Progress Report No. 8, dated May 1965. Since this table covers 26 single-spaced typewritten pages it is not reproduced in the current report.

At each site the following samples were collected:

(1) Original asphalt cement as it was supplied to the hot-mix plant (one gallon).

(2) Asphalt-aggregate mixture as it came from the plant (2 gallons).

(3) Asphalt-aggregate mixture when it was placed in the paving machine (2 gallons).

(4) A 2- x 2-foot sample of surfacing material taken one day after the pavement was laid and compacted.

(5) A 2- x 2-foot sample taken after 2 weeks.

(6) A 2- x 2-foot sample taken after 4 months.

(7) A 2- x 2-foot sample taken after 1 year.

(8) A 2- x 2-foot sample taken after 2 years.

(9) A 2- x 2-foot sample taken after 3 years.*

*Only for the first 13 sites.

TABLE 1

DATA ON ORIGINAL MIXTURES AND FACTORS AFFECTING CONDITION OF PAVEMENTS[a]

Site	Date	ADT 1965	Mix Temp °F	Hveem Stability	Hveem Cohesion	Condition After (Years)	
1	5-26-63	5,040	320	32	156	Good (4)	(3)
2	6-6-63	1,780	-	41	154	Base failure - minor	(2)
3	6-21-63	1,820[b]	300	50	389	Excellent(1)	(3)
4	7-1-63	1,700	270	31	63	Base failure - Minor	(3)
*							
6	7-22-63	3,700	250	36	90	Excellent ✓	(3)
7	7-23-63	1,200	275	42	134	Transverse cracks	(2)
8	7-22-63	3,590	325	50	214	Emulsion Applied	(2)
9	7-31-63	3,140	325	32	287	Very good	(3)
10	8-6-63	1,350	270	38	158	Some Cracks	(3)
11	8-22-63	4,580	265	36	126	Seal Coated	(3)
12	9-23-63	3,305	275	51	159	Excellent	(2)
13	10-24-63	2,000	300	34	-	Good - few reflection cracks	(3)
14	2-6-64	6,970[b]	325	58	-	Original surface excess asphalt, surface dry and bleached after	(3)
15	5-2-64	2,590	300	-	-	Good	(2)
16	7-24-64	3,630	280	-	-	Excellent	(2)
17	8-4-64	9,240	305	-	-	Excellent	(2)
18	8-11-64	3,230	300	-	-	Good to excellent. Some cracks due to soft subgrade	(2.5)

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* Site 5 was not investigated.

[a] 85-100 penetration asphalts used except at sites 17 and 18 where AC-20 was used.

[b] 1966.

2. Data on Surfacing Material Before and After Removal from the Pavements

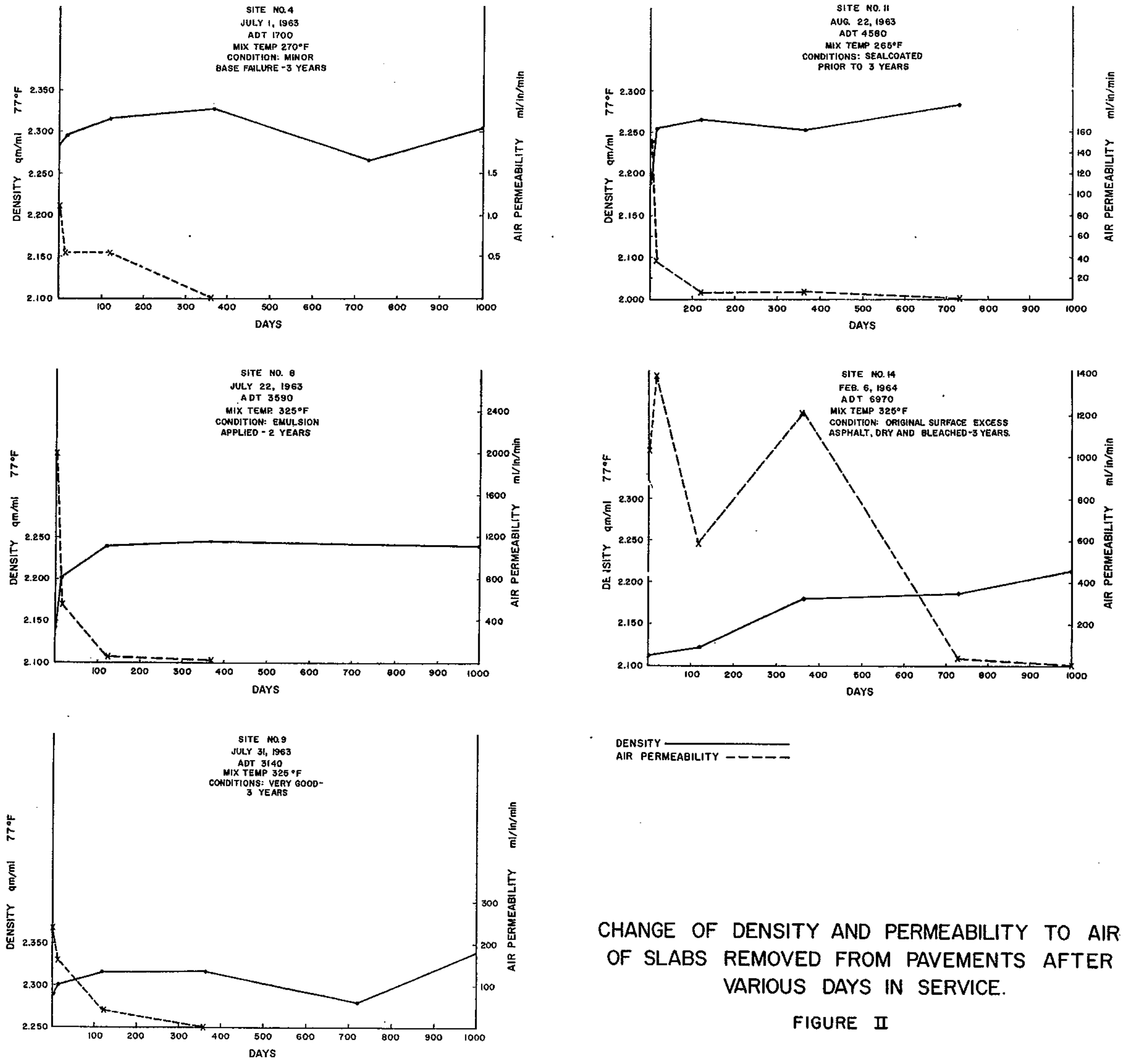
Table 1, facing, gives data on the season of the year (date) during which the pavement was laid, average daily traffic to which the pavement was subjected, temperature of the mixture as it came from the hot-mix plant, Hveem stability and cohesion for a number of the mixtures used and, finally, an indication of the condition of the road after 2 to 3 years of service.

Season of the year in which the pavement was laid appears to have little effect on its condition as determined by inspection after 2 or 3 years of service. The same appears to be true for Hveem stability and cohesion--although if more data were available a trend might be detected.

The 12 roads that showed excellent to good condition after 2 or 3 years carried an average of 4,000 + vehicles per day. Those surfacings that were showing distress after service carried an average of 2,100 + vehicles per day. This indicates that reasonable amounts of energy put into the pavement by mechanical action (traffic) reduces the time hardening of the asphalt binder and thus retards cracking and deterioration of the pavement. An exaggerated situation of this kind is a dead end street where the bituminous pavement receives little or no traffic and is badly cracked.

The surfaces that rated excellent to good were made from mixtures that flowed from the plant at an average temperature of 300°F. An average mix temperature of 280°F was observed at the plant for those pavements which were reported as less than good after 2 or 3 years of service. The highest temperature used was 325°F and the lowest 265°F. I do not believe that these differences in temperature of the freshly prepared mixtures are, in themselves, too significant but those prepared at an average of 300°F may have been in a better condition for compacting on the road. Certain highly qualified road builders believe that temperature during compaction is of considerable importance. It is unfortunate that temperatures of the mixtures were not taken during laying and compacting.

When suitable slabs of pavement were available, density and permeability to air measurements were made on a series of samples from each site. Data obtained are shown in Table A-2 of the Appendix. Plots of time (days) versus



CHANGE OF DENSITY AND PERMEABILITY TO AIR OF SLABS REMOVED FROM PAVEMENTS AFTER VARIOUS DAYS IN SERVICE.

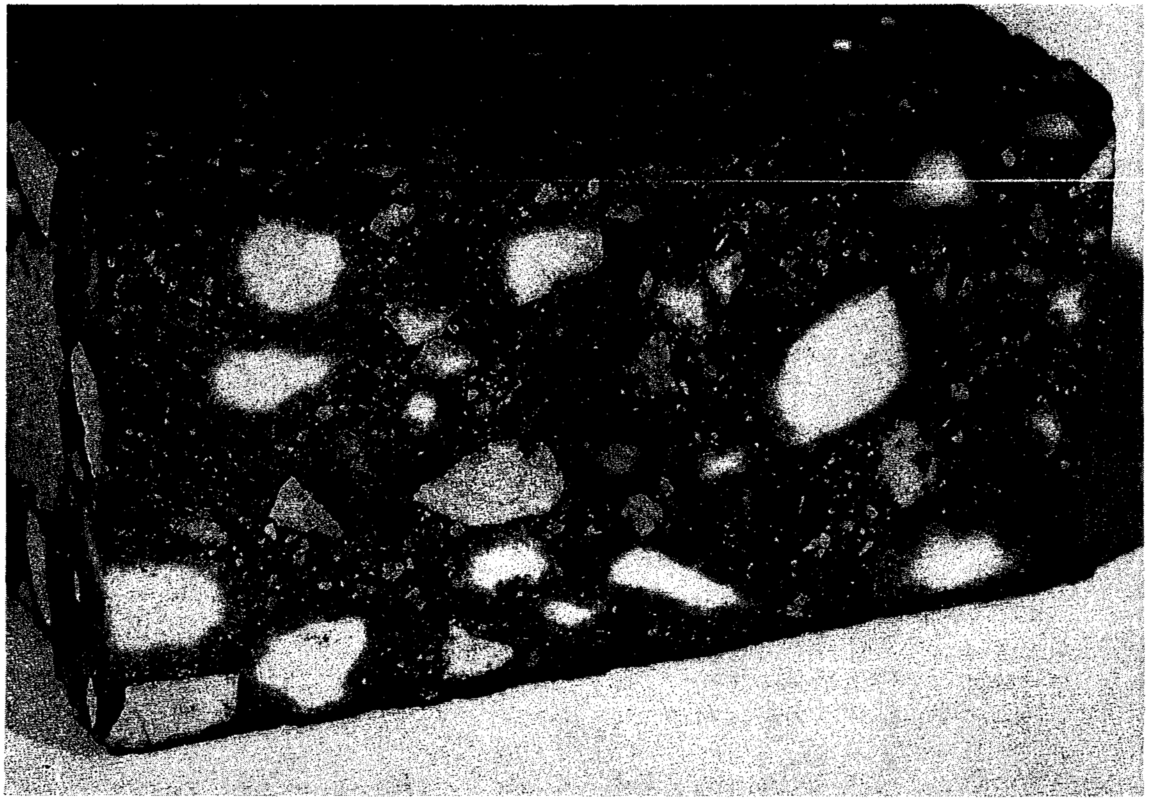
FIGURE II

density and air permeability data on slabs taken at intervals are shown in Figure II (facing) for sites 4, 8, 9, 11 and 14. Date the pavement was laid, ADT, temperature of the freshly prepared mixture and condition of the pavement after service are shown for each of the five sites.

