

Reaction Rates and Hardening Susceptibilities as Determined from Pressure Oxygen Vessel Aging of Asphalts

C. K. LAU, K. M. LUNSFORD, C. J. GLOVER, R. R. DAVISON,
AND J. A. BULLIN

Many attempts have been made to simulate road aging with laboratory tests. These have generally not been successful. One reason is that other causes of road failure, such as heavy traffic or construction deficiency, can obscure the effect of the asphalt binder. But other reasons are that an adequate model relating laboratory-measured properties to changes in the road is not available and that the oven test temperatures that have been used may be too high to relate oven aging to road aging. Use of the lower-temperature pressure oxygen vessel offers promise of approximating road conditions. It is shown that no test run at a single elevated temperature can be used to simulate road aging and that at road temperature the rate is still too low for practical testing. It is also shown, however, that the oxidative aging mechanism is constant at temperatures up to 82.2°C and that it approximates aging of the asphalt binder in laboratory cores. If this is borne out with road cores, extrapolation of higher temperature data may provide more accurate predictions of road aging.

From the standpoint of understanding and predicting asphalt performance, the properties of aged asphalt are more important in many respects than the properties of the original material. Years after placement, when a roadway is deteriorating, the asphalt properties may be greatly different from the original properties, and, as a result, if failure is due to the asphalt cement, it will be the properties of this aged material rather than those of the original that are causing the problem. This has been recognized for many years, and many test procedures have been devised to predict aging.

Aging really occurs in two modes: first, there is the very rapid aging that occurs in the hot-mix plant at high temperature on the aggregate surface. This is followed by the much slower, low-temperature aging in the road. It is likely that the mechanisms in these modes of aging are quite different. This is borne out by the persistent failure of tests that were developed to predict hot-mix aging to predict road aging accurately.

There are now two widely recognized hot-mix aging procedures: the thin film oven test (TFOT), ASTM D1754-83, and the rolling thin film oven test (RTFOT), ASTM D2872-80. Numerous variations have been made on the TFOT and RTFOT methods with many intended to simulate road aging as well. Vallerga et al. (1) proposed tilting the oven used for the TFOT and provided an effect similar to that of the RTFOT.

The recognition that thinner films may be desired to simulate hot-mix aging led to the development of microfilm tests (2,3). Thinner films correspond better to actual pavement mixtures, harden faster, and lessen any diffusion effects that may be present in thicker films. The microfilm durability test of Griffin et al. (4) was designed to simulate road aging but has been correlated with aging from the hot-mix process as well as road aging (5,6).

Schmidt and Santucci (7) developed a rolling microfilm test in which benzene-dissolved asphalt was cast in bottles and aged in a manner similar to that used in the RTFOT. The modification of adding a circulating fan, as suggested by Schmidt (8), has been maintained and is called the rolling microfilm circulating (RMFC) test. Schmidt (9) has also used several modifications to the rolling microfilm test at various temperatures and rates of air circulation as well as in the presence of aggregate in an effort to simulate hot-mix and road aging.

Kemp (10) studied several durability tests and their relationship to field hardening. Kemp stated that no laboratory process seemed to predict performance satisfactorily. The void content of the mix is the only parameter that seems to give any prediction of the aging process for pavements with more than 50 months of service. This leads to the conclusion that voids must be constant before an aging test can predict actual field hardening. The best correlation with the field data came from the RMFC test for 48-hr exposures. The RMFC test was modified by Petersen (11). The test is run at 112.8°C using a larger sample and is purported to represent about 10 years of field aging.

Evidence exists that aggregate can catalyze aging (12-17). Jennings (12) studied the aging process using gel permeation chromatography (GPC). Jennings stated that the change in the molecular size distribution of an asphalt due to aging is a function of aggregate, additives, exposure, and time.

The most realistic road aging procedures appear to be the low-temperature (compared with the hot-mix temperatures) pressure oxygen vessel (POV) tests, in which lower temperatures can be partly compensated for with respect to required test duration by higher oxygen pressure, and there are several reports using this kind of device. Lee (18) simulated the hot-mix step first by TFOT aging. This material, still in the TFOT dish, was placed in a pressure vessel and aged at 65°C at various pressures. A standard procedure was proposed by Lee (19) in which TFOT residues were aged at 65°C and 300 psi for 1,000 hr. They used a variety of criteria to correlate aging

including viscosity, penetration, softening point, microductility, and percent asphaltenes. Though the results are promising, the rankings of the POV-aged asphalts by each criterion were not very consistent with the rankings of the field-aged asphalts, indicating that actual road aging was not being duplicated. They also found an overwhelming effect of voids that complicated relating properties to age. Jamieson and Hattingh (20) reported that POV results using 7-mm films at 65°C and 300 psi did not agree with road performance.

Edler et al. (21) reported results in which 30- μ m asphalt films were contacted with oxygen at 300 psi at 65°C for 96 hr, but the results were not compared with field data. Kim et al. (22) aged compacted cores at 100 psi and 60°C up to 5 days. Comparison with field cores was on the basis of the resilient modulus and Corbett analysis of extracted material. There was generally fair agreement between the laboratory and field cores for two of three sets of cores, but they reported some problems with diffusion into cores of low air voids. It is likely that much of this work is complicated by diffusion of oxygen into cores and problems with extraction and recovery (23) of the asphalt binder from laboratory and field cores.

