Modulus Properties of Plasticized Sulfur Mixtures

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Plasticized sulfur is a total replacement for asphalt cement. As a result, the potential for using plasticized sulfur in lieu of asphalt cement in paving mixtures is attractive in this day of capricious supply and pricing of petroleum products. The modulus properties of selected plasticized sulfur mixtures were studied. The plasticized sulfur binders consist of about 70 percent elemental sulfur and 30 percent of a combination of hydrocarbons that chemically react with molten sulfur. The results of modulus characterization include resilient modulus, creep stiffness, dynamic modulus, flexural modulus, and relaxation modulus. They also include a comparison with asphalt cement modular properties. The study concluded that plasticized sulfur mixtures exhibit similar modulus properties and are stiffer than asphalt concrete. They also exhibit well-defined viscoelastic response at higher temperatures. These conclusions were consistently substantiated by each of the moduli evaluated.

The modulus properties of the materials that make up flexible pavement layers are an indispensable part of most up-to-date structural pavement design techniques. In fact, the most commonly used failure criteria in flexible pavement design are tensile strain in the stiffest layer and vertical compressive strain in the subgrade layer. These criteria are extremely sensitive to the respective modulus properties of the pavement layers. Thus, the pavement engineer must not only seek an accurate estimate of the modulus but also the proper definition of modulus for the intended purpose.

Viscoelastic materials such as plasticized sulfur concrete and asphalt concrete add another dimension of difficulty to the task of selecting the correct modulus. These materials have modulus properties that are affected by time (duration of loading) and temperature. Detailed description of the properties and uses of several plasticized sulfur binders and mixtures, including those used in this work, are available elsewhere (*1–5*).

Van der Poel (*6*) has defined the modulus of asphalt cement as stiffness:

$$S(t, T) = \text{stress/strain} \quad (1)$$

where t is the time of loading and T is the temperature.

Figure 1 is a simplified illustration of the time of loading dependency of idealized asphalt concrete at a selected temperature. It is easy to trace the change in behavior from an elastic response at short loading times, through a delayed elastic behavior zone, and finally to a region where the stiffness is totally a function of the viscous properties of the binder. Because of the time-temperature superposition properties of asphalt and plasticized sulfur, the abscissa in Figure 1 could be changed to temperature if stiffness were measured at a selected duration of loading.

In this study, the modulus properties were measured in five forms:

1. Resilient modulus (M_R),
2. Creep stiffness,
3. Dynamic modulus (E_{flex}),
4. Flexural modulus, and
5. Relaxation modulus.

RESILIENT MODULUS

The resilient moduli, defined as the ratio of induced stress to recoverable strain, were measured by the Mark IV device developed by Schmidt (*7*). The device applies a 0.1-sec load pulse once every 3 sec across the vertical diameter of a cylindrical specimen (Marshall-type specimen) and senses by linear variable transformers the resultant deformation across the horizontal diameter. The shape of the load impulse is shown in Figure 2.

The resilient modulus was used throughout as a quality assurance measure. Resilient moduli data were recorded from aging studies, water susceptibility studies, and mixture design studies. In this study, laboratory mixtures prepared with different binders were aged for 6 days at 50°F and tested at four temperatures: −10°F, 32°F, 73°F, and 104°F. The 6-day cure period was selected based on an aging study that revealed that the resilient modulus does not appreciably change in the laboratory following 6 days of curing at 50°F. The properties of the different binders used are summarized in Table 1.

In addition to the laboratory-molded specimens, field cores from a project in San Antonio, Texas (Loop 1604), were tested over the same temperature range. These data are plotted in Figure 3 and summarized in Table 2.

Table 2 and Figure 3 indicate that the resilient modulus versus temperature relationship is very similar for binders Mix 1, Mix 2, Mix 3, and the field cores from Loop 1604. These cores were 2 months old at the date of testing.

Table 2 and Figure 3 also indicate that mixtures with Mix 4 and Mix 5 proved to have lower resilient moduli than the other plasticized sulfur binders across the test temperature range.