Site No. 4 had a low ADT count (1,700) and a low mix temperature (270°F). After 3 years the road showed some distress, thought to be caused by minor base failure.

Site No. 11 had a high ADT (4,580) and the lowest mix temperature (265°F). It was necessary to seal coat this surfacing prior to its third anniversary.

Site No. 14 made from the same asphalt, but a radically different aggregate than that used at Site No. 11, had next to the highest ADT (6,970) of the 17 roads examined and the highest mix temperature (325°F) used. Site No. 14 is the only pavement laid during cold weather (February 1964). Another difference between Sites 11 and 14 (both made with asphalt cement from supplier No. 6) is the notable difference in aggregate used. Site No. 11 was made using quartz and chert mixed with oyster shell (new reef) whereas Site No. 14 was prepared with an aggregate composed predominately of porous limestone. The nature of the stone in this situation probably explains the statement shown in Table 1 that the original pavement surface showed excess asphalt but after 3 years the surface appeared to be dry and bleached. Figure III is a photograph of a cut surface of a slab from Site No. 14. The absorption of low viscosity but dark colored oils from the asphalt cement by the porous limestone is clearly evident. Later in this report it will be shown that a considerable increase occurred in the viscosity of the oils present in the asphalt extracted from this pavement surface after 2 years in service. Some of the extremely low viscosity oils were retained by the porous stone during the extraction process.



SLAB CUT FROM A BITUMINOUS PAVEMENT SHOWING ABSORPTION OF OIL FROM THE ASPHALT CEMENT BY POROUS LIMESTONE AGGREGATE

FIGURE III

3. Extraction and Recovery of Asphalt Cements from Mixtures and Pavements Samples

About 20-25 pounds of asphalt-aggregate mixture or surfacing removed from the road were placed in large Colorado type percolator extractors for separation of the asphalt from the stone. In order to insure, as nearly as possible, complete removal of all asphaltic components from the aggregate surface, a mixture of 6 parts benzene and 1 part 95 per cent ethyl alcohol was used to dissolve the asphalt. When the benzene-alcohol solution flowing from the bottom cone of the extractor showed a very light straw color, the heavy solution of extracted asphalt was centrifuged at 770 times gravity to remove most of the fine minerals that could have passed through the filter paper used in the extraction apparatus. The essentially mineral-free solution was distilled by the standard Abson procedure until a large portion of the benezene-alcohol was removed. This concentrated solution was then transferred to a rotary film evaporator and the remaining solvent removed at 115-150°F and 15 mm of mercury pressure. By this procedure, the final removal of solvent was accomplished by exposing the asphalts to temperatures only a few degrees above their softening points. Figure A-II Appendix, is a photograph of the evaporator without the vacuum lines and radiant heaters.

With 13 sites investigated over a period of 3 years and 4 sites for 2 years there should have been 8 mixtures or pavement samples extracted for each of 13 locations and 7 extracted for each of 4 sites making a total of 132 recovered asphalts. Actually only 116 extracted asphalts were obtained because 16 samples of mixtures or pavement slabs were lost, damaged or contaminated in some way. One source of contamination was spillage of lubricating or other oil on the pavement at the point at which the sample was taken (see Figure A-I, Appendix). This type of contamination was found amoung 4 of the samples taken after one or more years of service. A few samples taken from the truck or at the paver were contaminated, apparently by diesel fuel used to clean the truck bed. Some diesel fuel would remain in dents in the metal floor of the truck and we were unfortunate in receiving a sample from this particular area.

TABLE 2

RELATIVE VISCOSITIES OF THE 17 ASPHALTS (1963 and 1964 SERIES) HARDENED UNDER VARIOUS CONDITIONS AND TIMES

Temperature 77°F

Sites	Lab Test	Plant	Paver	1 Day	2 Wks	4 Mos	1 Yr	2 Yrs	3 Yrs
1	4.2	1.65	-	2.7	-	8.95	9.1	9.25	13.5
2	2.7	-	2.1	3.1	3.6	9.3	13.3	-	-
3	2.7	2.3	2.7	2.8	3.6	6.0	-	7.45	-
4	2.7	1.7	1.75	2.0	2.2	9.7	10.65	12.8	22.0
6	6.35	2.6	2.7	3.1	5.6	21.5	25.0	40.0	41.5
7	3.2	1.75	1.85	2.85	4.35	13.2	14.5	21.5	23.0
8	5.1	2.7	2.8	3.25	7.6	14.3	17.5	33.0	35.0
9	4.7	2.4	2.3	2.3	4.7	7.4	10.2	13.3	14.4
10	2.7	1.15	1.55	1.9	2.85	8.3	13.8	20.5	27.4
11	3.2	2.6	-	-	3.6	15.2	22.0	27.0	-
12	3.2	1.4	1.6	2.4	2.6	5.7	15.3	29.0	32.5
13	2.55	2.2	2.5	-	2.8	5.75	9.85	22.0	23.0
14	2.8	1.8	-	1.9	-	3.3	10.6	12.7	20.0
15[a]	3.35	2.2	-	2.2	4.3	12.0	14.2	35.0	-
16	3.4	2.1	2.8	2.9	5.0	9.0	19.0	23.5	-
17	3.8	2.1	-	3.4	4.3	11.4	20.8	27.8	-
18	2.7	2.1	-	2.3	2.9	7.7	14.0	18.8	-
Ave.	3.50	2.05	2.25	2.60	4.00	9.90	15.00	22.10	25.25

[a] This surfacing was 0.75 inch thick. The other 16 surfacings were about 1.50 inch thick.

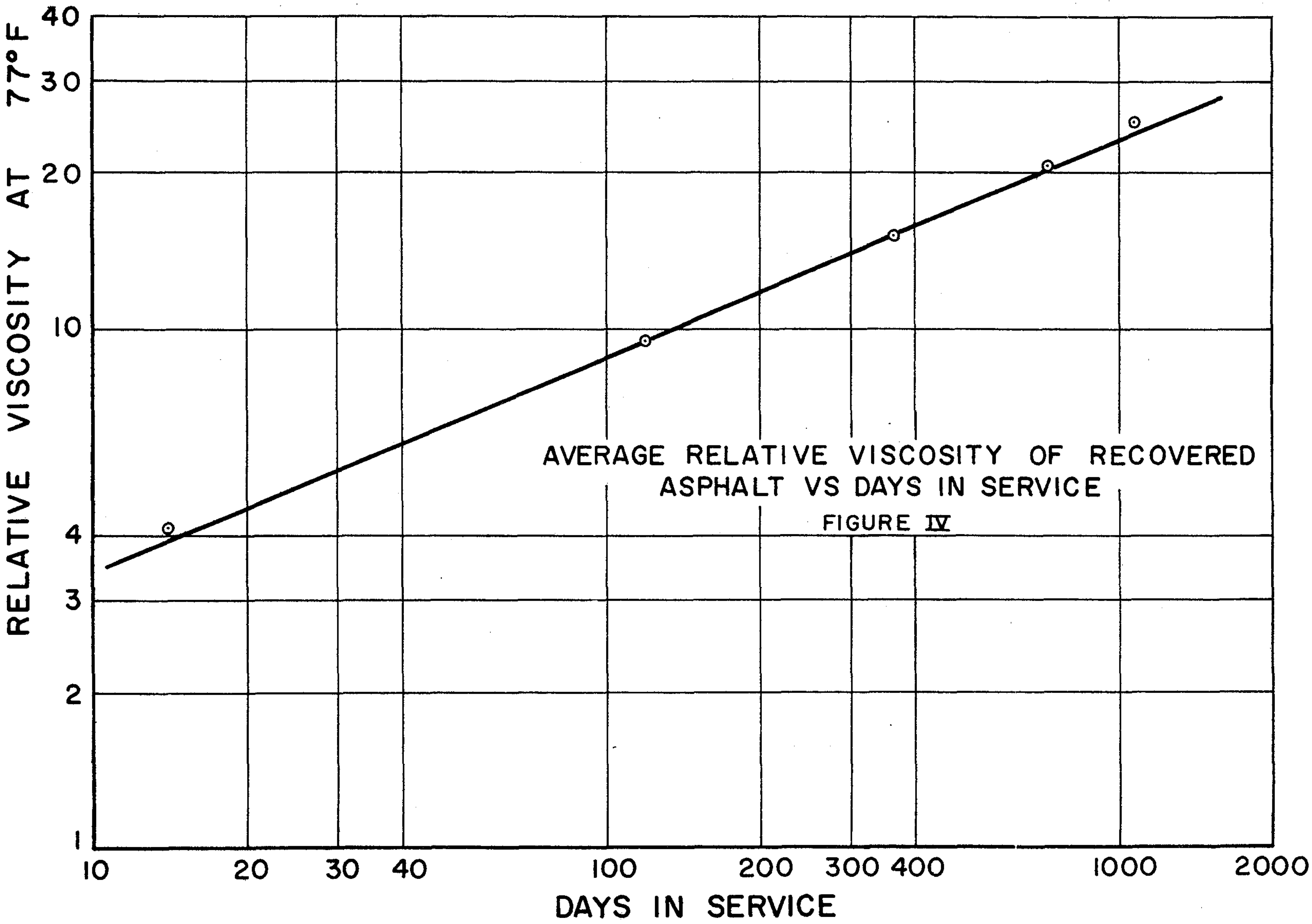
4. Flow (Rheological) Properties of the Original and Extracted Asphalt Cements

ASTM penetrations at 77°F, 100 gms/5 secs. and viscosities at 77°, 95°, 140° and 275°F were determined on the 17 original asphalts and on each recovered asphalt. Measurements at 77° and 95°F were made using the thin film Hallikainen viscometer (ASTM Book of Standards, Part 11, 831, March 1967), and values were calculated at 5×10^{-2} sec^{-1} rate of shear. Viscosities at 140° and 275°F were determined in Cannon-Manning vacuum capillary tube apparatus (ASTM Designation D-2171-66, Part 11, pp. 648-52, March 1967).