OBJECTIVES

As indicated, most road aging tests, which are lower-temperature variations of the oven tests that were designed to simulate the hot-mix process, do not appear to correlate to road aging. One likely reason is that the tests are still conducted much above field temperatures, so that the aging rates and mechanisms are not relevant to field aging. Another difficulty with correlation is that an adequate model relating laboratory-measured properties to changes in the road is not available.

The primary objective of this work was to determine the significance of aging temperature with respect to oxidation rate and mechanism. By so doing, a rationale would exist for designing a laboratory test that would be optimal with respect to the ease and time to conduct the test and with respect to the ability of the test to indicate or predict road aging.

APPARATUS AND PROCEDURES

The POV and sample holder designed for this work are shown in Figures 1 and 2. The vessel is made of 4-in.-diameter standard stainless steel pipe. It is 2 ft long and flanged at each end. The top flange is equipped with a 1½ in., 400-psig rupture disk with the operating pressure in the tests limited to 300 psia. The pressure vessel is wrapped with heating tape, which with a variable transformer is used to control the temperature. A thermocouple is inserted through the bottom flange to detect the temperature, which is read from a digital indicator. A control panel houses the temperature indicator as well as vacuum and pressure gauges. The pressure vessel is located behind a metal shield. All control valves are specially cleaned and teflon coated for oxygen service. Valves and tubing as shown in Figure 1 are stainless steel. The sample holder is also stainless steel. It fits inside the pressure vessel and can support 88 asphalt samples.

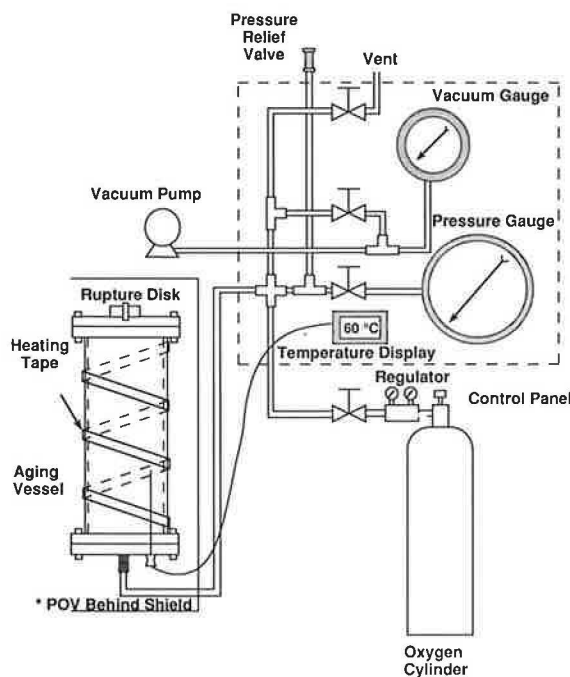


FIGURE 1 POV apparatus.

Asphalt samples for the POV oxidation test are prepared by depositing about 20 g of molten asphalt between one sheet of polyvinylchloride and one sheet of triacetate. The asphalt is then pressed between two glass plates until the asphalt between the plastic sheets formed a 6-in. circular disk to obtain a film thickness of about 1000 μ m. If the asphalt cools down to the point that the pressing process becomes difficult, the whole setup is heated in an oven at a temperature of about 90°C to 100°C until the asphalt becomes soft again. A thinner film can be obtained by pressing the asphalt into a bigger disk if desired. The composite sheet is then cooled to room temperature and is cut into rectangular pieces of 1.5 in. by 2.5 in. The plastic sheets are removed from the asphalt film after

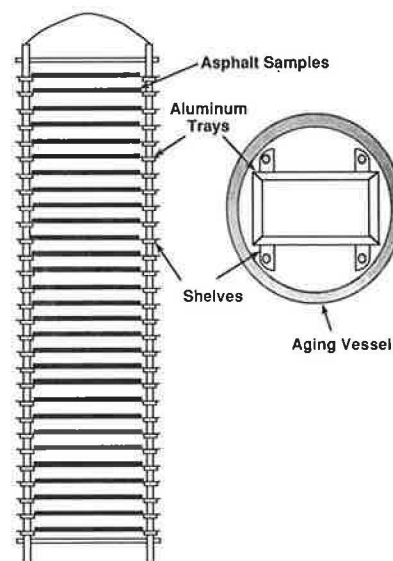


FIGURE 2 Sample holder for POV.

cooling the composite film in a freezer. The asphalt films are then deposited in 1.5-in. by 2.5-in. aluminum trays and are ready for the POV experiment.

