creasing traffic. At low traffic levels, the mixture with 2 percent rubber has significantly higher permeability but is similar to the mixture with 0 percent rubber at higher volumes. The air permeability of the mixture with 1 percent rubber is low and nearly constant regardless of traffic. This implies that the mixture with 1 percent rubber compacts more completely under the roller and that no further compaction occurs under traffic.

The average densities of the pavement with 0 percent and that with 1 percent rubber were the same at 142 lb/ft^3. Since the voidless unit weight of the mix with 1 percent rubber is lower, this indicates a greater degree of compaction. The material with 2 percent rubber averaged 1.5 percent less at 139.8 lb/ft^3. The relationship between density and traffic volume (Figure 9) is that of increasing density with increasing traffic level. The two locations where the original pavements were portland cement concrete recorded the highest nuclear densities. Either the gauge is influenced by the concrete or the more rigid base aids in compaction. Both had been overlaid previously by using bituminous concrete.

A comparison of the average nuclear density at one year with the average Marshall density at the time of construction provides an insight into the characteristics of the materials. At one year, the average wheel-track nuclear density for the sections with 0 percent rubber was 92 percent of the original Marshall density. For the material with 1 percent rubber this comparison was 95 percent and for that with 2 percent rubber, 94.6 percent.

Based on the average nuclear densities, the void content of the three types of pavement at one year would be 10.4 percent for the material with 0 percent rubber, 9.2 percent for that with 1 percent rubber, and 8.1 percent for that with 2 percent rubber.

Changes in paving materials always raise concerns about skid resistance. Skid-resistance numbers determined by using equipment that meets current federal highway standards provide realistic comparisons between surfaces. Those for the rubber-modified overlays are listed in Table 8. Extensive statistical computations by using the Student's t-test show few significant differences. The only comparisons that resulted in confidence levels about 95 percent were those in 1977 when, at the 95 percent level, it could be said that those mixes that contained rubber were superior to those without and that 2 percent rubber mixes were better than mixes with either 0 or 1 percent rubber. At an 85 percent level, the mixes that had 2 percent rubber were higher than the mixes with 1 percent rubber in 1978. There is a weak trend for all materials to improve with age. For all test sections, the skid numbers compare well with the nonrubber control, and skid resistance should not be a problem.

CONCLUSIONS

The rubber-modified mixes are performing as a class better than the nonrubberized mixes do. Cracking has been reduced and there has been no reduction in skid resistance. The lower permeability and higher density of the rubber-modified mixture imply a probability of longer life.

ACKNOWLEDGMENT

The views expressed are mine and may not represent the official position of the University of Connecticut or the supporting agencies, ConnDOT and the Federal Highway Administration.

REFERENCES

1. J.E. Stephens and S. Mokrzewski. The Effect of Reclaimed Rubber on Bituminous Paving Mixtures. Univ. of Connecticut, Storrs, Civil Engineering Department Rept. CE 74-75, Feb. 1974.
2. J.L. McRae and B.D. Lagrone. Effect of Modified Reclaimed Rubber and Ground Vulcanized Rubber on the Physical Properties of Bituminous Pavements as Evaluated by the Gyratory Testing Machines. HRB, Highway Research Record 361, 1971, pp. 14-19.
3. Standard Specifications for Roads, Bridges, and Incidental Construction--Form 811. Connecticut Department of Transportation, Wethersfield, 1974.

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Methods of Increasing Fracture Toughness of Asphalt Concrete

R.T. WOODHAMS

Experience with rubber-modified asphalt concrete in the Canadian environment during the past seven years has yielded only marginal improvement with respect to cracking. The most effective rubbers for reducing the brittle temperature of asphalt concrete are those that have low glass transition temperatures, although reclaim tire rubber is preferred since it costs the least. Chopped nylon or polyester tire cord or integrated rovings can impart large increases in fracture toughness at temperatures below freezing. For maximum efficiency, the chopped cord should be near its critical pullout length. Calculations indicate that fracture toughness can be increased 20-fold under ideal conditions with the addition of only 1 percent fiber. Moisture damage can be minimized by the addition of a minor proportion of ferric oxide or iron naphthenate as an adhesion promoter. The combined use of rubber, chopped fibers, and an adhesion promoter should help to improve the durability of most asphalt concretes in cold climates. Paving trials are needed to indicate the long-term economic benefit of these modifications.