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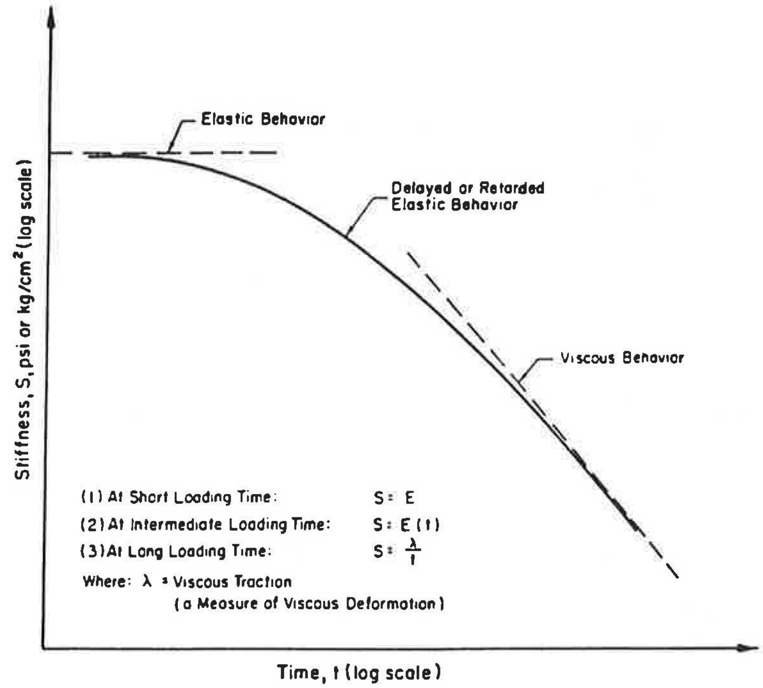

FIGURE 1 Simplified illustration of components of stiffness: elastic, viscoelastic, and viscous.

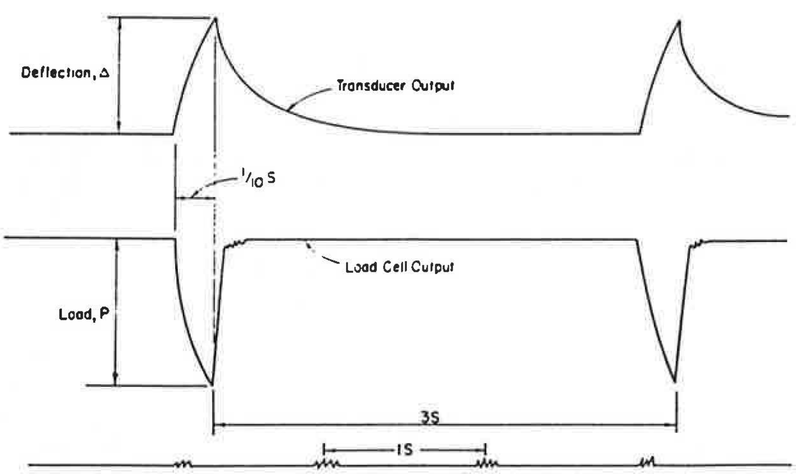

FIGURE 2 Load pulse of resilient modulus device.

The data also indicate that both the laboratory mixture of AC-10 plus crushed limestone and the field cores from Loop 1604 of AC-20 and crushed limestone produced similar M_R versus temperature relationships. It is clear that the plasticized sulfur mixtures are substantially stiffer across the temperature range. Obviously, resilient moduli will vary based on the effects of aggregate type, gradation, void content, and such, but the unmistakable trend is that plasticized sulfur mixtures under rapid load rates are much stiffer than asphalt concrete. However, plasticized binders display a very definite temperature susceptibility and viscoelastic response. It should be mentioned that variations in the number of specimens tested in the various cases tested should not cast doubt on the accuracy of comparing the different materials because a minimum of three specimens will provide adequate accuracy.

Figure 3 indicates that the viscoelastic behavior of plasticized sulfur mixtures actively begins at about 32°F. Below about 32°F, the modulus is probably very nearly linearly elastic. However, the accuracy of diametral resilient moduli determinations at low temperatures (32°F) is poor (*7*). The resilient modulus data in Figure 3 indicate a dominating viscous response at high temperatures, perhaps above 100°F. The stiffness data for all mixtures containing crushed limestone and basaltic aggregate indicate substantial decrease in stiffness with increased time of loading, which reflects the dominating viscous response at high temperatures because high-

TABLE 1 FORMULATION AND PROCESSING CONDITIONS FOR BINDERS ANALYZED

Binder Identification	Formulation	Reaction Temperature, °F	Reaction Time, Hours	Penetration at 77°F, (1 Day old), dmm	Viscosity at 275°F, (1 day old), poises
Mix 1	68% Sulfur 12% DCPD** 12% DP*** 8% Vinyl Toluene Catalyst (1% of mix)	302	6.5	35	10.2
Mix 2	70% Sulfur 12% DCPD 10% DP 8% Vinyl Toluene Catalyst (1% of mix)	338	6.5	165	4.0
Mix 3	70% Sulfur 12% DCPD 10% DP 8% Vinyl Toluene Catalyst (1% of mix)	302	6.5	22	5.8
Mix 4	70% Sulfur 12% DCPD 10% DP 8% Vinyl Toluene Catalyst (1% of mix)	320	6.5	78	2.5
Mix 5	68% Sulfur 12% DCPD 10% Solvenol 10% Vinyls Toluene Catalyst (1% of mix)	300-350	4.5	40	11.0