Asphalt samples are placed in the stainless steel sample holder. The sample holder is placed inside the aging vessel and the flange is bolted down securely. With the venting valve, oxygen feed valve, and high-pressure gauge valve closed and the vacuum valve opened, a vacuum is applied to the aging vessel to remove the air inside. The vacuum valve is then closed. The vessel is slowly pressurized to 300 psia oxygen from an oxygen cylinder with the oxygen feed valve and high-pressure gauge valve open. When the desired oxygen pressure is reached, the oxygen feed valve is closed. Power to the heating tape is then turned on to heat the pressure vessel to the desired aging temperature. During the heating process, excess pressure above the aging pressure can be vented off by opening the venting valve momentarily. When the desired period of oxidation time is attained, oxygen in the vessel is slowly vented to the atmosphere. Asphalt samples are taken from the vessel for further analysis at specified time intervals. The short time required to depressurize, remove the samples, and return to operating pressure and temperature is small compared with the total aging time.

Infrared analyses were made of all samples with a Nicolet 60 SXB Fourier transform infrared spectrometer (FT-IR) using an attenuated total reflectance (ATR) procedure described by Jemison et al. (24). The growth in the carbonyl band (from 1820 cm^{-1} to 1650 cm^{-1}) was used as a measure of oxidation. Dynamic viscosities were measured at several temperatures using a dynamic mechanical spectrometer RMS-800 RDSII from Rheometrics Inc. Measurements were made with 0.984-in. (25-mm) parallel plates operating in a dynamic mode with a 0.0394-in. (1-mm) gap. Operation and data acquisition are monitored by an IBM PS-2 Model 30 computer with a RECAPIII computer analysis package, also from Rheometrics Inc. The complex dynamic viscosities were measured at low enough frequencies (at 0.1, 0.01, or 0.005 sec^{-1} , depending on the asphalt stiffness) to approach the low-frequency limiting values designated as η_0^* .

RESULTS AND DISCUSSION

It was expected that the failure of aging procedures to represent road aging resulted primarily from using temperatures far above those experienced by the asphalts in road service, but the question remained how high a temperature could be used while still being representative of road aging. Experiments in the POV at 25°C showed extremely low aging rates, and it was obvious that some compromise must be made with higher temperatures.

To study this effect, five asphalts (AC-20, viscosity grade) were aged in the POV at 300 psia oxygen pressure at temperatures of 60°C, 71.1°C, and 82.2°C, and one asphalt, Ampet, was aged at 93.3°C. Samples were withdrawn periodically for viscosity measurements and infrared analysis. A typical infrared spectrum is shown in Figure 3. Figure 4 shows the limiting complex dynamic viscosity, η_0^* , measured at 60°C (140°F) versus aging time for Ampet at all four aging temperatures. The limiting complex dynamic viscosity, η_0^* , is defined as the limiting, essentially frequency-independent value

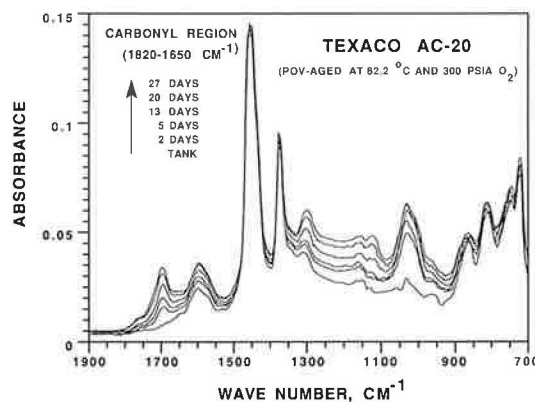


FIGURE 3 Typical FT-IR spectra using the ATR method for asphalts aged in the POV for various lengths of time.

that is reached at sufficiently low frequencies. The range of shear rate defining η_0^* is material dependent. The linear relation of $\log \eta_0^*$ versus aging time is characteristic of all asphalts studied, though with different slopes and intercepts. Figure 5 shows a plot for the same asphalt of carbonyl growth (the area increase above the neat asphalt between 1820 to 1650 cm^{-1}) versus time exhibiting a constant rate of carbonyl growth.

As expected, a cross plot of η_0^* versus carbonyl is linear (Figure 6). It can be seen that data from different aging temperatures combine to give a single line, strongly indicating that whereas the rate of oxidation changes with temperature, the mechanisms of oxidation do not. All five asphalts gave similar results as shown in Figure 7.

The linear relation between \log viscosity and carbonyl formation was reported previously by Martin et al. (25) for asphalt extracted and recovered from road-aged asphalt. Jemison et al. (26) showed that it also applied to TFOT and RTFOT aging, but the function was not the same as that resulting from hot-mix plant aging. The POV data further confirm that this is a universal characteristic of asphalt oxidation for each asphalt as long as the aging mechanism is unchanged.

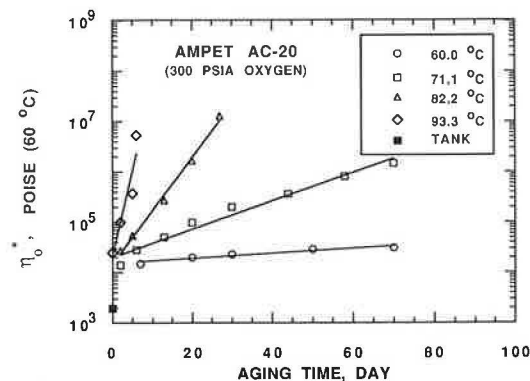


FIGURE 4 Dynamic viscosity hardening of an Ampet AC-20 asphalt with time at four temperatures.