Relative viscosity for each recovered asphalt was calculated by dividing the viscosity of the recovered asphalt by the viscosity of the original asphalt at the same temperature. The resulting quotient indicates how many fold the asphalt increased in hardness because of the treatment or service it had encountered. Use of relative viscosities in comparing and discussing hardening of asphalts makes for an easier understanding of the changes because the original viscosities varied considerably (0.57 to 1.18 megapoises at 77°F).

A laboratory hardening test based on ASTM Book of Standards, Part 11, pp. 831-35, March 1967, was also made on each of the 17 original asphalts. A 15-micron film of the bitumen was placed on 4 cm x 4 cm glass plates and exposed in a dark air oven at 225°F for 2 hours. The cool, hardened film was removed from the glass surface by a razor blade, the asphalt placed between the plates used in the thin film viscometer, and viscosity measured at 77°F and calculated at 5×10^{-2} sec^{-1} rate of shear. Viscosity of the hardened asphalt was divided by the viscosity of the original asphalt to obtain the Relative Viscosity after the laboratory test. Viscosities on all asphalts and Relative Viscosities on all recovered samples at 77°, 95°, 140° and 275°F are given in Table A-3 of the Appendix.

The Relative Viscosities at 77°F, shown in this table, have been assembled and are shown in Table 2, facing. This tabulation gives a condensed picture of the differences in hardening of the asphalt cements over a period of two years at 16 sites and for three years at 10 sites. The asphalts used at sites 6 and 8 showed the greatest relative viscosities in the laboratory thin film test (2 hours at 225°F in a dark air oven) and also showed the highest relative viscosities (41.5 and 35.0 respectively) after recovery from the pavement at the end of 3 years in service. A log-log plot of the Average Relative Viscosities given in the last line of Table 2 versus time in days is shown in Figure IV following. This chart indicates that, in general, hardening of asphalt with time in service is a log-log relationship.



AVERAGE RELATIVE VISCOSITY OF RECOVERED ASPHALT VS DAYS IN SERVICE

FIGURE IV

The ranges of Relative Viscosities for the asphalts from the 17 sites are shown in the tabulation below. For example, the values obtained by the laboratory hardening test varied from 2.55 to 6.35 times harder than the original asphalts. The values for the asphalts extracted after 3 years of service varied from 13.5 to 41.5 times harder than the original asphalt.

Age of Sample	Range of Relative Viscosity Values Obtained from 17 Sites
Lab. Test	2.55 - 6.35
Plant	1.15 - 2.70
Paver	1.55 - 2.80
1 Day	1.90 - 3.40
2 Weeks	2.20 - 7.60
4 Months	3.30 - 21.50
1 Year	9.10 - 25.00
2 Years	7.45 - 40.00
3 Years	13.50 - 41.50

The data shown in Table 2 and the above range of Relative Viscosity values indicate that, with temperature of the hot-mix not exceeding 325°F, hardening during handling, laying and compacting is not as great as commonly believed.

Progress Report No. 9, issued during August 1966, gives rheological data not repeated herein. Included were (a) viscosities of the 17 original asphalts at 50° and 60°F (b) shear susceptibility (non-Newtonian flow) of the same 17 asphalts at 50° and 60°F (c) microductility tests on the original and extracted asphalts at 60° and 77°F and (d) the relationship between viscosity and ductility.

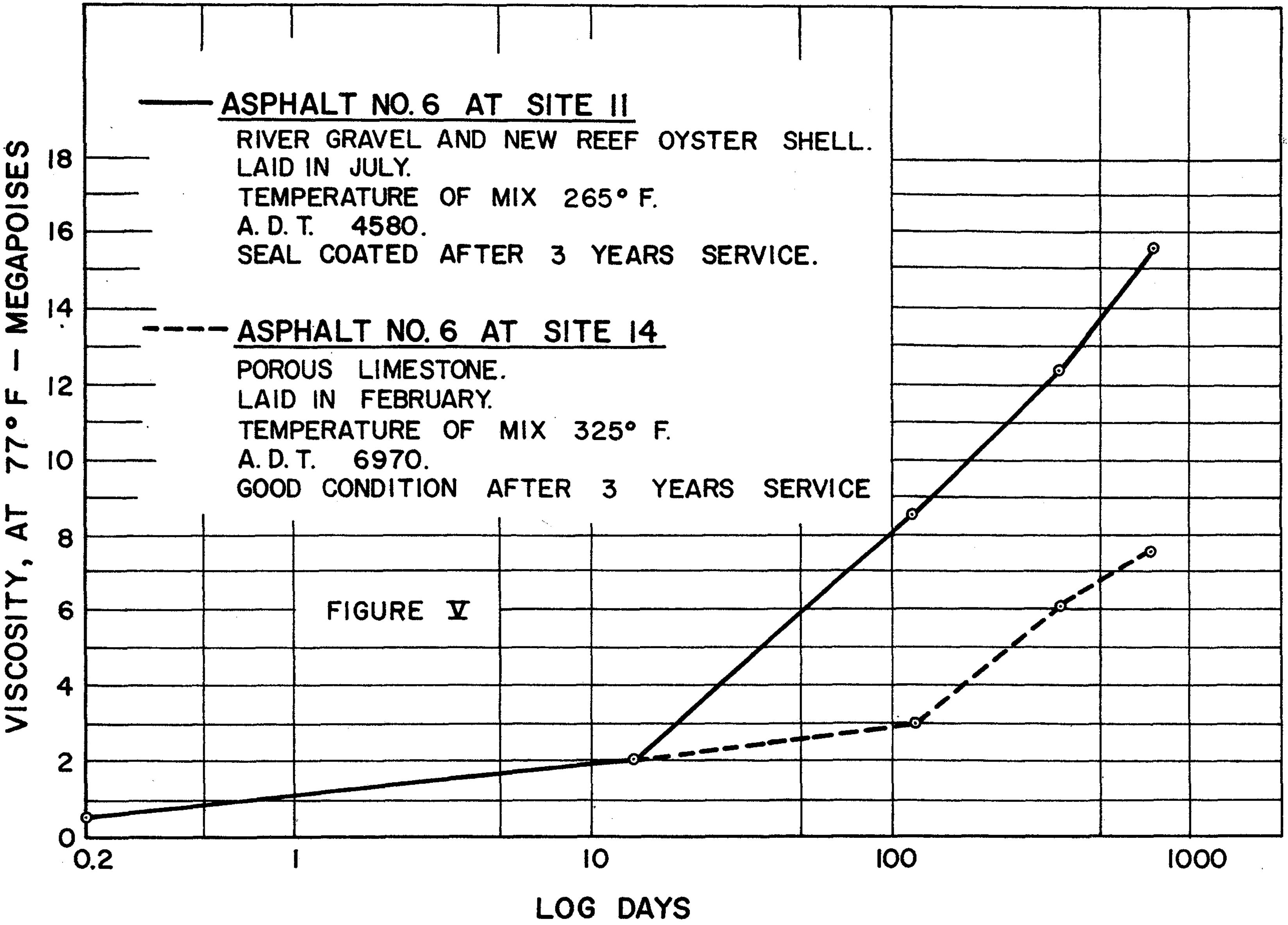


FIGURE V

ASPHALT NO. 6 AT SITE 11
RIVER GRAVEL AND NEW REEF OYSTER SHELL.
LAID IN JULY.
TEMPERATURE OF MIX 265° F.
A. D. T. 4580.
SEAL COATED AFTER 3 YEARS SERVICE.
ASPHALT NO. 6 AT SITE 14
POROUS LIMESTONE.
LAID IN FEBRUARY.
TEMPERATURE OF MIX 325° F.
A. D. T. 6970.
GOOD CONDITION AFTER 3 YEARS SERVICE
VISCOSITY, AT 77° F — MEGAPOISES
0
2
4
6
8
10
12
14
16
18
0.2
1
10
100
1000
LOG DAYS

5. Variation in Hardening (Viscosity) of a Particular Asphalt at Different Sites

An important fact has been noted from this investigation. Some asphalts during service for a particular period of time harden (increase in relative viscosity) much more than others. The cause for excessive increases in viscosity of certain asphalts during service beyond doubt is related to their composition, nature of the aggregate used in construction of the pavement, effect of actinic radiation, and other variables such as temperature of the asphalt-aggregate mixture during compaction and later under traffic.

Two sites (No's 11 and 14) are interesting because of questions raised by the data. Figure V, facing, is a plot of viscosity at 77°F versus log days, showing the hardening of the asphaltic binder at the two sites. The asphalts came from the same supplier and had almost identical original flow properties. The viscosities of the original asphalts were so near the same that it was not necessary to use Relative Viscosity to obtain a satisfactory comparison of the two situations. Aggregates used at the 2 sites were entirely different. For the first 2 weeks in service, the viscosities of the recovered samples were about the same, but from then on the hardening at Site 11 was greater than at Site 14.