* Expressed as percent by weight.
** DCPD = Dicyclopentadiene
*** DP = Dipentene

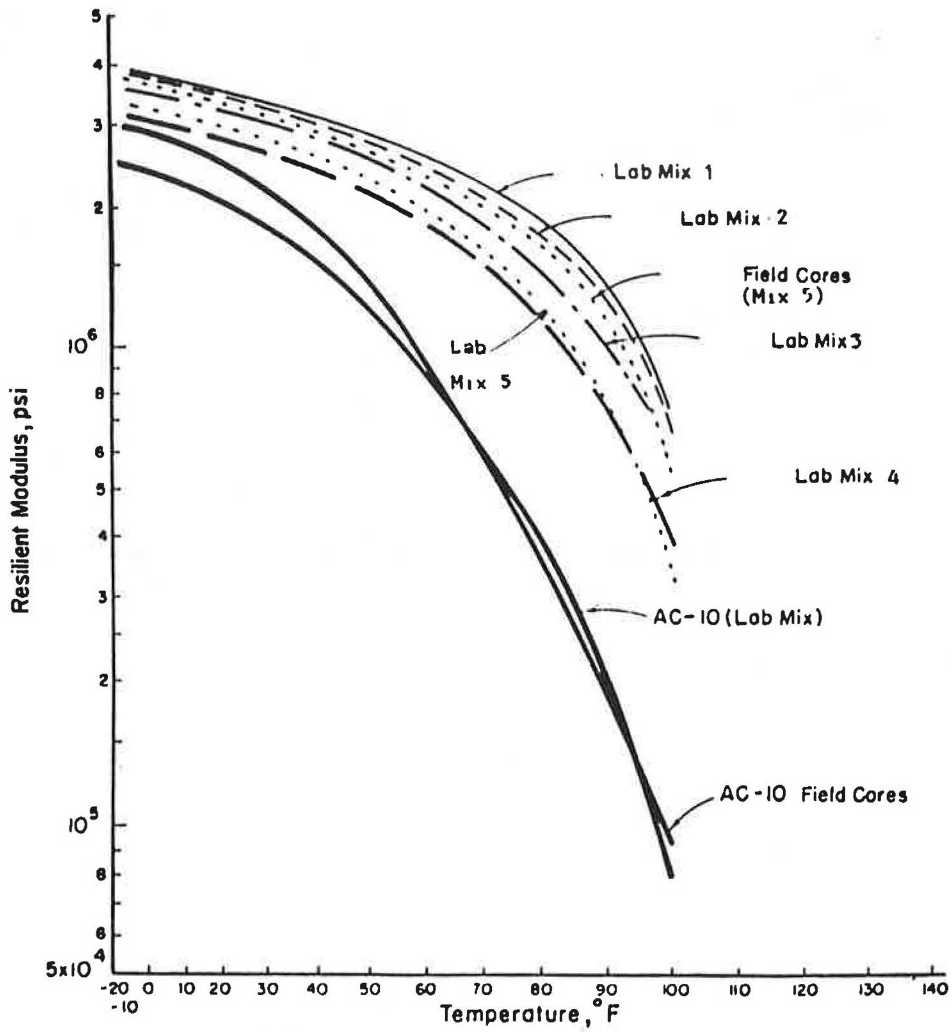

FIGURE 3 Resilient moduli versus temperature for all mixtures fabricated with crushed limestone.

TABLE 2 SUMMARY OF RESILIENT MODULI

Binder	Aggre-gate	Type of Mixture	Mean Resilient Modulus, psi x 10^6 -10 F	73°F	104°F	No. of Spec.
Mix 1	CLS*	Lab	4.01	2.22	0.790	50
Mix 2	CLS	Lab	4.00	2.16	0.699	50
Mix 3	CLS	Lab	3.62	1.82	0.670	20
Mix 4	CLS	Lab	3.19	1.29	0.540	20
Mix 5	Basalt	Lab	3.29	1.50	0.300	9
Mix 5	CLS	Field	3.81	2.01	0.710	20
AC-10	CLS	Lab	2.66	0.489	0.077	20
AC-10	Basalt	Lab	2.51	0.321	0.102	9
AC-20	CLS	Field	3.00	0.501	0.092	20

*CLS : Crushed limestone

temperature effect on viscous materials can be viewed as similar to the effect of long duration of loading. Another point of interest is the generally lower stiffness values of mixtures prepared by using basalt aggregates. This is also true, but to a lesser extent, of field versus laboratory data. General conclusions related to the effect of aggregate type on moduli and stiffness values will not be justified here because of the limited available data related to this specific factor. The lower stiffness and moduli values associated with field data may be attributed to the lesser degree of control in the preparation of field mixtures.