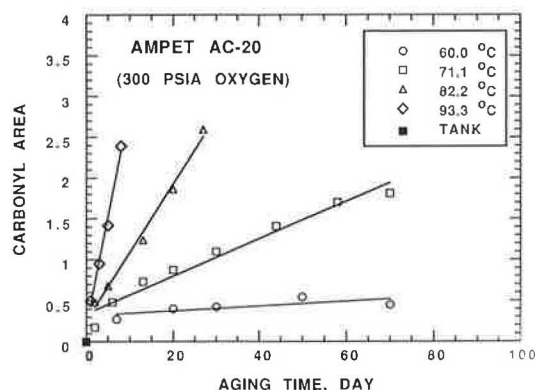


FIGURE 5 Growth of the infrared carbonyl band with time at four temperatures.

This relation also appears to hold for viscosity measurements over a wide range of temperatures. Figure 8 shows log viscosity at other temperatures versus carbonyl area. This of course means that changes in viscosity temperature susceptibility are independent of the aging temperature within this range, further confirmation of a constant aging mechanism.

This does not necessarily mean that the aging mechanism of the neat asphalt in the POV is the same as that on the road. The results of Jemison et al. (26) indicate that, at least in the hot-mix plant, the aggregate may play a role. Tables 1 and 2 give results obtained with two asphalts used in the Strategic Highway Research Program (SHRP). The asphalts were aged in a laboratory hot-mix process in the presence of aggregate (either SHRP RL or RB), compacted into cores, and then aged either in a POV or in a forced-draft oven (27). The aging times and temperatures are given in the table. The cores were then subjected to physical property tests (not the subject of this paper) and subsequently to asphalt extraction and recovery using a recently developed procedure designed to minimize solvent aging of the asphalt and to maximize asphalt extraction and subsequent solvent removal (23,28,29). The recovered asphalt was compared with neat samples of the same asphalts after aging in the POV at 93.3°C and 300 psia oxygen (Figures 9 and 10). As can be seen, the differences, if any, are small, indicating little difference be-

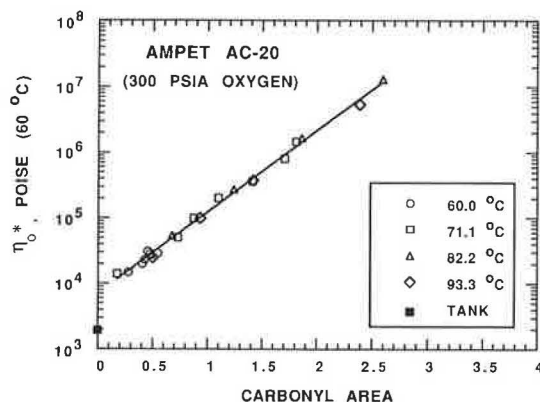


FIGURE 6 Hardening of the dynamic viscosity with growth of the carbonyl band at three temperatures.

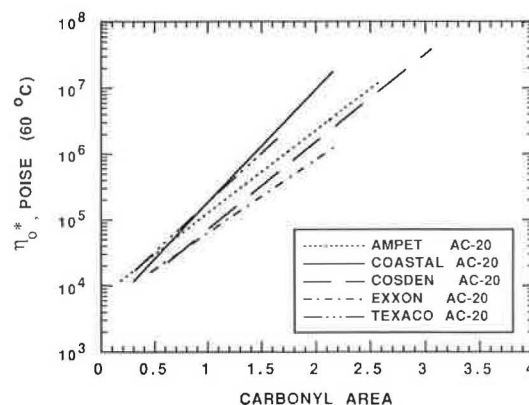


FIGURE 7 Comparison of the hardening susceptibilities of five asphalts.

tween POV aging of neat asphalt and core aging at temperatures below about 93.3°C.

The SHRP AAG-1 asphalt is remarkable in that when oxidized to the same extent as the other asphalts, the viscosity has not increased nearly as much. For example, when the carbonyl area of AAG-1 has increased by 2 units, then it has hardened to a viscosity of about 30 kPoise (Figure 9). On the other hand, for the same carbonyl growth, AAK-1 (Figure 10) has hardened to about 10,000 kPoise. There are very dramatic differences.

These results of Figures 4 through 10 show that hardening as the asphalt ages involves two important and independent factors. The first, the slope of the log viscosity versus carbonyl area plot, as in Figures 6 and 7, is a measure of the sensitivity of the asphalt viscosity to carbonyl formation. This is called the hardening susceptibility. The second, the slope of the carbonyl area versus time plot, as in Figure 5, is the rate of oxidation, which, as seen in Figure 5, is a strong function of temperature. An asphalt may oxidize relatively rapidly but not harden a great deal, compared with other asphalts, or it may harden considerably while oxidizing at a slower rate. Which situation holds can be an important consideration in assessing an asphalt's durability.