Although Site 11 was laid in hot weather, the temperature of the mix was 60°F lower than at Site 14 which was laid in cool weather. ADT at Site 11 was 4580 compared with 6970 at Site 14. Because of the porous nature of the limestone used at Site 14 some light oil was drawn from the films of asphalt and held within the capillaries of the limestone. This effect is dealt with in Section 7, "Special Tests on Original and Extracted Asphalts."

6. Apparent Effect of Mineral Surfaces on Hardening of Asphalt Cements During Service

One outstanding difference between Sites 11 and 14, both of which contained the same asphalt cement, was the nature of the aggregate used. This fact, together with an increasing awareness of the possible effect of mineral surfaces on inter- and intra-molecular rearrangements in the asphalt films led to a series of experiments which it was thought might yield some valuable information.

Two hundred gram lots of AC-20 (Asphalt No. 11) were blended with 265 grams of the following mineral powders passing 200 and retained on 270 sieves:

Pure Silica
New Reef Oyster Shell
Old Reef Oyster Shell
Iron Ore Soil (from Site 10)
Gravel and New Reef Oyster Shell (Site 11)
New Reef Oyster Shell (Site 12)
Dolomite (Site 13)
Porous Limestone (Site 14)

The fine mineral powders were used to expose a large surface area of each mineral to the films of asphalt. Hot, dry mineral powder was carefully dispersed in hot, fluid asphalt to prevent entrainment of air. Mixtures were gently stirred until the mass became so viscous that the powders could not settle to the bottom of the tin can in which the mixture was prepared. The eight mixtures and a can of the original AC-20 were each covered with a tight lid and placed in an air oven maintained at about 45°C (113°F) to accelerate any reactions that might take place. After 2 months, the 9 cans were removed from the oven and the mixtures dissolved in a blend of 6 parts benzene to 1 part 95% ethyl alcohol and refluxed through a Colorado type extractor. The solutions of asphalt were centrifuged at 770 times gravity to remove fine particles that passed through the filter paper in the extractor. The essentially mineral-free asphalt was then recovered by removing the solvent in a rotary, thin film, flash evaporator (Figure A-II, Appendix). The recovered asphalts were tested for penetration and viscosity at 77°F. The Hallikainen sliding plate viscometer was used to measure the viscosities which were calculated at 5×10^{-2} sec^{-1} rate of shear.

A similar series was prepared, treated and recovered in the same manner except AC-20 (Asphalt No. 6) was used and the original asphalt and the 8 blends were maintained at 45°C (113°F) for 6 months prior to extraction and recovery. Table A-4 in the Appendix gives the values obtained for both series. No significant differences in hardness were found for the asphalts exposed to the different mineral surfaces. These negative findings indicate that thermal energy (heat) in the absence of air and sunlight does not greatly stimulate activity at the aggregate-asphalt interface.

After much thought and discussion concerning the significance of these negative results I decided that similar sets of mixtures should be exposed (without lids on the containers) at a uniform temperature and time to short wave length visible and to ultra violet (chemically active) light. The asphalt would then be separated from the mineral powders, the solvent removed and the asphalt tested for any differences in hardening caused by changes in the bitumen at the mineral surface. These experiments, which are listed under Future Work, should indicate whether actinic energy in the presence of air can stimulate changes at the asphalt-aggregate interfaces and whether the kind of aggregate influences the degree of reaction.

TABLE 3

PERCENT ASPHALTENE CONTENTS

Site No.	Supplier No.	Original Asphalt	Asphalt Extracted After One Year	Asphalt Extracted After Two Years	Kind of Aggregate Used
1	3	22.6	24.0	25.0	Siliceous Gravel
9	3	25.8	30.6	26.6	Iron Ore Slag and Sand
11	6	4.9	8.2	11.6	Gravel and New Reef Oyster Shell
14	6	2.0	6.8	6.9	Porous Limestone
6	7	14.4	22.1	23.7	Gravel
8	7	20.0	25.4	21.0	Limestone
4	11	19.5	23.4	22.4	Gravel and Clay
10	11	20.5	24.1	25.2	Iron Ore
7	15	13.3	19.9	18.3	Hard Non-Porous Limestone
12	2	13.7	22.0	24.0	New Reef Oyster Shell
13	5	9.4	11.5	13.9	Hard Dolomite
	Average	15.0	19.8	19.9	

7. Special Tests on Original and Extracted Asphalts

Since samples of original and extracted asphalts were available from 10 different suppliers, the opportunity was taken to make special tests which would give some indication of changes occurring in the asphalt cements with time in service.

(a) Asphaltene Content

Original asphalts from eleven sites and samples recovered after one and two years of service were analyzed for pentane asphaltene content. The technique used was to fill alundum extraction thimbles (RA 84 Dense) with glass wool and pour about 2.5 grams of molten asphalt over the wool. The thimbles were weighted before and after the addition of the asphalt to obtain the precise weight of each sample. These alundum thimbles were placed in slightly larger paper thimbles which fitted into a Soxhlet Extractor. For each test one hundred and fifty ml of n-pentane were refluxed through the asphalt coated glass wool for about 24 hours. The resulting solution of petrolenes (pentane solubles) was stripped of solvent, the residue cooled and weighed to determine the amount of extracted petrolenes. This subtracted from the weight of the original sample gave the amount of n-pentane insolubles (asphaltenes). This procedure avoided the weighing of asphaltenes in air, thereby precluding errors caused by the rapid oxidation of unprotected asphaltene. Data given in Table 3, facing, show that asphaltene contents increased in each case after one year of service. However, the samples recovered after two years of service showed some variation that is hard to explain. It could be experimental errors but all samples were analyzed in triplicate and if trouble was encountered an additional two or three analyses were made. The numbers given in Table 3 are averages of closely checking values. By a study of the table, it will be noted that there are four pairs of samples, each pair having the same source of asphalt but used at two different sites. In three of these groups, the samples from one of the sites were lower in asphaltene content after two years than for one year. These cases are underscored. The reverse was true for the other member of the pair. The situation with the sites using Asphalt No. 6 are "normal." This behavior indicates how little we know about the changes in chemical composition that take place in asphalt cements during service in roads.

(b) Viscosity of Petrolenes

Petrolenes from the original asphalts and from the bitumens extracted after two years of service in pavements were measured for viscosity at

TABLE 4

VISCOSITIES (POISES) OF PETROLENES AT 77^{o}F

Site No.	Supplier No.	Viscosities of Petrolenes Poises, at 77^{o}F, from		Ratio Vis (2)/Vis (1)
		Original Asphalt (1)	Asphalt Extracted After 2 years of Service in Pavement (2)	
1	3	5,220	8,600	1.65
9	3	7,600	9,600	1.25
11	6	47,600	278,000	5.85
14	6	75,000	1,600,000	20.10
6	7	17,000	21,000	1.25
8	7	10,800	25,000	2.30
4	11	4,780	-	-
10	11	4,040	4,700	1.15
7	15	16,700	26,000	1.55
12	2	29,800	31,200	1.05
13	3	70,000	206,000	2.95

77°F in the sliding plate viscometer. In most cases films 6 to 12 microns thick were used in these tests. Table 4, facing, gives the viscosity values obtained and shows that the ratio of viscosities between nine 2-year-old and original samples ranged from 1.05 to 5.85, with an average ratio of 2.1. The exception is the petrolenes from the 2-year-old sample from Site 14 which was 20 fold greater in viscosity than for the original asphalt. This is the pavement shown in Figure III page 13 where some of the light oils from the asphalt cement had been absorbed by the porous limestone used in the hot-mix surfacing. The removal of low viscosity oils from the asphaltic binder could be responsible for the change of appearance of the pavement at Site 14 mentioned in Table 1, page 9. Although the original surface showed evidence of excess asphalt, the surface appeared to be dry and bleached after three years of service.

About 10 pounds of aggregate recovered from Site 14 was treated as follows:

(1) Passed through a No. 8 sieve to remove small stones and mineral filler.

(2) Coarse stones were cleaned of adhering dust, placed in tin cans containing benzene and allowed to stand for 5 days at 77°F to extract oil from the capillaries of the stone.

(3) The resulting benzene solution of oil was decanted, filtered to remove any fine mineral particles and carefully distilled to remove all benzene from the oil.

(4) Six grams of a dark colored oil was recovered which had a viscosity of 1.2 stokes at 77°F.

The 6 grams of oil represent about 2 per cent of the asphalt that had been mixed with limestone to form the pavement. It is logical that the removal of 2 per cent of an extremely fluid oil could be responsible for the large viscosity increase (from 278,000 to 1,600,000 poises) of the petrolenes in Asphalt 6 recovered from Site 14 after 3 years of service.

(c) Oxygen Content

The importance of oxidation and photo-oxidation in the hardening of asphalts has been recognized for many years. Reactions involved in the oxidation process vary in degree and probably in kind with temperature.

At the high temperatures (425°-550°F) used in the production of air-blown asphalt for roof coatings and other hard, gel-type asphalts, hydrogen as water, carbon as CO_2 and low molecular weight hydrocarbon derivatives containing hydroxyl, carbonyl and carboxyl radicals are removed as fumes from the vessel. Small amounts of air-blown asphalt are sometimes blended into asphalt cements used in road building, but highly blown products are not considered satisfactory for pavement construction.

Experiments conducted in the past at various temperatures have shown that oxygen causes definite hardening of films of asphalt cements and the degree of hardening is increased if ultraviolet or short wave lengths from sunlight are present. Such hardening is very severe at the surface of the asphalt but may not progress to any great depth. However, the hardening of the surface can be so great that the bituminous surface is worn away by the action of rain, wind and traffic to reveal a fresh surface to the inroads of the combined action of oxygen and sunlight. Oxygen alone can penetrate for some distance into a bituminous pavement depending on variables such as temperature and porosity of the road surface. Information concerning the oxygen content of an asphalt prior to use and after subjection to environmental factors should assist in an understanding of the changes that take place in a bituminous surfacing.