Despite several serious deficiencies, bitumen continues to be favored as a low-cost binder in road paving formulations. The temperature susceptibility of bitumen is so pronounced that at elevated temperatures, creep and distortion of asphalt pavements

become excessive, whereas at temperatures below freezing, the pavement becomes so brittle that numerous cracks develop as a consequence of thermal shrinkage forces. It is difficult to tailor a bitumen that performs equally well at such environmental extremes and simultaneously resists oxidative hardening, moisture damage, stress fatigue, freeze-thaw cycles, thermal cycling, excessive loading, and unstable foundations. Therefore, in order to improve the performance of bitumen as a pavement binder it would be desirable to make the following changes:

1. Reduce the brittle temperature through the use of rubber additives and at the same time increase the resistance to flow at elevated temperatures;
2. Promote better adhesion to the aggregate, particularly in the presence of moisture, by incorporating suitable adhesion promoters; and
3. Increase the fracture toughness of asphalt concrete by adding a minor proportion of chopped fibers.

Previous investigators have demonstrated that the brittle temperature of bitumen and its mixtures can be lowered by the incorporation of rubber; the rubber is added to the bitumen as an uncured gum, thermoplastic elastomer, liquid latex, or powdered reclaim (1). The reduction in the brittle temperature will depend on the concentration of the added rubber and its glass transition temperature (Tg). Therefore, one of the most effective elastomeric materials should be cis-polybutadiene, which has a reported glass transition temperature of -120°C. For economic reasons, reclaim rubber powder obtained by the mechanical or cryogenic grinding of discarded tires has become the preferred additive even though it is not so efficient as other elastomers. Commercial methods have now been developed for paving rubber-modified asphalt concrete by the hot-mix process (2) or by seal coating (3). (Methods have also been reported by L.S. Brake, Sahuaro Petroleum and Asphalt Co., P.O. Box 6536, Phoenix, AZ 85005, and by E. Thompson, Pounder Emulsions, 1474 Wall Street, Winnipeg, Manitoba R3E 2S4, Canada.) These two methods have been used in Canada during the past several years with limited success. Despite rubber modification, cracking of the surface persists. Surface texture remains good after four years and there is negligible ravelling or stripping observable with Metro Toronto RA-3 rubberized asphalt (4,5).

The addition of rubber to an asphalt concrete does reduce the brittle temperature but has a minor effect on the tensile strength or toughness of the composite. Toughness can be increased through the addition of chopped cord or rovings of sufficient length and size to impart a large increase in fracture energy (6). The maximum toughness is obtained when the chopped cord or roving is near its critical length, a length that depends on the temperature, interfacial shear strength, and cord diameter. Chopped fibers have been used in the past to toughen portland concrete (7,8) and sulfur concrete (9). They often produce 20-fold increases in fracture toughness at fiber concentrations near 1 percent by weight (e.g., steel wires or glass, polypropylene, nylon, or polyester fibers). The availability of large quantities of waste tire cord makes this method of toughening especially attractive. This increase in fracture toughness is retained even at freezing temperatures when the asphalt concrete has become very brittle. With proper design, the fibers can induce multiple cracking so that large cracks are less likely to occur.

Various publications have drawn attention to the loss of adhesion at the interface between the bitumen binder and the aggregate when moisture is present. This phenomenon is sometimes referred to as water stripping, disbonding, or water damage. The loss of adhesion may occur after long immersion in water or prolonged exposure to a humid environment, after which the asphaltic concrete may be seriously weakened and thus become more susceptible to damage. Under moist conditions, asphalt concrete can lose up to 80 percent of its strength (10). Although adhesion promoters or antistripping agents (such as amines and lime) have been suggested as additives to help prevent this loss of adhesion, Scott (11) concluded that "cationic adhesion additives in the asphalt can delay but not prevent loss of adhesion," whereas "anionic types are only likely to be successful where there is an opportunity for water insoluble salt formation with tightly bound surface metal ions." Fromm (12) found that three commercial antistripping additives delayed the onset of stripping but actually caused accelerated stripping in the later stages, which also promoted increased emulsion formation in the asphalt. The most effective reagent found by Fromm for preventing stripping was an asphalt-soluble iron compound, iron napthenate, which is a commercially available paint drier (Nuodex DMR Iron 6%, Nuodex Canada, Ltd., 34 Industrial Street, Toronto, M4G 1T9, Canada).