There is considerable evidence that the hardening susceptibility is primarily a function of compatibility. Asphalt AAG-

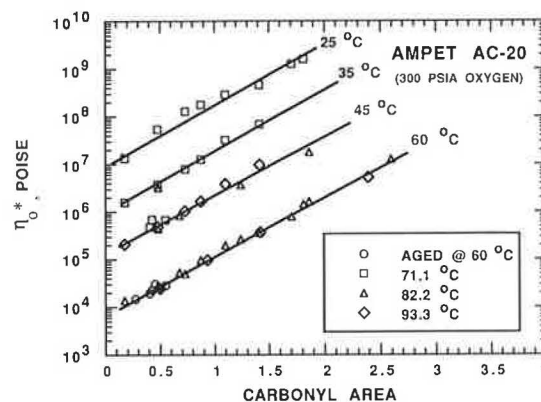


FIGURE 8 Dynamic viscosity versus carbonyl area for a single asphalt at four temperatures.

TABLE 1 LIMITING COMPLEX DYNAMIC VISCOSITY AND CARBONYL AREA FOR SHRP ASPHALT AAK-1 FOR DIFFERENT AGING CONDITIONS

Sample ^a Aggregate	Air Voids (%)	Pre-Aging		Aging		Oven ^b	Properties		Carbonyl Area
		Time (days)	Temp. (°C)	Time (days)	Temp. (°C)		η_o^* (25°C, 0.01s ⁻¹) (10 ⁶ poise)	η_o^* (60°C, 0.1s ⁻¹) (10 ³ poise)	
Neat	---	0	---	0	---	---	1.987	3.992	0.000
RL	9.20	2	40	0	---	---	11.98	20.00	0.209
RL	9.10	2	40	7	107	---	270.2	1976.	1.350
RB	8.00	2	40	0	---	---	11.16	17.36	0.229
RB	8.90	2	40	7	107	---	183.4	868.0	1.318
RB	6.57	2	60	0	---	---	11.19	17.92	0.271
RB	6.42	2	60	7	107	---	150.9	560.9	1.071
RL	---	0	---	2	60	300	30.47	46.86	0.378
RL	---	0	---	7	25	300	15.45	34.43	0.291
RB	---	0	---	7	60	100	40.46	62.34	0.537
Neat	---	0	---	0.5	92.2	300	60.00	88.00	0.568
Neat	---	0	---	1.0	92.2	300	170.0	300.0	0.933
Neat	---	0	---	2.0	92.2	300	---	40,000	2.736

^a Aggregates are SHRP RB or RL.

^b The numbers indicate O₂ pressure in the POV. Where no number is given, the aging was done in the FDO.

TABLE 2 LIMITING COMPLEX DYNAMIC VISCOSITY AND CARBONYL AREA FOR SHRP ASPHALT AAG-1 FOR DIFFERENT AGING CONDITIONS

Sample ^a Aggregate	Air Voids (%)	Pre-Aging		Aging		Oven ^b	Properties		Carbonyl Area
		Time (days)	Temp. (°C)	Time (days)	Temp. (°C)		η_o^* (25°C, 0.01s ⁻¹) (10 ⁶ poise)	η_o^* (60°C, 0.1s ⁻¹) (10 ³ poise)	
Neat	---	0	---	0	---	---	2.448	2.500	0.000
RL	7.91	2	60	0	---	---	13.80	8.334	0.490
RL	7.54	2	60	7	107	---	165.0	83.95	2.071
RB	---	0	---	2	60	300	22.43	12.00	1.165
RB	---	0	---	7	25	300	11.78	8.132	0.940
RL	---	0	---	7	60	100	28.00	13.77	1.278
Neat	---	0	---	0.5	92.2	300	35.00	16.00	1.369
Neat	---	0	---	1.0	92.2	300	54.00	19.00	1.682
Neat	---	0	---	3.0	92.2	300	130.0	45.00	2.527
Neat	---	0	---	5.0	92.2	300	164.0	64.00	2.831
Neat	---	0	---	8.0	92.2	300	250.0	106.0	3.228

^a Aggregates are SHRP RB or RL.

^b The numbers indicate O₂ pressure in the POV. Where no number is given, the aging was done in the FDO.

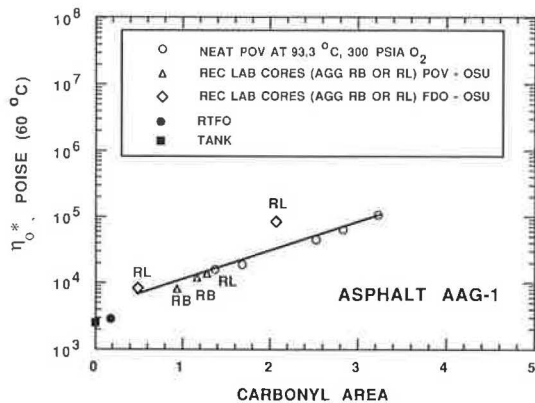


FIGURE 9 Hardening susceptibility for SHRP Asphalt AAG-1 as determined by both POV aging of neat asphalt and either POV or forced draft oven aging of laboratory-prepared cores. (L and B symbols refer to SHRP Aggregates RL and RB.)