For many years the difficulties encountered in making a direct measurement of oxygen in bituminous materials forced the asphalt technologist to be satisfied with analyses for C, H, N, and S, followed by subtracting the sum of these 4 values from the original sample. The remainder was called oxygen content, but in fact the value determined by difference represented the oxygen and metals content, and accumulated errors from the C, H, N and S analyses. In recent years direct chemical methods have been developed for the determination of oxygen, but for some materials the results are not very accurate. However, if nuclear activation equipment is available, oxygen content can be determined rapidly and accurately without destruction of the sample provided no appreciable amounts of boron or fluorine are present. Nuclear activation analysis is a method of quantitative and qualitative chemical analyses which is based upon measurement of the characteristic radiation emanating from isotopes formed by bombarding a sample of material by nuclear particles.

Texas A&M University has very fine activation analyses laboratories and equipment. The availability of this equipment and the expert personnel of the Activation Analysis Research Laboratory were made available to us for the testing of some of the asphalt samples mentioned in this report.

TABLE 5

OXYGEN CONTENTS OF 85-100 PENETRATION ASPHALT DETERMINED BY NUCLEAR ACTIVATION ANALYSIS

		Percent Oxygen (Values to Nearest 0.05)				
			Extracted Asphalt After			
Site No.	Supplier No.	Original Asphalt	4 Months	1 Year	2 Years	After Lab Test[a]
4	11	1.05	1.85	1.60	1.45	1.40
10	11	1.05	2.15	1.50	1.55	1.45
11	6	0.55	1.40	1.15	1.20	2.20
14	6	0.60	1.50	1.45	1.50	2.60
6	7	0.80	1.50	1.45	1.35	1.70
8	7	0.75	1.70	1.65	1.55	2.00
1	3	0.30	1.20	0.90	0.90	2.95
9	3	0.30	0.80	0.70	0.75	2.05
2	18	0.55	1.40	1.05	1.25	2.25
3	8	0.50	1.20	1.10	0.95	1.85
7	15	0.65	1.25	1.20	1.20	1.85
12	2	0.75	1.40	1.25	1.50	1.95
13	5	0.45	0.95	1.00	1.15	2.65

[a] Fifteen-micron films were heated at 225°F in a dark air oven for 2 hours.

About 1 gram of each asphalt (weighed to the 4th decimal place) was placed in a small polyethylene container which had been thoroughly cleaned. Duplicate samples were prepared for each original and recovered asphalt. As many as 5 tests were made on the contents of a single container. A block diagram of the neutron activation analysis system is shown in Figure A-III of the Appendix.

Data obtained from these exploratory experiments are shown in Table 5, facing. It will be noted that the oxygen content of the nine original asphalts varied from 0.30 to 1.05 weight per cent. Samples from four suppliers were obtained from two sites. Two pairs of duplicates (Sites 4 and 10 and 1 and 9) showed the same oxygen content; the other two pairs (Sites 11 and 14 and 6 and 8) showed differences of 0.05 per cent. Oxygen contents of asphalts from the 13 sites increased after 4 months of service but the increase was not uniform for the above mentioned pairs. This would be expected because of the stone used in the pavement and other factors affecting the presence of oxygen in the asphaltic binder. Oxygen content of the asphalt extracted from the pavement after one year of service, except for that from producer No. 5 (Site 13), was slightly less than for the sample obtained after 4 months. Analyses obtained on the asphalts after two years service in the roads varied considerably with reference to the one-year old samples. Of the 13 asphalts tested after two years, seven increased, four decreased, and two were the same in respect to oxygen content after one year. All of the original asphalts heated in the form of 15-micron films in a dark air oven for 2 hours at 225^{O}F showed considerably higher oxygen content than any of the materials extracted from the pavements except for the two asphalts from Supplier No. 11. For this asphalt the oxygen in the sample subjected to the laboratory test was about the same as that in the one- and two-year old samples.

The following tabulation shows the per cent increase in oxygen for each of the nine different asphalts after two years in the pavement. Values for each of the 4 pairs were averaged to obtain the values shown for asphalts 3, 6, 7 and 11.

Asphalt No. 11 which had the most oxygen in the original asphalt showed the least increase in oxygen content after two years.

Asphalt from Supplier No.	Percent Increase in O_2 After 2 Years in Pavement
3	165
18	125
6	120
5	115
8	110
2	100
15	85
7	80
11	45

There are several factors involved in the extraction of an asphalt from a pavement that must be carefully examined before the movement of oxygen can be fully explained. However, the exploratory data shown in Table 5 indicate the potential usefulness of the rapid, easily repeated, non-destructive test afforded by nuclear activation analysis in the tracing of oxygen occurrence in road asphalts during service and in any laboratory test used to evaluate susceptibility of the bitumens to hardening by heat and air. This method can also be used to determine the amounts of trace elements, such as vanadium, zinc, etc., in asphalts.

The asphalts tested by activation analyses and discussed above were also analyzed by a reputable commercial analytical laboratory for oxygen using an established chemical procedure (the so-called carbonate method). The oxygen values obtained by this method were higher than were found by activation analysis. Table A-5 of the Appendix gives the data obtained by activation and chemical methods. Oxygen content by the chemical procedure varied from 7.5 to 200 per cent greater than by the activation method. The literature contains a number of references stating that activation analysis is more accurate for low percentages of oxygen than are the chemical procedures.

Progress Report No. 10 of Research Study No. 2-8-59-9 "Oxygen Consumption by Asphalt Films and Resulting Viscosity Changes" gave information which led to the need for measurement of oxygen in asphalt by nuclear activation analysis.

VIII. APPENDIX

TABLES

FIGURES

TABLE A-1

GENERAL INFORMATION CONCERNING SEVENTEEN SITES[a]

Site No.	District	County	Highway	Project	Stations	Asphalt Supplier No.	Date Pavement was Laid	Temperature °F of Fresh Mix
1	9	McLennan	U. S. 84	E55-8-30	755+41-755+66	3	May 26, 1963	320
2	18	Kaufman	S. H. 34	C-173-4	22+00-24+50	18	June 6, 1963	-
3	4	Hartley	U. S. 54	F 608(8)	29+89-30+14	8	June 21, 1963	300
4	11	Sabine	U. S. 96	E 64-6-12	569+75-570+30	11	July 1, 1963	270
6[b]	5	Hale	U. S. 87	F 546(30)	474+90-475+15	7	July 22, 1963	250
7	24	El Paso	U. S. 62 & 180	374-2&3,19&13	768+92-768+67	15	July 23, 1963	275
8	8	Taylor	U. S. 83 & 84	F 90(15)34-1-29	907+00-907+72	7	July 22, 1963	325
9	19	Upshur	U. S. 259	S 75(8)392-2	780+65-781+25	3	July 31, 1963	325
10	12	Montgomery	FM-1314	1986-1-4	908+81-909+06	11	Aug. 6, 1963	270
11	13	Wharton	U. S. 59	C 89-6-19	270-271	6	Aug. 22, 1963	265
12	16	Nueces	I. H. 37	I 137-1(14)001	105+70-106+45	2	Sept. 23, 1963	275
13	20	Hardin	S. H. 105	339-4-10	189+57-189+82	5	Oct. 24, 1963	300
14	14	Williamson	I. H. 35	15-8-43	482+60-482+85	6	Feb. 6, 1964	325
15[c]	9	McLennan	S. H. 31	162-2-2-13	406+75-407	3	May 2, 1964	300
16	21	Hidalgo	U. S. 83	F 308(20)	2594+70-2595+45	16	July 24, 1964	280
17	9	Bell	I. H. 35	C-15-4-22	335+58-335+33	3	Aug. 4, 1964	305
18	21	Hidalgo	U. S. 281	C-255-7-35	403-479	6	Aug. 11, 1964	300

[a] All asphalts used were 85-100 Penetration Grade except those at Sites 17 & 18 which were AC-20 Grade.

[b] Site No. 5 was not evaluated because of technical difficulties.

[c] This surfacing was 0.75 inch thick; all others were about 1.5 inches thick.

TABLE A-2

DENSITY AND PERMEABILITY OF SAMPLES REMOVED FROM PAVEMENT

Site No.	Age of Sample	Density gm/ml 77°F.	Permeability to air, ml/in/min
1	1 day	2.309	103
	4 months	2.372	Impermeable
	1 year	2.374	"
	2 years	2.381	"
	3 years	2.382	"
2	1 day	2.347	36.3
	2 weeks	2.400	2.1
	4 months	2.400	Impermeable
	1 year	2.414	"
3	1 day	2.148	200
	2 weeks	2.158	41
	4 months	2.230	0.4
	2 years	2.258	Impermeable
4	1 day	2.287	1.12
	2 weeks	2.295	0.53
	4 months	2.318	0.61
	1 year	2.326	Impermeable
	2 years	2.263	"
	3 years	2.306	"
6	1 day	2.243	160
	2 weeks	2.292	65
	4 months	2.316	4.5
	1 year	2.325	Impermeable
	2 years	2.329	"
	3 years	2.314	"
7	1 day	2.161	1820
	2 weeks	2.242	272
	4 months	2.273	46
	1 year	2.250	Impermeable
	2 years	2.225	"
	3 years	2.270	"

Table A-2 (Continued)

8	1 day	2.142	2260
	2 weeks	2.202	473
	4 months	2.240	49
	1 year	2.245	11
	3 years	2.240	Impermeable
9	1 day	2.291	236
	2 weeks	2.302	164
	4 months	2.331	40.6
	1 year	2.331	Impermeable
	2 years	2.277	"
	3 years	2.339	"
10	1 day	2.283	129
	2 weeks	2.317	61.5
	4 months	2.337	35
	1 year	2.309	39
	3 years	2.305	Impermeable
11	1 day	2.199	158
	2 weeks	2.254	36
	4 months	2.263	8
	1 year	2.252	8
	2 years	2.284	Impermeable
12	1 day	1.946	269
	2 weeks	1.999	132
	4 months	1.994	39
	1 year	2.114	Impermeable
	3 years	2.089	"
13	1 day	2.243	14
	2 weeks	2.259	1510 (many cracks)
	4 months	2.265	41
	1 year	2.302	13
	3 years	2.310	Impermeable
14	1 day	2.134	1033
	2 weeks	2.131	1380
	4 months	2.141	588
	1 year	2.179	1240
	2 years	2.184	29
	3 years	2.214	Impermeable

Table A-2 (Continued)