The following investigation describes attempts to evaluate the effectiveness of these three types of additives--rubber, fibers, and iron naphthenate--on the performance of asphaltic paving materials by using diagnostic laboratory tests. A more complete evaluation awaits further tests and paving trials.

EXPERIMENTAL

The dynamic mechanical properties of the bitumen-rubber mixtures were measured on a Rheovibron viscoelastometer (Model DDV-II) in a shear mode. The sample holder and measurement technique have been described previously (1). Viscosity measurements were conducted with a Brookfield viscometer while the samples were maintained at constant temperature in a heated bath.

The measurement of contact angles was carried out by the technique of Neumann and Good (13, Chapter 2, p. 31). The sessile drops of each bitumen sample were allowed to equilibrate in an air oven at 100°C for 15 min and were then removed for measurement of the contact angle by using a calibrated goniometer.

The asphalt concrete test beams were prepared in the laboratory by using a special roller equipped with a hydraulic stage (2). Rectangular slabs 30 cm long, 19 cm wide, and 5.2 cm thick could be prepared with a void content of less than 1 percent. A standard paving formulation (Metro Toronto HL-1 mix) was used as a control. Each compressed slab was cut into several rectangular beams for flexural testing. The full procedure has already been described (2).

Peel forces were measured at a 90° angle by using glass fiber tapes adhered by various bitumen mixtures to horizontal glass plates. Hot bitumen mixtures were spread onto clean glass plates and the glass fiber tapes (15x6.5 cm) were embedded into the liquid bitumen. A sheet of paper was placed over the glass tape so that a second glass plate could be positioned over the assembly without adhering to the bitumen. Thin aluminum shims (0.6 mm thick) placed between the parallel glass plates ensured a uniform reproducible film thickness. A weight was placed on top of the assembly and each specimen was allowed to equilibrate for one week before the assembly was subjected to complete immersion in distilled water or exposed to the atmosphere. [Details of this pro-

Figure 1. Setup for 90°-angle peel testing.

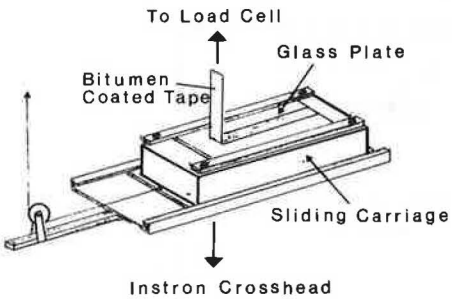
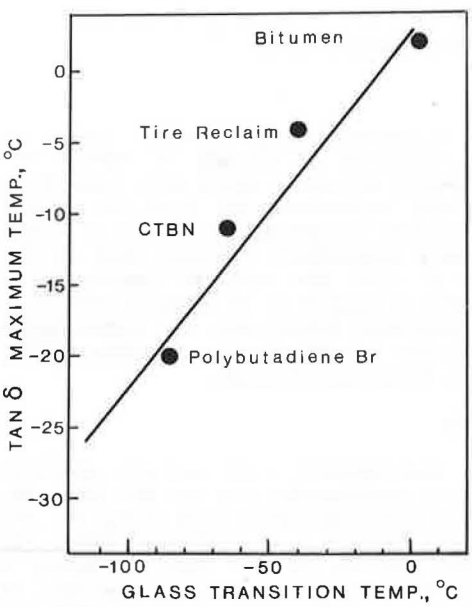


Figure 2. Effect of addition of 20 parts of rubber to 100 parts of bitumen on brittle temperature of mixture.



cedure may be obtained from the thesis of Varevorakul (14); see ASTM B533-70.] Figure 1 shows the experimental rig for 90°-angle peel testing, which attaches to an Instron tester. All peel tests in this series were conducted at a temperature of 16.5°C and a peel rate of 1 cm/min. The sliding carriage may be heated or cooled to provide data over a wide range of temperatures if necessary.

RUBBER-MODIFICATION RESULTS

The addition of rubber (whether in particulate form or as a liquid) to bitumen lowers the brittle temperature of the mixture. The extent of lowering was proportional to the amount of rubber and its glass transition temperature. This trend is indicated in Figure 2 in which, from dynamic mechanical tests (1), the tan δ maximum for these rubber-bitumen blends shows a linear proportionality to the glass transition temperatures of the added rubbers at 20 parts of rubber. It is likely that cis-polybutadiene (not shown), which has a glass transition temperature of -102°C, would be the most effective rubber for reducing the brittle temperature of asphalt concrete at this concentration.