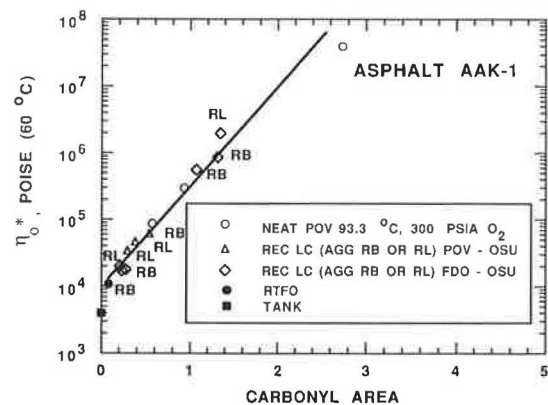


FIGURE 10 Hardening susceptibility for SHRP Asphalt AAK-1 as determined by both POV aging of neat asphalt and either POV or forced draft oven aging of laboratory-prepared cores. (L and B symbols refer to SHRP Aggregates RL and RB.)

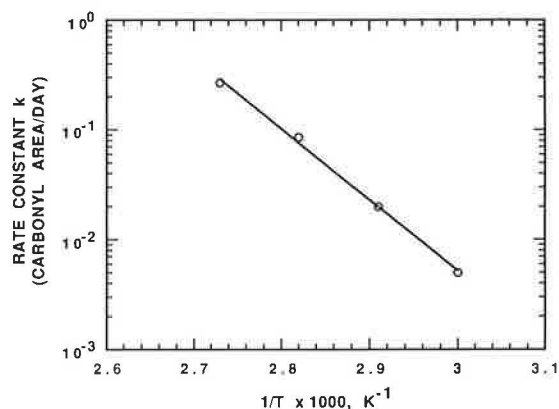


FIGURE 11 Arrhenius plot for the Ampet AC-20 asphalt.

1, for instance, is exceptionally low in both saturates and asphaltene content, which are mutually insoluble. Work by Stegeman et al. (30) has shown that the hardening ratio before and after RTFOT aging can be significantly reduced by separating asphalt into fractions on the basis of solubility and then formulating asphalts from the more mutually soluble fractions.

Chemical reaction rate constants generally vary as the exponent of the reciprocal absolute temperature. The rate of reaction is then given by this constant times a function of reactant concentrations. At the conditions in the POV, oxygen pressure is constant, and the actual oxygen uptake by the asphalt is so small compared with the number of available reaction sites that effectively the reactant concentrations are constant. If the rate of oxidation can be considered proportional to the growth rate in carbonyl are, then at temperature T

$$\frac{dA}{d\theta} \sim k \sim \exp(-E/RT)$$

where

- A = carbonyl area,
- k = the rate constant,
- θ = time,
- E = a constant energy of activation unique to the reaction taking place, and
- R = gas constant (1.987 cal/g mol K).

From this equation

$$\ln k \sim \frac{E}{R} \left(\frac{1}{T} \right)$$

and hence, one would expect a plot of $\ln k$ versus $(1/T)$ to be linear.

Figure 11 shows this expected linear relationship for the Ampet asphalt and Figure 12 shows it for all five asphalts. The striking thing is not that the rates are different for each asphalt but that the slopes are different. This suggests that asphalts having different slopes are reacting with oxygen by different mechanisms. As can be seen, some asphalts that

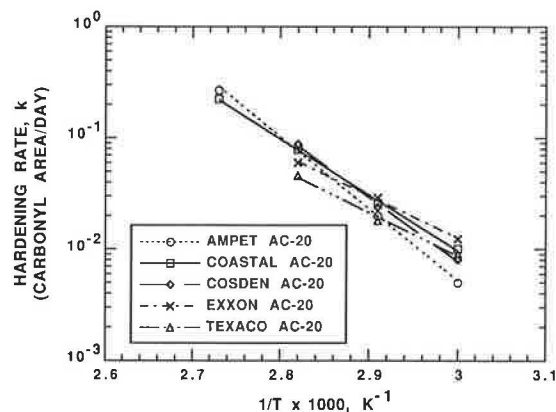


FIGURE 12 Comparison of the Arrhenius plots for the five asphalts studied. Note presence of crossover points where the relative aging rates of two asphalts interchange.

show higher rates of oxidation at elevated temperatures actually have lower oxidation rates at temperatures expected during life on the road. This reversal can carry over to physical properties as well, as shown in Figure 13, which shows the oxidative hardening of the Ampet and Texaco asphalts at 60°C and 80°C. At the lower temperature the Ampet asphalt hardens more slowly, whereas at the higher temperature the Texaco hardens more slowly. Thus no test run at a single elevated temperature is valid as a measure of expected road performance.