16	1 day	2.224	42.3
	2 weeks	2.218	46.6
	4 months	2.286	Impermeable
	2 years	2.319	"
17	1 day	2.282	668
	2 weeks	2.306	285
	4 months	2.307	12.6
	1 year	2.330	Impermeable
	2 years	2.342	"
18	1 day	2.169	42
	2 weeks	2.220	10
	4 months	2.236	Impermeable
	2 years	2.241	"

TABLE A-3

RHEOLOGICAL PROPERTIES OF ORIGINAL AND RECOVERED ASPHALTS

				Sliding Plate Viscometer				Vacuum Capillary Tube				
Site	Producer	Age of Sample	Pen.@ $77^{\circ}F$	Vis.@ $77^{\circ}F$ $S=5x10^{-2}sec^{-1}$ Megapoises	R.V.	Vis.@ $95^{\circ}F$ $S=5x10^{-2}sec^{-1}$ Megapoises	R.V.	Vis.@ $140^{\circ}F$ Stokes	R.V.	Vis.@ $275^{\circ}F$ Stokes	R.V.	Lab. Oxid. R.V.
1	3	Original	84	1.15	-	0.15	-	1830	-	4.35	-	4.2
		Plant	66	1.90	1.65	0.20	1.35	2040	1.1	4.80	1.1	-
		1 day	52	3.15	2.70	0.36	2.4	3610	2.0	5.35	1.2	-
		4 months	39	10.30	8.95	0.55	3.7	6120	3.3	6.20	1.5	-
		1 year	36	10.40	9.10	0.67	4.5	7070	3.9	6.05	1.4	-
		2 years	35.5	10.60	9.25	0.95	6.2	8820	4.8	6.80	1.55	-
		3 years	32.5	15.60	13.50	1.50	9.9	13940	7.6	8.25	1.9	-
2	18	Original	86	0.90	-	0.09	-	1410	-	3.35	-	2.7
		Paver	52	1.90	2.1	0.24	2.7	2450	1.75	4.15	1.25	-
		1 day	44.5	2.80	3.1	0.31	3.4	3200	2.3	4.35	1.3	-
		2 weeks	40.5	3.20	3.6	0.39	4.3	3340	2.4	4.65	1.4	-
		4 months	35	8.35	9.3	0.45	5.0	4510	3.2	4.90	1.45	-
		1 year	32	12.00	13.3	1.06	11.8	8210	5.8	6.00	1.8	-
		2 years*	-	-	-	-	-	-	-	-	-	-
		3 years*	-	-	-	-	-	-	-	-	-	-
3	8	Original	77	0.98	-	0.13	-	2650	-	9.95	-	2.7
		Plant	66	2.30	2.3	0.30	2.3	4340	1.6	12.85	1.3	-
		Paver	60	2.65	2.7	0.35	2.7	4930	1.8	12.70	1.3	-
		1 day	58.5	2.78	2.8	0.34	2.6	4980	1.9	12.30	1.2	-
		2 weeks	53	3.50	3.6	0.54	4.1	6690	2.5	13.55	1.35	-
		4 months	47	5.94	6.0	0.59	4.5	7865	3.0	14.40	1.45	-
		1 year*	-	-	-	-	-	-	-	-	-	-
		2 years	44.5	7.30	7.45	0.62	4.8	9225	3.5	14.70	1.5	-
		3 years*	-	-	-	-	-	-	-	-	-	-
4	11	Original	85	1.17	-	0.13	-	1620	-	2.60	-	2.7
		Plant	66	1.96	1.7	0.23	1.75	2270	1.4	3.20	1.2	-
		Paver	64	2.06	1.75	0.26	2.0	2400	1.5	3.35	1.3	-
		1 day	59	2.36	2.0	0.28	2.15	2520	1.55	3.85	1.5	-
		2 weeks	59	2.56	2.2	0.38	2.9	2790	1.7	3.60	1.4	-
		4 months	40.5	11.40	9.7	0.98	7.5	6130	3.8	4.25	1.6	-
		1 year	39.5	12.40	10.65	1.06	8.2	7570	4.7	4.40	1.7	-
		2 years	33	15.00	12.8	1.85	14.2	15100	9.3	5.50	2.1	-
		3 years	30.5	26.00	22.0	2.05	15.8	24000	14.8	6.50	2.5	-
6	7	Original	79	0.96	-	0.11	-	1150	-	2.45	-	6.35
		Plant	54.5	2.50	2.6	0.22	2.0	1955	1.7	3.10	1.25	-
		Paver	51	2.60	2.7	0.23	2.1	2010	1.75	3.20	1.3	-
		1 day	49	3.00	3.1	0.25	2.3	2215	1.9	3.25	1.35	-
		2 weeks	47	5.35	5.6	0.46	4.2	2500	2.2	3.35	1.4	-
		4 months	27	20.80	21.5	1.06	9.6	5820	5.1	4.40	1.8	-
		1 year	24.5	23.60	25.0	1.52	10.4	7650	6.6	4.70	2.0	-
		2 years	21.5	38.20	40.0	2.34	21.2	11680	10.1	5.00	2.05	-
		3 years	20.0	39.80	41.5	3.70	32.6	22600	19.5	6.40	2.60	-

Table A-3 Continued.

7	15	Original	92.5	1.00	-	0.11	-	1165	-	2.75	-	3.2
		Plant	75	1.75	1.75	0.17	1.55	1660	1.4	3.35	1.2	-
		Paver	66	1.85	1,85	0.24	2.2	2060	1.75	3.55	1.3	-
		1 day	58	2.85	2.85	0.31	2.8	2430	2.1	3.70	1.35	-
		2 weeks	54	4.35	4.35	0.33	3.0	2960	2.5	3.75	1.35	-
		4 months	36.5	13.20	13.2	0.86	7.8	5370	4.6	4.45	1.6	-
		1 year	33.5	14.50	14.5	1.28	11.6	8100	6.9	5.00	1.8	-
		2 years	29	21.50	21.5	2.20	20.0	15110	13.0	5.90	2.15	-
		3 years	29	23.00	23.0	2.50	22.8	18600	16.0	7.00	2.55	-
8	7	Original	80	0.86	-	0.11	-	1160	-	2.40	-	5.1
		Plant	50.5	2.35	2.7	0.34	3.1	1990	1.7	3.00	1.25	-
		Paver	48	2.40	2.8	0.22	2.0	2040	1.8	3.00	1.25	-
		1 day	47	2.80	3.25	0.38	3.5	2200	1.9	3.10	1.3	-
		2 weeks	38.5	6.56	7.6	0.47	4.3	3320	3.0	3.45	1.4	-
		4 months	30.5	12.40	14.3	0.73	6.6	4560	3.9	3.90	1.6	-
		1 year	26	15.00	17.5	1.04	9.5	7390	6.4	4.46	1.85	-
		2 years	20.5	28.6	33.0	2.26	20.5	12380	10.7	4.80	2.0	-
		3 years	18.5	30.0	35.0	3.20	29.0	21120	18.3	6.55	2.7	-
9	3	Original	81.5	1.18	-	0.17	-	1870	-	4.00	-	4.7
		Plant	53.5	2.85	2.4	0.34	2.0	3860	2.1	5.30	1.3	-
		Paver	54.0	2.82	2.3	0.43	2.5	4090	2.2	5.55	1.4	-
		1 day	55.0	2.80	2.3	0.41	2.4	3910	2.1	5.60	1.4	-
		2 weeks	54.5	5.56	4.7	0.48	2.8	4970	2.7	5.59	1.5	-
		4 months	44.5	8.76	7.4	0.84	7.1	6710	3.6	6.40	1.6	-
		1 year	36.5	12.00	10.2	1.44	8.5	10400	5.5	8.83	2.2	-
		2 years	32.5	13.30	11.3	-	-	11190	6.0	-	-	-
		3 years	30.0	17.00	14.4	1.45	8.5	13800	7.4	8.8	2.2	-
10	11	Original	88.5	1.12	-	0.13	-	1660	-	2.85	-	2.7
		Plant	82	1.28	1.15	0.16	1.2	2060	1.25	3.15	1.1	-
		Paver	71	1.76	1.55	0.20	1.55	2370	1.4	3.45	1.2	-
		1 day	68.5	2.10	1.9	0.22	1.7	2630	1.6	3.75	1.3	-
		2 weeks	56	3.20	2.85	0.38	2.9	3560	2.15	4.20	1.5	-
		4 months	41	9.30	8.3	0.76	5.8	6255	3.7	4.45	1.55	-
		1 year	33	15.40	13.8	1.84	14.1	16690	10.0	6.10	2.1	-
		2 years	28	23.00	20.5	2.46	18.9	20200	12.6	6.1	2.1	-
		3 years	25	29.60	27.4	3.50	20.6	28000	16.8	8.4	2.9	-
11	6	Original	99.5	0.565	-	0.053	-	1110	-	2.70	-	3.2
		Plant	62.5	1.45	2.6	0.14	2.6	2020	1.8	3.45	1.25	-
		2 weeks	54	2.05	3.6	0.20	3.8	2440	2.2	3.80	1.4	-
		4 months	41	8.60	15.2	0.34	6.4	3570	3.2	4.20	1.55	-
		1 year	29.5	12.40	22.0	0.63	11.9	5610	5.1	4.75	1.75	-
		2 years	27.5	15.40	27.0	0.71	13.4	6540	5.9	5.00	1.85	-
		3 years**	-	-	-	-	-	-	-	-	-	-
12	2	Original	77	0.86	-	0.088	-	1190	-	2.25	-	3.2
		Plant	68	1.20	1.4	0.116	1.3	1430	1.2	2.80	1.1	-
		Paver	62	1.40	1.6	0.130	1.5	1780	1.5	2.95	1.15	-
		1 day	55	2.05	2.4	0.196	2.2	1995	1.7	3.15	1.25	-
		2 weeks	53.5	2.20	2.6	0.210	2.4	2175	1.8	3.17	1.25	-
		4 months	39.5	4.95	5.7	0.400	4.5	3340	2.8	3.70	1.45	-
		1 year	25.5	13.20	15.3	0.690	7.8	8775	7.4	5.16	2.0	-
		2 years	19.0	25.00	29.0	2.300	26.0	15600	13.0	6.97	2.75	-
		3 years	18.0	28.00	32.5	2.460	28.0	17980	15.0	7.34	2.9	-

Table A-3 Continued.