The argument has often been put forth that plasticized sulfur layers should be designed as rigid pavement layers because they probably crystallize rapidly in the thin-film arrangement found in mixtures, and they are probably very nearly linearly elastic in normal pavement conditions. However, the pronounced viscous effects at higher temperatures and longer loading rates and especially a combination of the two effects demand thoughtful consideration of the consequences of these effects in pavement design applications.

CREEP STIFFNESS

The diametral resilient modulus is often subjected to criticism because of the light load used, the conditions of biaxial stressing, and the rigid assumptions that should be but are not closely adhered to in order for the cylindrical, diametrally loaded specimen to respond elastically. To more precisely establish the modular properties of plasticized sulfur under different conditions of loading and different states of stress, other forms of moduli were computed.

Creep stiffness is simply the inverse of the creep compliance. For purposes of comparison creep stiffness was calculated at 0.1 sec of load duration at 40°F, 70°F, and 100°F during the compression creep test. The resulting values are tabulated in Table 3.

As expected, these moduli do not closely agree with the resilient moduli. However, the same trends are evident as were established with resilient moduli data. These moduli are plotted in Figure 4.

DYNAMIC MODULUS AND FLEXURAL MODULUS

The dynamic moduli and flexural moduli are also presented in Table 3. The dynamic modulus is defined here as the ratio of stress applied during repeated load permanent deformation testing to the dynamic strain at the 200th load application. The flexural modulus is defined as the modulus of the flexural fatigue beams at the 200th load application. The modulus is more clearly defined as follows:

TABLE 3 SUMMARY OF CREEP STIFFNESSES, DYNAMIC MODULI, AND FLEXURAL MODULI FOR ALL MIXTURES

Binder	Aggregate	Creep Stiffness, psi x 10^6 40 °F	70°F	100°F	Dynamic Modulus, psi x 10^6 40°F	70°F	100°F	Flexural Modulus psi x 10^6 70°F
Mix 1	CLS**	3.51*	1.90	0.320	4.00	2.69	0.290	0.350
Mix 2	CLS	8.00	2.90	0.300	4.76	2.22	0.495	0.675
Mix 4	CLS	2.20	1.30	0.210	2.75	1.30	0.190	0.330
Mix 5	Basalt	5.00	2.00	0.150	4.00	1.02	0.200	------
AC-10	CLS	1.10	0.310	0.060	2.10	0.397	0.086	0.056
AC-10	Basalt	1.50	0.400	0.059	2.50	0.700	0.090	------

* Each Value represents the mean of three data points.