On the other hand, by running the POV at several temperatures, the resulting linear relations can be extrapolated to a realistic average road temperature or integrated over a typical road temperature history. To convert the rate data into a hardening (or other physical property) prediction, extrapolated rates, as could be obtained from Figure 12, can be combined with hardening susceptibilities (or analogous physical-

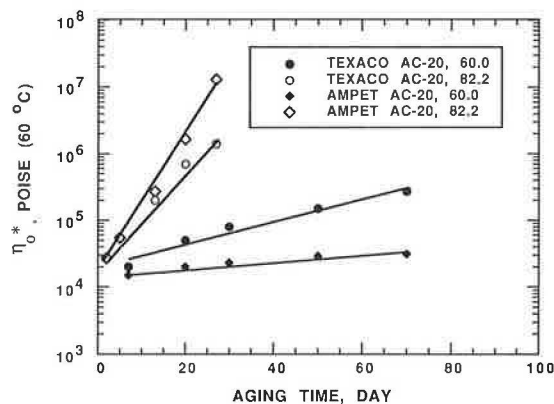


FIGURE 13 Comparison of the hardening of the Texaco and Ampet asphalts with time at two POV temperatures. At the lower temperature (140°F) the Ampet hardens more slowly, whereas at the higher temperature (180°F) the order is reversed and the Texaco hardens more slowly. The differences are significant, and relative aging of the two asphalts predicted on the basis of the higher temperature data would be grossly in error at highway conditions.

chemical property relation), such as those in Figure 7. In this way the relative expected hardening rates could be calculated from the expected road temperature profile.

CONCLUSIONS

POV aging (300 psia O₂) of asphalt films (1000 μ m thick) and subsequently measured FT-IR (ATR) spectra and complex dynamic viscosities have led to the following conclusions:

1. The degree of oxidation, as measured by the increase in the carbonyl peak area relative to the unaged neat asphalt, increases linearly with oxidation time at a given temperature, over a range from 60°C to 82.2°C (at least). The slope of this relation is indicative of the chemical reaction rate.
2. The degree of hardening due to oxidation increases essentially linearly with time and, hence, is linearly related to the carbonyl peak area. The hardening susceptibility is a key measure of the effect of oxidative aging on a physical property. This effect is probably related to the compatibility of the asphalt.
3. Different asphalts exhibit different reaction rates at the same temperature and, just as important, exhibit different activation energies (different slope of $\ln k$ versus $1/T$). As a result, one asphalt may age faster than another at one temperature, whereas it may age slower at a different temperature. This crossover in aging rate is of profound significance in designing an asphalt road aging test. Such a test must be conducted at more than one temperature to accurately predict pavement aging.
4. Asphalts aged in contact with two different aggregates suggest that the reaction mechanisms are not significantly changed by the aggregate up to even 107°C.

ACKNOWLEDGMENTS

Support for this work by the State Department of Highways and Public Transportation (SDHPT, Texas), in cooperation with the U. S. Department of Transportation and the Federal Highway Administration, is gratefully acknowledged. Helpful discussions with Don O'Connor and Darren Hazlett of SDHPT and the technical contributions of Penny Bartnicki are greatly appreciated.

The authors are grateful to Chris Bell of the Oregon State University Transportation Research Institute and the Strategic Highway Research Program for making available the aged asphalt-aggregate mixes.