The extent of reduction of the brittle temperature ΔT may be predicted from the rule of mixtures for any concentration by using the following simple formula:

$$\Delta T = (T_a - T_r)\phi \quad (1)$$

where $T_a - T_r$ is the difference between the glass transition or brittle transition T_a of the asphalt cement and the glass transition T_r of the added rubber. The symbol ϕ is the volume fraction of added rubber in the mixture. The glass transitions of most elastomers are known or can be estimated from the tables (15). The brittle temperature of bitumen is near 0°C at 30 Hz.

The addition of rubber to an asphalt mix also increases the viscosity so that, in practice, longer mixing times are required to achieve the same degree of aggregate dispersion. In the case of powdered reclaim rubber, an additional 10 s in a production-sized pug mill was necessary for proper mixing. When the temperature of the pug mill was increased by 20°C (about 1°C for each part of added rubber), no additional mixing time was required. Since the time in the pug mill is usually quite short (less than 1 min), no appreciable swelling or reaction can take place between the particulate rubber and the bitumen. There is evidence to suggest, however, that over a longer period of time (about one year) at ambient temperatures, a chemical reaction (probably initiated by air oxidation) takes place after the mixture has been applied to the road surface. This chemical interaction may lead to improved bonding (adhesion) of the bitumen onto the rubber phase and a concordant increase in the toughness of the rubber-modified concrete as it ages. According to Fromm and McDougall of the Ministry of Transport and Communications in Downsview, Ontario, samples of rubberized asphalt concrete removed from three-year-old pavements in Metro Toronto after recompaction gave Marshall flow values (0.01 in) between 16 and 40 that correspond to Marshall stability values of 2765 and 1350 lbf. This surface texture of the road was rated excellent and there was negligible evidence of stripping or ravelling. Good adhesion to the portland concrete base was noted although the usual number of cracks (both longitudinal and transverse) was apparent. The experience with reclaim rubber in hot-mix paving in the Metro Toronto area now extends more than six years, and although all pavements have been rated in good condition, the improvement gained through the addition of rubber thus far is considered marginal. The incorporation of reclaim rubber into a paving mixture does require a corresponding increase in the asphalt concentration, which in view of the increasing price of asphalt has tended to deter further trials.

Laboratory samples of rubberized asphalt concrete containing vulcanized tire rubber (minus-30 mesh size) showed a slight gain in strength and toughness after one year's immersion in tap water. It is not known to what extent the other elements and compounds usually present in tire compounds (sulfur, zinc oxide, accelerators, antioxidants, antiozonants, oil extenders, stearic acid, processing aids, carbon black, etc.) influence the long-term performance of asphalt paving materials, but they could well have a beneficial effect.

In a series of experiments, chopped nylon and chopped polyester tire cords were incorporated into a standard hot-mix formula as shown below:

Material	Percent by Weight
Trap rock (1 cm coarse)	42.0
Trap-rock screenings	12.8
Sand	38.6
Asphalt	6.0
Chopped cord (1.2 cm)	0.6

Figure 3. Stress-strain response of sample that contained 1.2 volume percent chopped nylon tire cord 1.3 cm long.

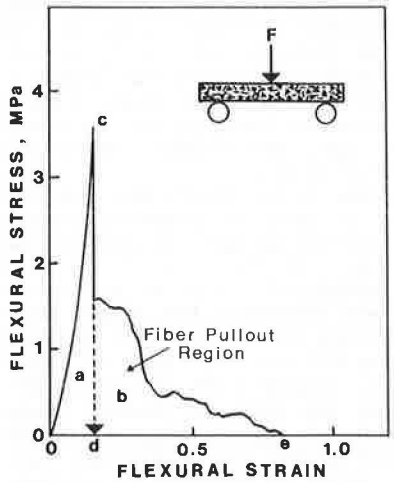
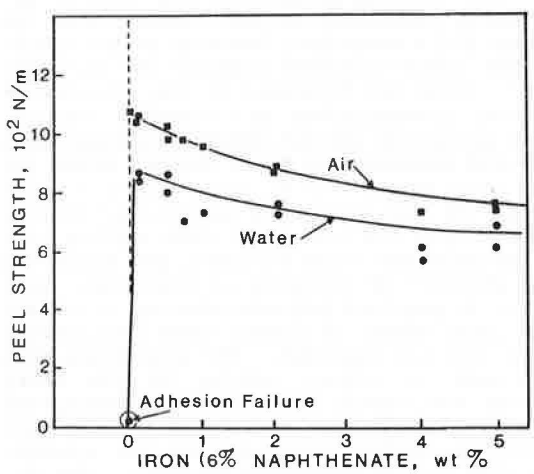
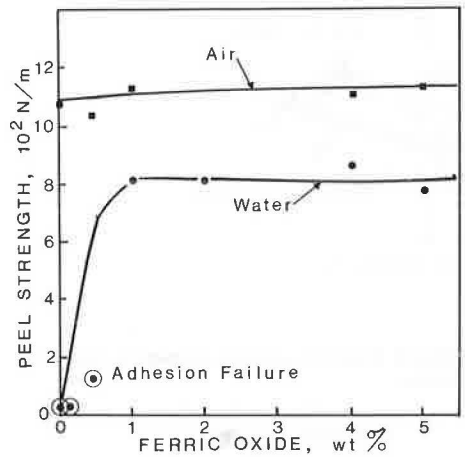