** CLS : Crushed Limestone

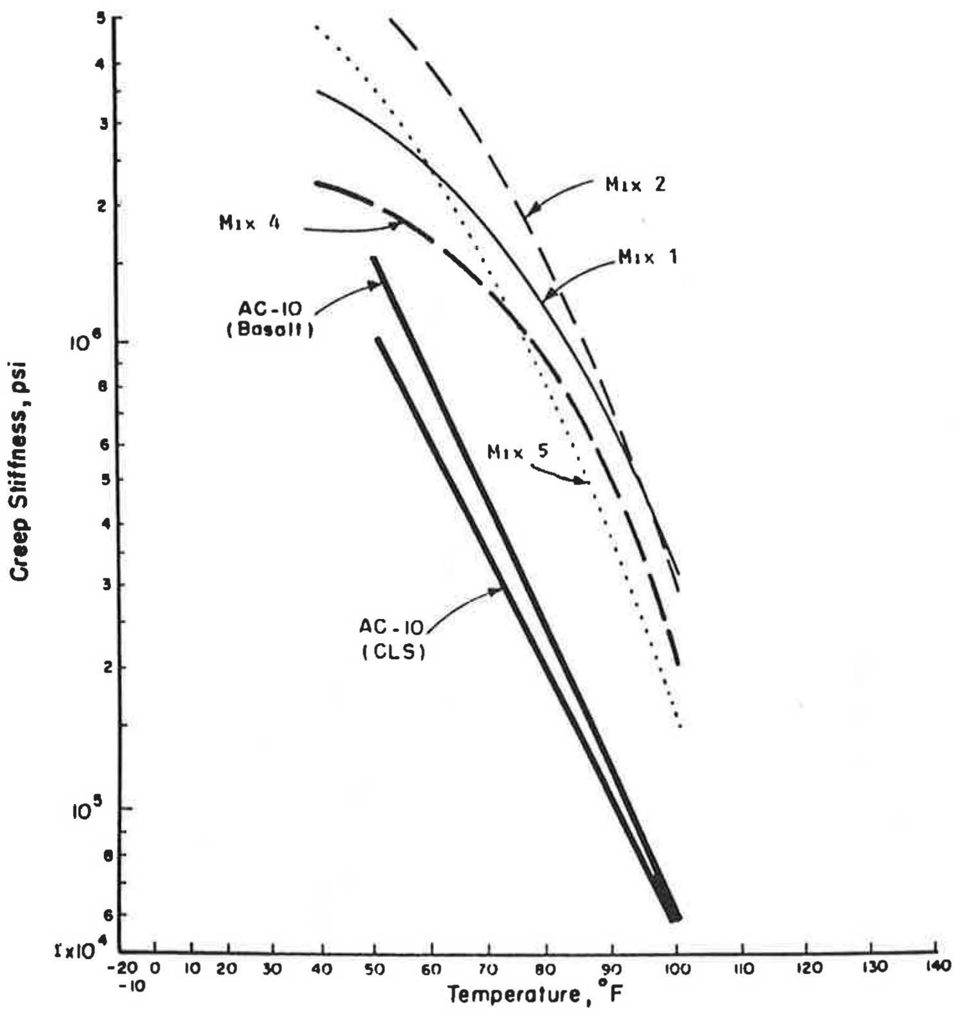

FIGURE 4 Creep stiffness versus temperature for all mixtures.

$$E_{\text{flex}} = \frac{Pa(3L^2 - 4a^2)}{48\ (I)(DELTA)} \quad (2)$$

where

P = dynamic load applied to deflect the beam,
a = $1/(2L-4)$,
L = reaction span length,
I = specimen moment of inertia, and
$DELTA$ = dynamic beam deflection at the center point.

The dynamic moduli are plotted in Figure 5.

RELAXATION MODULI

The relaxation modulus was measured by applying a compressive stress that induced an initial strain of 100 μin./in. in specimens 4 in. in diameter and 8 in. high. Strains were measured with linear variable transformers. Strains were monitored across the middle 4 in. of the cylindrical specimens. The resulting relaxation moduli are presented in Table 4.

MIXTURE PROPERTIES

Although the purpose here is to compare differences in binders, it is appropriate to indicate average mixture properties as a point of reference. For this purpose, aggregates gradation curves, Marshall stability, Marshall flow, percent air voids, and percent voids in mineral aggregate (VMA) for mixtures tested are shown in Figures 6, 7, 8, 9, and 10, respectively.

SUMMARY

From this study of modulus properties, the following conclusions are established:

1. Plasticized sulfur binders Mix 1, Mix 2, Mix 3, and Field Mix 5 generally exhibit very similar modulus properties over the temperature range that a typical pavement is expected to experience.
2. Although substantially stiffer than asphalt concrete over the normal pavement temperature range, all plasticized sulfur binders exhibit well-defined viscoelastic response as well as dominant viscous response at higher temperatures. These