REFERENCES

1. B. A. Vallerger, C. L. Monismith, and K. Grantham. A Study of Some Factors Influencing the Weathering of Paving Asphalts. *Proc. AAPT*, Vol. 26, 1957.
2. D. F. Fink. Research Studies and Procedures. *Proc. AAPT*, Vol. 27, 1958.
3. R. N. Traxler. Changes in Asphalt Cements During Preparation, Laying, and Service of Bituminous Pavements. *Proc. AAPT*, Vol. 36, 1967.
4. R. L. Griffin, R. K. Miles, and C. J. Penther. Microfilm Durability Test for Asphalt. *Proc. AAPT*, Vol. 24, 1955.
5. J. J. Heithaus, and R. W. Johnson. A Microviscometer Study of Road Asphalt Hardening in the Field and Laboratory. *Proc. AAPT*, Vol. 27, 1958.
6. W. C. Simpson, R. L. Griffin, and T. K. Miles. Correlation of the Microfilm Durability Test with Field Hardening Observed in the Zaca-Wigmore Experimental Project. *ASTM STP*, 277, 1959.
7. R. J. Schmidt and L. E. Santucci. The Effect of Asphalt Properties on the Fatigue and Cracking of Asphalt Concrete on the Zaca-Wigmore Test Project. *Proc. AAPT*, Vol. 38, 1969.
8. R. J. Schmidt. Laboratory Measurement of the Durability of Paving Asphalts. *ASTM STP*, 532, 1973.
9. R. J. Schmidt. The Rolling Thin-Film Circulating Oven—An Improved Rolling Thin-Film Oven Test. *ASTM STP*, 532, 1973.
10. G. R. Kemp. Asphalt Durability Tests and Their Relationship to Field Hardening. *ASTM STP*, 532, 1973.
11. J. C. Petersen. A Thin Film Accelerated Aging Test for Evaluating Asphalt Oxidative Aging. AAPT preprint volume, Session II, Feb. 1989.
12. P. W. Jennings. Uses of High Pressure Liquid Chromatography to Determine the Effects of Various Additives and Fillers on Characteristics of Asphalt. Report FHWA/MT-82/001. Montana Department of Highways, 1982.
13. J. C. Petersen, F. A. Barbour, and S. M. Dorrence. Catalysis of Asphalt Oxidation by Mineral Aggregate Surfaces and Asphalt Components. *Proc. AAPT*, Vol. 43, 1974.
14. J. C. Petersen, E. K. Ensley, and F. A. Barbour. Molecular Interactions of Asphalt in the Asphalt-Aggregate Interface Region. In *Transportation Research Record 515*, TRB, National Research Council, Washington, D.C., 1974.
15. H. Plancher, S. M. Dorrence, and J. C. Petersen. Identification of Chemical Types in Asphalts Strongly Adsorbed at the Asphalt-Aggregate Interface and Their Relative Displacement by Water. *Proc. AAPT*, Vol. 46, 1977.
16. E. K. Ensley, J. C. Petersen, and R. E. Robertson. Asphalt-Aggregate Bonding Energy Measurements by Microcalorimetric Methods. *Thermochimica Acta*, Vol. 77, 1984.
17. A. F. Barbour, R. V. Barbour, and J. C. Petersen. A Study of Asphalt-Aggregate Interactions Using Inverse Gas-Liquid Chromatography. *Journal of Applied Chemical Biotechnology*, Vol. 24, 1974.
18. D. Y. Lee. Development of a Laboratory Durability Test for Asphalts. In *Highway Research Record 231*, HRB, National Research Council, Washington, D.C., 1968.
19. D. Lee. Asphalt Durability Test Correlations in Iowa. In *Highway Research Record 468*, HRB, National Research Council, Washington, D.C., 1973.
20. I. L. Jamieson and M. M. Hattingh. The Correlation of Chemical and Physical Properties of Bitumens with Their Road Performance. *Proc., Australian Road Research Conference*, Vol. 5, 1970, pp. 293–324.
21. A. C. Edler, M. M. Hattingh, V. P. Servas, and C. P. Marais. Use of Aging Tests to Determine the Efficacy of Hydrated Lime Additions in Retarding Oxidative Hardening. *Proc. AAPT*, Vol. 54, 1985, pp. 118–139.
22. O. Kim, C. A. Bell, J. E. Wilson, and G. Boyle. Development of Laboratory Oxidative Aging Procedures for Asphalt Cements and Asphalt Mixtures. In *Transportation Research Record 1115*, TRB, National Research Council, Washington, D.C., 1987, pp. 101–112.
23. B. L. Burr, R. R. Davison, C. J. Glover, and J. A. Bullin. Solvent Removal from Asphalt. In *Transportation Research Record 1269*, TRB, National Research Council, Washington, D.C., 1990, pp. 1–8.
24. H. B. Jemison, B. L. Burr, R. R. Davison, J. A. Bullin, and C. J. Glover. Application and Use of the ATR, FT-IR Method to Asphalt Aging Studies. *ACS Preprints, Division of Petroleum Chemistry*, Vol. 35, No. 3, 1990, pp. 490–495.
25. K. L. Martin, R. R. Davison, C. J. Glover, and J. A. Bullin. Asphalt Aging in Texas Roads and Test Sections. In *Transportation Research Record 1269*, TRB, National Research Council, Washington, D.C., 1990, pp. 9–19.
26. H. B. Jemison, R. R. Davison, C. J. Glover, and J. A. Bullin. Evaluation of Standard Oven Tests for Hot-Mix Plant Aging. In *Transportation Research Record*, TRB, National Research Council, Washington, D.C. (forthcoming).

27. C. A. Bell, Y. AbWahab, and M. E. Cristi. Investigation of Laboratory Aging Procedures for Asphalt-Aggregate Mixtures. In *Transportation Research Record*, TRB, National Research Council, Washington, D.C. (forthcoming).
28. B. L. Burr, R. R. Davison, H. B. Jemison, C. J. Glover, and J. A. Bullin. Asphalt Hardening in Extraction Solvents. In *Transportation Research Record*, TRB, National Research Council, Washington, D.C. (forthcoming).
29. C. A. Cipione, R. R. Davison, B. L. Burr, C. J. Glover, and J. A. Bullin. Evaluation of Solvents for the Extraction of Residual Asphalt from Aggregates. In *Transportation Research Record*, TRB, National Research Council, Washington, D.C. (forthcoming).
30. J. R. Stegeman, A. L. Kyle, B. L. Burr, H. B. Jemison, R. R. Davison, C. J. Glover, and J. A. Bullin. Compositional and

Physical Properties of Asphalt Fractions Obtained by Supercritical Extraction. *ACS Preprints, Division of Petroleum Chemistry*, Vol. 35, No. 3, 1990, pp. 431-439.

The contents of the report reflect the views of the authors, who are responsible for the facts and the accuracy of the data presented herein. The contents do not necessarily reflect the official views or policies of the Federal Highway Administration or the Texas Department of Highways and Public Transportation. This report does not constitute a standard, specification, or regulation.

Publication of this paper sponsored by Committee on Characteristics of Bituminous Materials.