13	5	Original	90	0.81	-	0.074	-	940	-	2.6	-	2.55
		Plant	59	1.78	2.2	0.200	2.7	1590	1.7	3.3	1.25	-
		Paver	57.5	1.98	2.5	0.200	2.7	1920	2.0	3.4	1.3	-
		2 weeks	52	2.26	2.8	0.244	3.3	1990	2.1	3.5	1.35	-
		4 months	48	4.66	5.75	0.250	3.4	2200	2.3	3.65	1.4	-
		1 year	30	7.96	9.85	0.660	8.9	5690	6.0	4.35	1.65	-
		2 years	26.5	17.80	22.0	1.160	15.7	7790	8.3	5.2	2.0	-
		3 years	23.0	18.60	23.0	1.300	17.6	8775	9.3	5.5	2.1	-
14	6	Original	98.5	0.58	-	0.075	-	1280	-	3.55	-	2.8
		Plant	76.5	1.03	1.8	0.096	1.3	1780	1.4	4.05	1.1	-
		1 day	70.5	1.10	1.9	0.118	1.55	1910	1.5	4.25	1.2	-
		4 months	52.5	2.92	5.0	0.168	2.25	2740	2.1	4.55	1.3	-
		1 year	37.5	6.20	10.6	0.304	4.05	3905	3.0	5.55	1.6	-
		2 years	34.0	7.24	12.7	0.530	7.10	6815	5.3	6.60	1.95	-
		3 years	30.0	11.60	20.0	1.000	13.30	8980	7.0	8.35	2.35	-
15	3	Original	81.5	1.26	-	0.19	-	2390	-	4.2	-	3.35
		Plant	57.0	2.80	2.2	0.57	3.0	4740	2.0	5.5	1.3	-
		1 day	56.0	2.74	2.2	0.45	2.4	4640	1.9	5.2	1.25	-
		2 weeks	42.0	5.40	4.3	0.83	4.4	7240	3.0	6.25	1.5	-
		4 months	33.5	15.00	12.0	1.62	8.5	16640	7.0	8.85	2.1	-
		1 year	27.0	18.00	14.2	2.46	13.0	-	-	9.35	2.2	-
		2 years	22.5	44.00	35.0	4.90	26.0	-	-	17.30	4.1	-
16	16	Original	93.0	0.66	-	0.075	-	875	-	2.22	-	3.40
		Plant	61.0	1.40	2.1	0.118	1.6	1570	1.80	2.85	1.30	-
		Paver	57.0	1.86	2.8	0.158	2.1	1920	2.20	3.14	1.40	-
		1 day	53.0	1.90	2.9	0.172	2.3	2020	2.30	3.17	1.45	-
		2 weeks	43.5	3.30	5.0	0.300	4.0	2920	3.35	3.35	1.50	-
		4 months	36.5	5.92	9.0	0.370	4.9	3660	4.20	3.78	1.70	-
		1 year	30.0	12.60	19.0	0.720	9.6	5910	6.75	4.70	2.10	-
		2 years	27.0	15.50	23.5	0.780	10.4	8750	10.00	6.12	2.75	-
17	3	Original	80.0	1.48	-	0.175	-	2290	-	4.45	-	3.8
		Plant	50.0	3.16	2.1	0.530	3.00	6940	3.0	6.60	1.5	-
		1 day	50.0	3.40	2.3	0.500	2.85	6810	3.0	6.65	1.5	-
		2 weeks	48.0	4.30	2.9	0.510	2,90	6860	3,0	-	-	-
		4 months	40.0	11.40	7.7	0.960	5.50	10570	4.6	7.25	1.6	-
		1 year	31.5	20.80	14.0	1.600	9.10	15830	6.9	9.25	2.1	-
		2 years	26.5	27.80	18.8	2.860	13.3	28000	12.2	12.75	2.8	-
18	6	Original	57.0	1.68	-	0.119	-	2380	-	4.20	-	2.7
		Plant	40.0	3.5	2.1	0.286	2.40	3970	1.65	5.30	1.25	-
		1 day	38.0	4.2	2.5	0,424	3.55	4390	1.85	5.50	1.30	-
		2 weeks	29.5	6.3	3.75	0.510	4.30	5720	2.40	6.10	1.45	-
		4 months	26.0	13.2	7.9	0.880	7.40	9170	3.85	7.15	1.70	-
		1 year	19.0	28.2	16.8	1.560	13.10	11790	4.90	8.05	1.90	-
		2 years	18.0	33.2	19.8	2.060	17.80	14800	6.20	10.40	2.50	-

*Contaminated sample.

**Surface sealed prior to 3 years of service.

TABLE A-4

200 GRAMS OF ASPHALT 11, AC-20 BLENDED WITH 265 GRAMS OF VARIOUS MINERAL POWDERS PASSING 200 AND RETAINED ON 270 SIEVES. MIXTURES HEATED IN CLOSED CANS IN AIR OVEN FOR 2 MONTHS AT 113°F. ASPHALT EXTRACTED, RECOVERED AND TESTED

	Original Asphalt		Recovered Asphalt After 2 Months		
Type of Mineral	Pen @ 77°	Megapoises @ 77°F	Pen @ 77°	Megapoises @ 77°F	R. V.
None	76.5	1.6	63	2.60	--
Pure Silica	--	--	60	2.85	1.10
New Reef Oyster Shell	--	--	60	2.30	0.90
Old Reef Oyster Shell	--	--	56.5	2.95	1.15
Iron Ore Soil Lot 10	--	--	60.3	2.60	1.00
New Oyster Shell Lot 11	--	--	58.5	2.80	1.05
New Oyster Shell Lot 12	--	--	60.0	2.60	1.00
Dolomite Lot 13	--	--	57.0	2.65	1.02
Porous Limestone Lot 14	--	--	60.5	2.40	0.92

TABLE A-4 (CONT'D)

200 GRAMS OF ASPHALT 6, AC-20 BLENDED WITH 265 GRAMS OF VARIOUS MINERAL POWDERS PASSING 200 AND RETAINED ON 270 SIEVES. MIXTURES HEATED IN CLOSED CANS IN AIR OVEN FOR 6 MONTHS AT 113°F. ASPHALT EXTRACTED, RECOVERED AND TESTED

	Original Asphalt		Recovered Asphalt After 6 Months		
Type of Mineral	Pen @ 77°	Megapoises @ 77°F	Pen @ 77°	Megapoises @ 77°F	R. V.
None	44.0	2.3	43.5	3.0	--
Pure Silica	--	--	46.0	3.1	1.05
New Reef Oyster Shell	--	--	41.5	3.0	1.00
Old Reef Oyster Shell	--	--	43.5	3.0	1.00
Iron Ore Soil Lot 10	--	--	37.5	3.4	1.15
New Oyster Shell Lot 11	--	--	43.0	2.8	0.95
New Oyster Shell Lot 12	--	--	35.5	3.2	1.05
Dolomite Lot 13	--	--	37.0	3.2	1.05
Porous Limestone Lot 14	--	--	40.5	3.2	1.05

*Original Asphalt After Heating 6 Months at 113°F but Not Dissolved and Extracted (Hot Sample Was Stirred Before Testing) Had a Viscosity of 3.7 Megapoises @ 77°F, R.V. = 1.6.

TABLE A-5

OXYGEN CONTENT OF ORIGINAL 85-100 PENETRATION ASPHALTS BY ACTIVATION AND CHEMICAL ANALYSES

Supplier No.	Samples Tested	Percent Oxygen by Activation	Percent Oxygen by Chemical	Percent Increase by Chemical Compared to Activation Analysis
11	2	1.05	1.13	7.5
7	2	0.80	0.94	17.5
8	1	0.50	0.70	40.0
6	2	0.55	1.07	95.0
5	1	0.45	1.05	113.0
3	2	0.30	0.91	200.0

TABLE A-6

RESEARCH REPORTS ISSUED FOR RESEARCH STUDY NO. 2-8-59-9, AREA 8 (FORMERLY PROJECT 15) FROM FEBRUARY, 1961 THROUGH OCTOBER, 1967