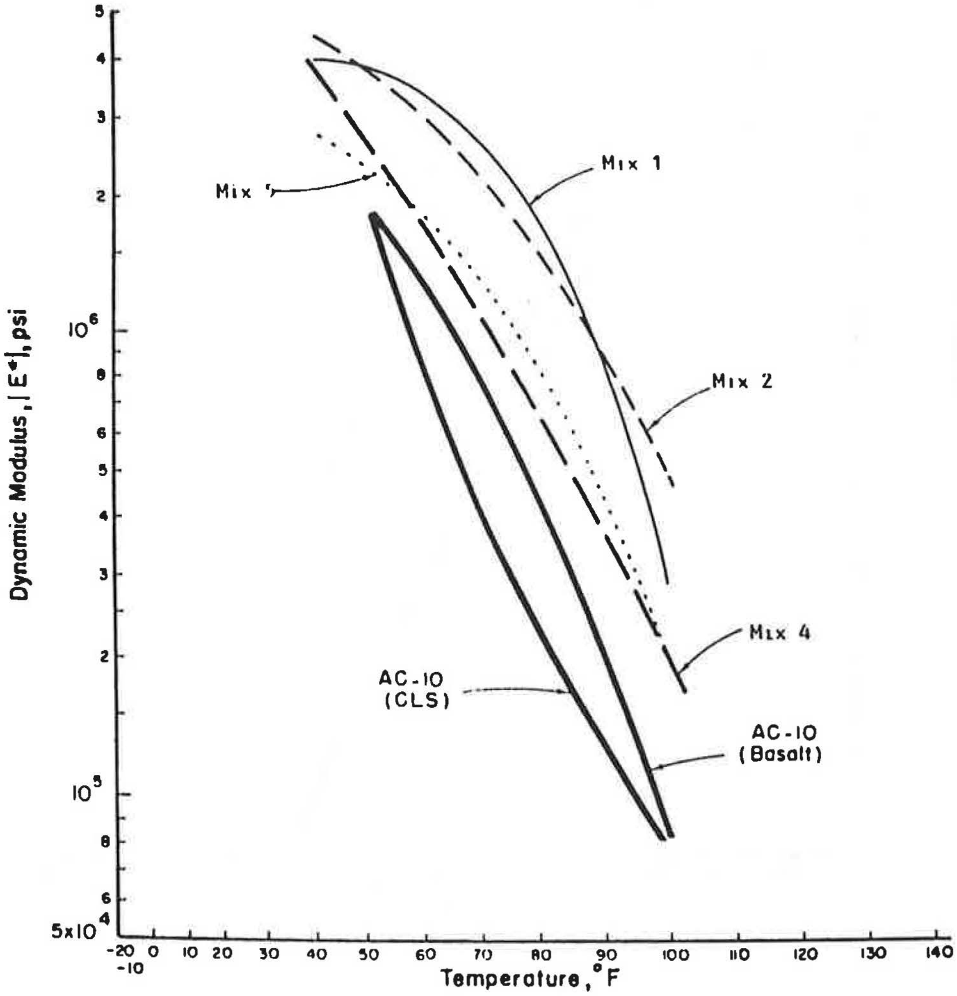

FIGURE 5 Dynamic moduli versus temperature for all mixtures.

TABLE 4 RELAXATION MODULI AT 73°F, 0.1-sec LOAD DURATION

	Relaxation Modulus, Psi x 10^6		
Binder	Mean Value	Standard Deviation	Number of Samples
Mix 1	1.129	0.180	3
Mix 2	0.637	0.010	3
Mix 3	1.449	0.023	3
Mix 4	0.557	0.158	3
AC-10	0.433	0.095	3

*CLS: Crushed limestone

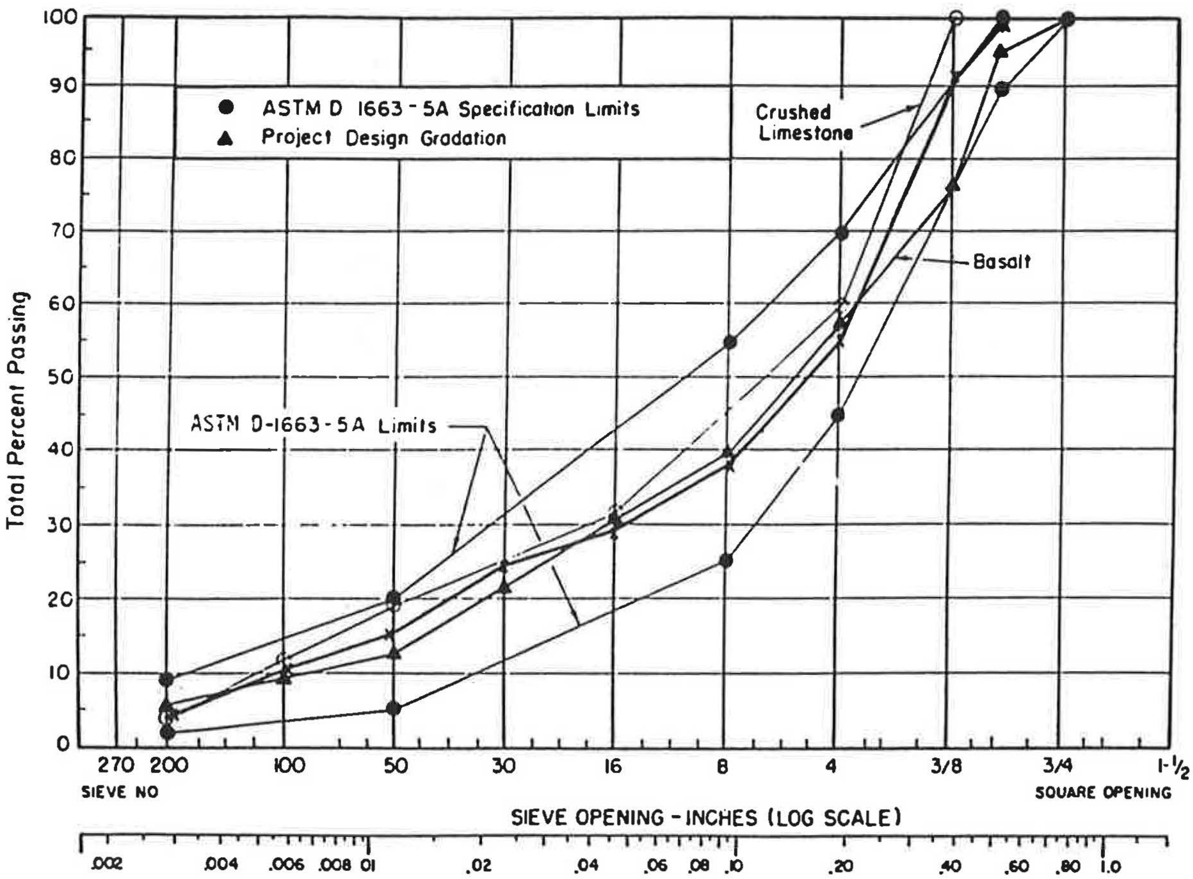

FIGURE 6 Gradation curves for aggregates used in mixtures.