Figure 4. Peel strength when iron naphthenate is added to asphalt mixture at 16.5° C.



In a three-point flexural test, the stress-strain response shown in Figure 3 was obtained at -20°C (which is well below the brittle temperature of the bitumen binder). The dotted line shows the path of the recorder pen when the chopped tire cord was absent. The influence of the tire cord on the stress-strain behavior is negligible up to the appearance of the first crack at c, after which the ultimate strain is increased from d to e. The area under the stress-strain curve is divided into two sections--that represented by a is the energy absorbed up to the first crack and that labeled b is attributable to debonding and fiber pullout energies. As the fiber length and fiber concentration increase, area b also increases without greatly influencing the first portion of the stress-strain curve. In other studies the optimum cord length was found to be 5 cm at fiber concentrations between 1 and 2 percent. When chopped roving is used instead of tire cord, the roving must be treated by using a binder to prevent dispersion of the individual filaments during the mixing stage. Attempts to incorporate loose reclaim tire cord filaments into the bitumen produced such large increases in viscosity that mixing with the aggregate became extremely difficult and would present problems in a pug mill. Even short filaments (a few millimeters long) produced large increases in the viscosity of bitumen at 120°C. The extent of toughening was quite small with such short filaments and this emphasizes the need for longer multifilament strands (or cord) that act as a single unit. Preliminary experiments indicate that in the brittle region at -20° the critical length for a typical tire cord is near 5 cm in asphalt concrete. Such chopped integrated fibers are now commercially available in Canada (Hartford Fibre Company, Ltd., Progress Avenue, Kingston, Ontario).

Figure 5. Peel strength when ferric oxide is added to asphalt mixture at 16.5° C.



ADHESION RESULTS

The peel-test measurements with asphalt mixtures that contain iron naphthenate (Nuodex DMR Iron 6%) substantiated the observations made by Fromm in 1974 (12). After long immersion in distilled water (25 days), the bond between asphalt that contained a small percentage of iron naphthenate (0.1 percent by weight) failed in a cohesive manner rather than adhesively (at 16.5°C) when applied to a borosilicate glass plate. Without the iron naphthenate, the peel strength was almost zero with adhesive failure. In a few experiments, ferric oxide powder (red, anhydrous) was added to the bitumen. Although not quite so effective as the soluble iron naphthenate, the iron oxide did confer good adhesion when immersed in water. These two additives are compared in Figures 4 and 5. It is apparent that the iron naphthenate is more effective at low concentrations. However, ferric oxide can be obtained as an inexpensive waste from iron foundries, so this additive may be more economically attractive. Since most bitumens are slightly acidic, it is possible that some of the ferric oxide reacted to form soluble iron salts.

The greater adhesion to a glass surface of bitumen that contained iron naphthenate was also confirmed from contact-angle measurements. Bitumen containing 1 percent iron naphthenate gave a contact angle of 11° compared with 17° without naphthenate. This simple diagnostic test may be used to check the relative performance of other additives and substrates by using very small samples, if preferred.