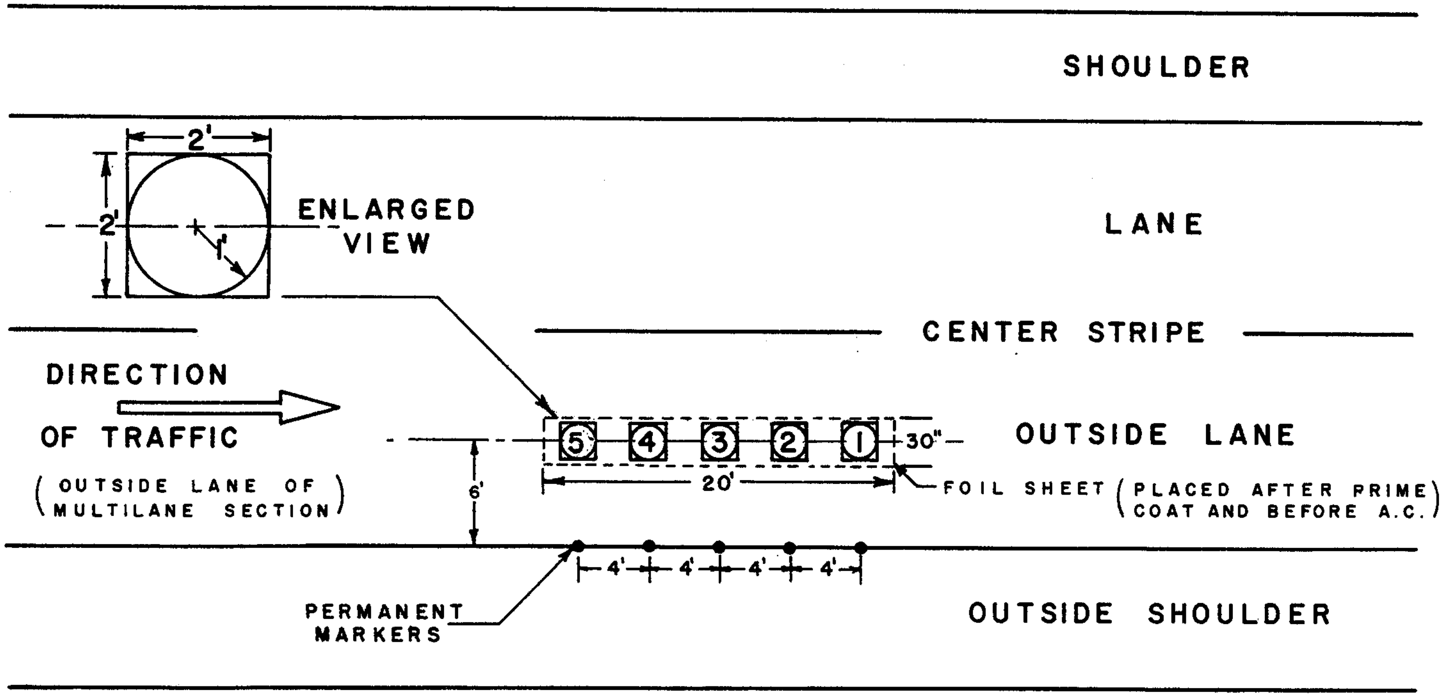
Report No. 1	"Hardening of Paving Asphalts and Relation to Composition," February, 1961.
Report No. 2	"Temperature -Viscosity Data on Asphalt Cements," September, 1962.
Report No. 3	"Antioxidants for Paving Asphalts," September, 1962.
Report No. 4	"Susceptibility of Paving Asphalts to Hardening by Heat, Oxygen and Sunlight," October, 1962.
Report No. 5	"Correlation of Composition with Rheology and Durability of Asphalts," December, 1962.
Report No. 6	"Viscosity Data for Asphalts Used by the Texas Highway Department," July, 1963.
Report No. 7	"Oxidation (Durability) Tests on Asphalts Used by the Texas Highway Department," August, 1963.
Report No. 8	"Hardening of 85-100 Penetration Asphalt Cements During Service in Pavement," May, 1965.
Report No. 9	"Additional Rheological Properties of 85-100 Penetration Asphalt Cements Used in Field Tests Initiated During 1963-64," August, 1966.
Report No. 10	"Oxygen Consumption by Asphalt Films and Resulting Viscosity Changes," March, 1967.
Report No. 11	"Hardening of Asphalt Cements During Service in Pavements, 1963-66 Program," October, 1967.

SCHEMATIC SAMPLING LAYOUT

INSTRUCTIONS FOR OBTAINING RESEARCH SAMPLES OF ASPHALTIC CONCRETE

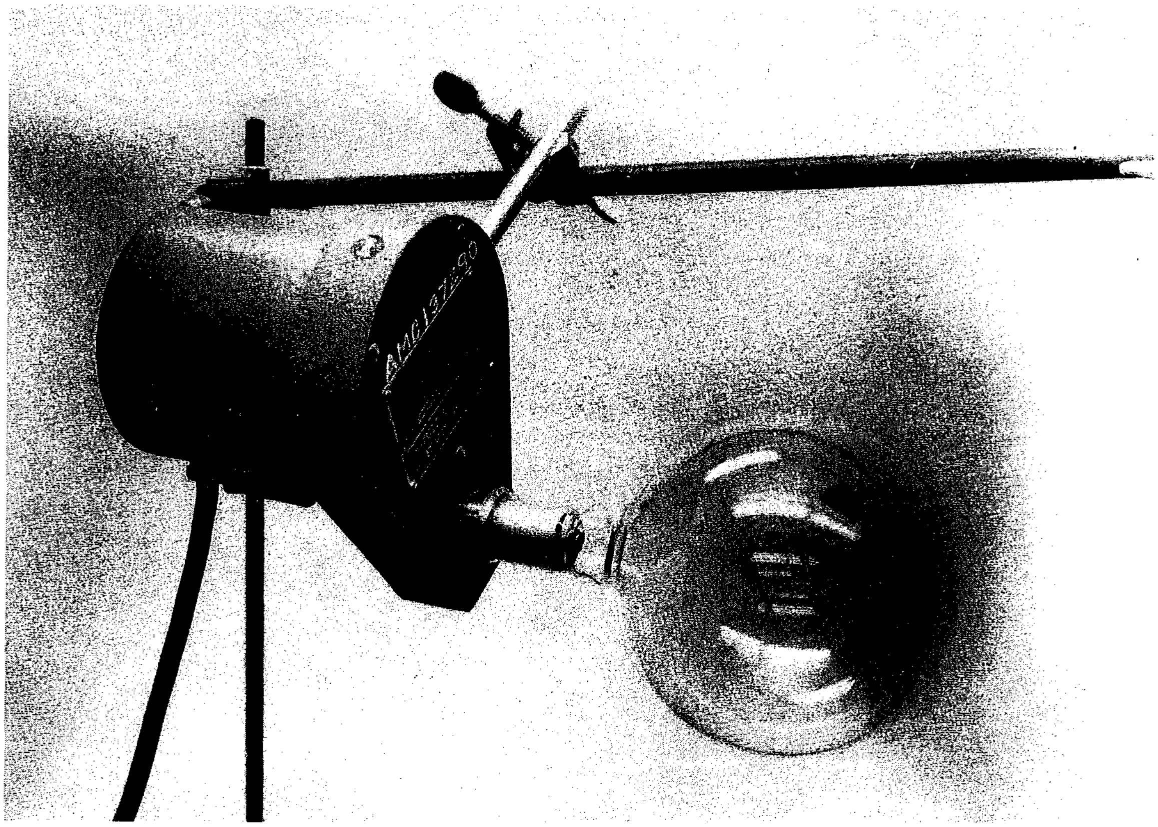
T.T.I. RESEARCH STUDY 2-8-59-9

FIGURE A-I



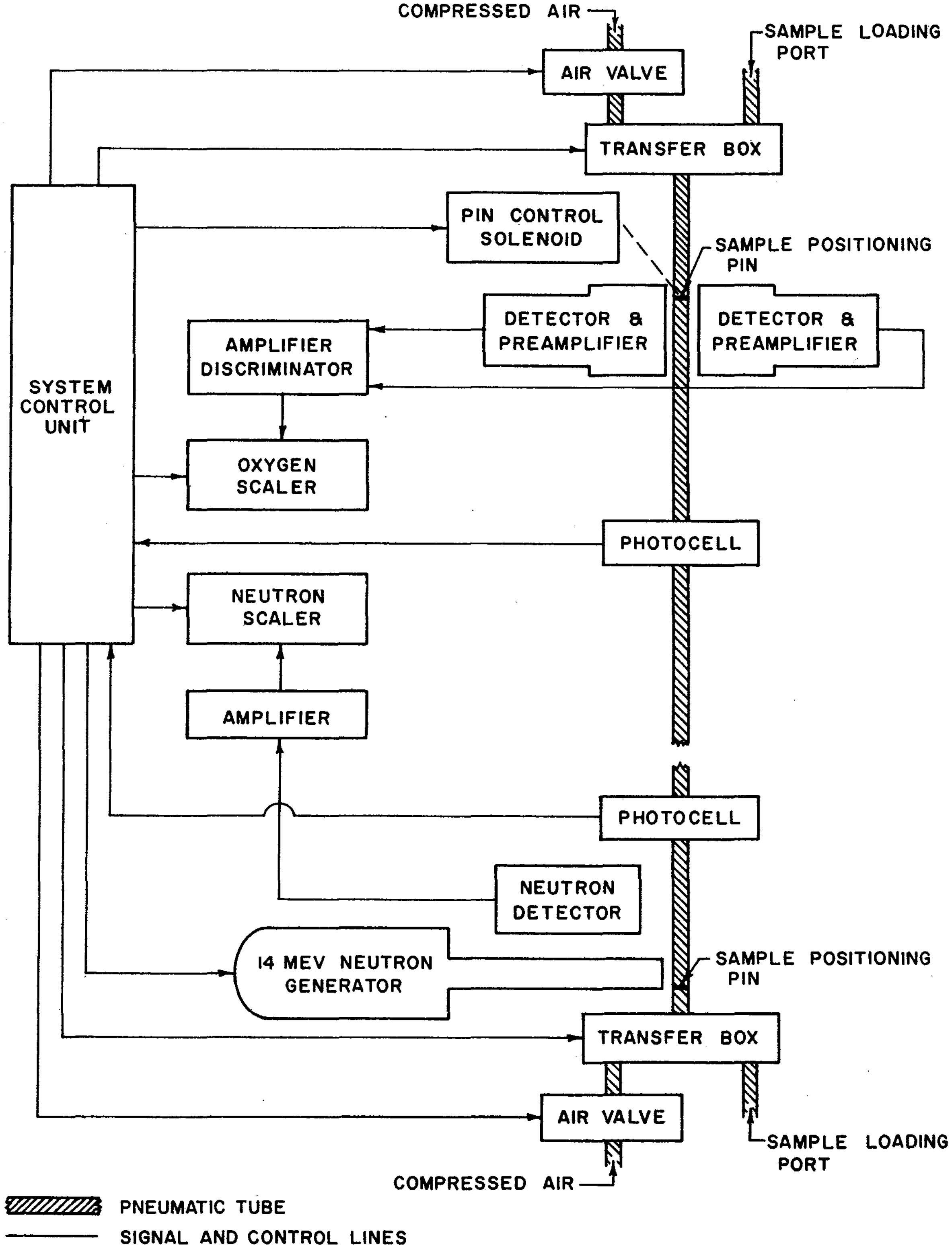
TEST SECTION SHOULD BE:

1) LEVEL SECTION OF ROAD
2) STRAIGHT ALIGNMENT
3) AWAY FROM AN INTERSECTION
4) SAMPLES SHOULD BE TAKEN IN THE SEQUENCE SHOWN
5) PATCHING SHOULD BE LIMITED TO THE TEST HOLE ONLY
6) TEST <u>AREA</u> SHOULD NOT BE OVERLAID UNTIL <u>ALL</u> SAMPLES ARE TAKEN



ROTARY FLASH EVAPORATOR

FIGURE A-II



BLOCK DIAGRAM OF NEUTRON ACTIVATION ANALYSIS SYSTEM

FIGURE A-III