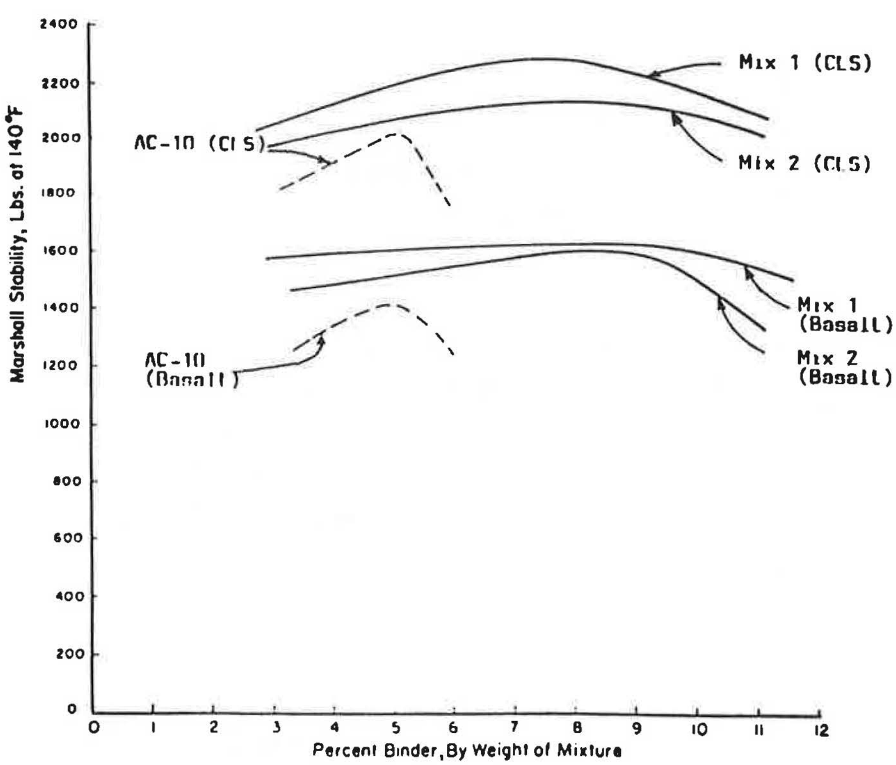

FIGURE 7 Marshall stability versus binder content for mixtures tested.

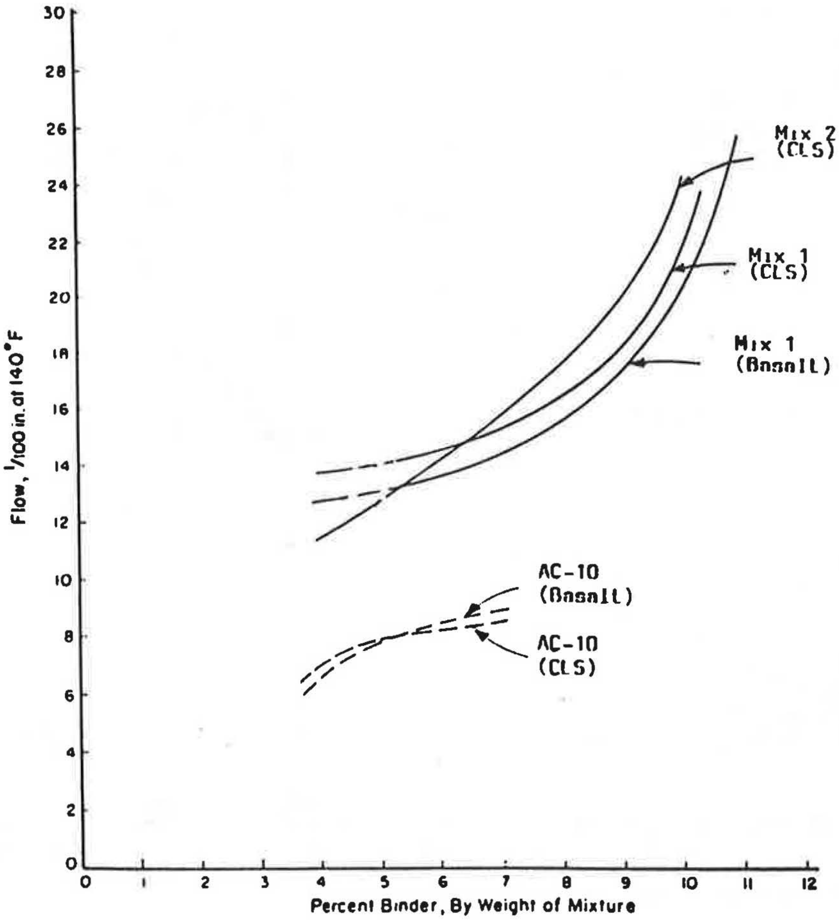

FIGURE 8 Marshall flow versus binder content for mixtures tested.

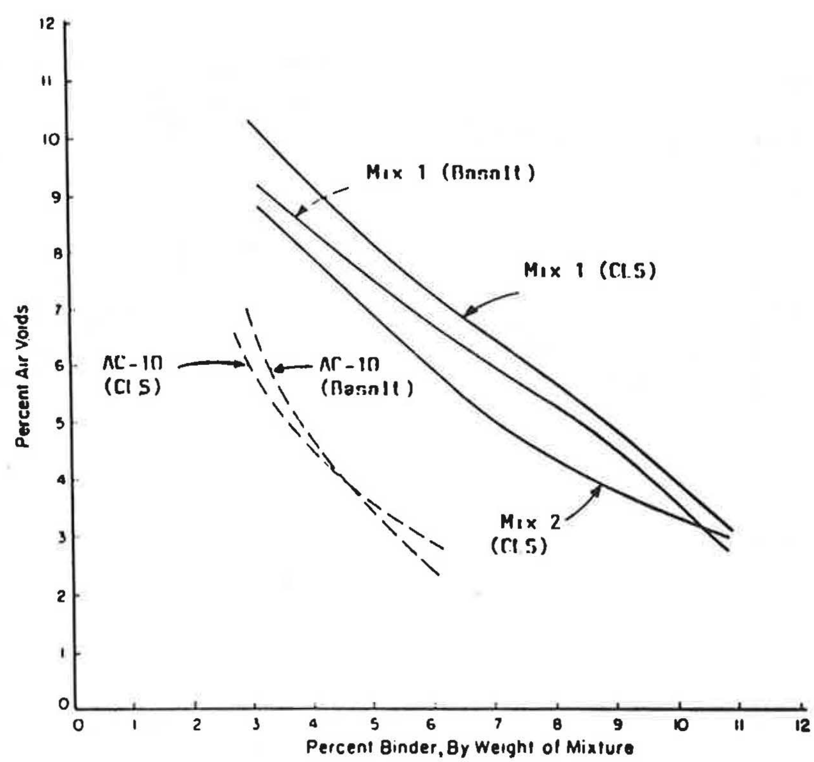

FIGURE 9 Percentage air voids versus binder content for mixtures tested.

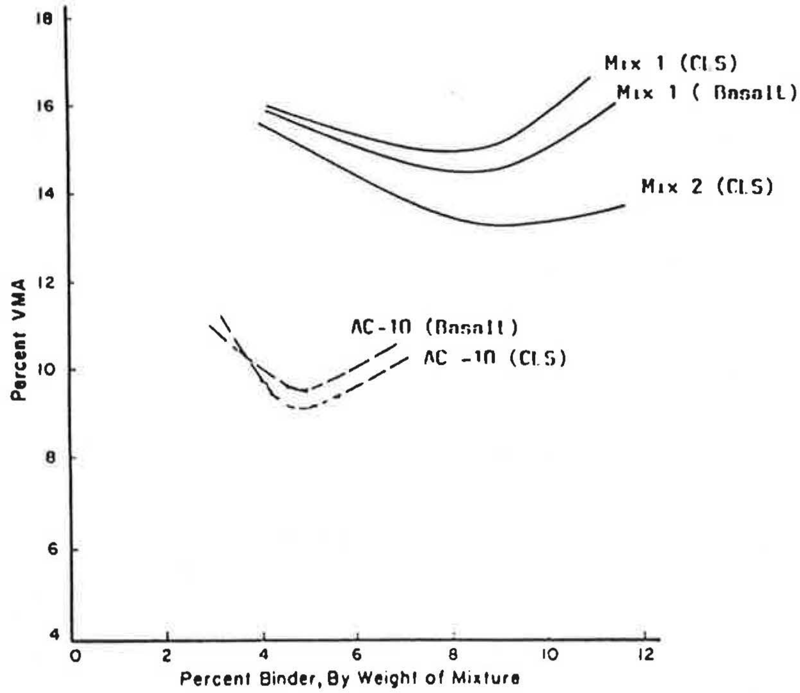

FIGURE 10 Percentage VMA for mixtures tested.

properties must be accounted for in structural pavement design considerations.

3. Binder Mix 4 is significantly softer than other plasticized sulfur binders across the temperature range to which pavements are normally subjected. Laboratory mixtures prepared with Mix 5 are also as soft as those prepared with Mix 4. However, the use of basalt aggregates in these mixtures may have contributed to their softness.

4. Resilient moduli, stiffness moduli, dynamic moduli, flexural moduli, and relaxation moduli were evaluated for each binder and the conclusions stated above consistently substantiated by each of the moduli versus temperature data.

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