DISCUSSION OF RESULTS

Although liquid or solid rubbers that have low glass transition temperatures are more efficient for reducing the brittle temperature of asphalt concrete, powdered reclaim rubber from discarded tires repre-

sents the only economically viable source of rubber for general use in road surfacing. Simple mixing in a conventional pug mill is adequate in order to disperse the rubber particles, which are preferably about 1 mm in size (2). Smaller particles are more difficult to disperse and do not appear to offer any advantages. Powdered tire rubber also contains antioxidants, antiozonants, sulfur accelerators, zinc oxide, carbon black, and other additives that can be beneficial for the long-term protection of the road surface by retarding embrittlement and increasing toughness. Some evidence suggests that long-term aging causes the bitumen molecules to interact with the rubber so that the rubber particles become more strongly bound to the bitumen. This slow reaction may be due to air oxidation or continued sulfur cross-linking during hot summer periods. This interaction may help to promote toughening in a manner analogous to the toughening of polystyrene and acrylics with rubber (16).

The fracture toughness G for a composite that contains randomly dispersed fibers wherein the fibers are all at their critical length L_c and with a volume fraction V_f and an ultimate tensile strength σ_{fu} will be given approximately by the following simple relationship:

$$G \simeq kV_f \sigma_{fu} L_c \quad (2)$$

where k is a proportionality factor that has a value near 0.1 for chopped nylon or polyester fibers. Thus for a volume fraction of 0.02 nylon cord that has a critical length of 5 cm and an ultimate tensile strength of 1 GPa, the total fracture energy is approximately 100 kJ/m² compared with an apparent fracture energy (including plastic deformation at the crack tip) for a typical asphalt concrete of 5 kJ/m² at -10°C. Thus, a relatively small proportion of either organic or inorganic fibers (that have an appropriate critical length) can increase the fracture toughness of asphalt concrete by a factor of 20 at low temperatures when the system is brittle. This toughening has been observed in experiments with other brittle materials such as portland concrete (7,8) and sulfur concrete (9). The above example would require about 20 lb of chopped fiber per ton of asphalt concrete mix, which would cost somewhere between $10 and $30/ton of mix. Therefore, the addition of even such a small proportion of fibrous material can add substantially to the total materials cost and may double the cost of a standard hot-mix formulation. Waste tire cord, if it can be recovered in the appropriate length, may represent a potential source of cheaper fibers for use in road construction.

Coupling agents are commonly used in the plastics industry to establish permanent bonds between inorganic fillers (such as glass) and polymeric organic molecules (17). The adverse effect of moisture on the mechanical properties of glass fiber composites is well known. Consequently, it is common practice to coat glass fibers with special organosilanes or chromium complexes (18) to establish strong covalent bonds that are less susceptible to the harmful effects of moisture. It is likely that such chemical reagents would be equally effective in asphalt concrete, except that cost prohibits their use.

Fortunately, there are cheap alternatives that are commercially available for this purpose. First observed by Fromm, the addition of iron oxide or iron derivatives to asphalt in minute quantities helps to prevent the loss of adhesion between silica and silicate-type aggregates in the presence of moisture. Iron naphthenates, commonly employed as driers in paints, are effective at concentrations of less than 0.1 percent in asphaltic bitumen. Waste iron oxides from iron smelters, which are recovered as finely divided powders, are also effective. The latter are available at prices near $50/ton and, at the small concentrations required, would help to ensure pavement stability without incurring significant added costs. The influence of iron compounds on limestone aggregates is unknown. However, contact-angle measurements may help to anticipate the influence of water on limestone and other aggregates.

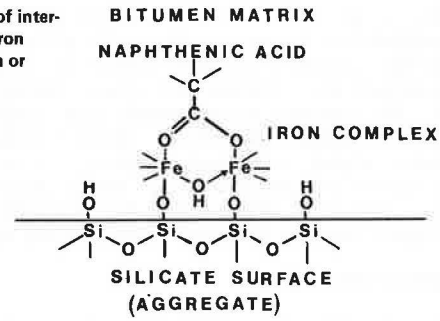


Figure 6. Schematic of interfacial bond between iron naphthenate and silica or silicate surface.

The unusual effect of iron compounds on the adhesion of bitumen to silicate or glass materials is attributed to strong coordinate or covalent bonding, as suggested in Figure 6. The cohesive bond is largely made up of van der Waals forces. Similar interfacial structures have been identified in chromium derivatives that are used in the treatment of glass fibers (18).

A cautionary note should be added concerning the long-term effect of iron naphthenate on the oxidative hardening of asphalt. Since iron naphthenate is a commercial paint drier, it therefore promotes air oxidation of unsaturated hydrocarbons and may cause premature embrittlement of paving concretes. On the other hand, Fromm observed that asphalt pavements that contain a reddish-brown aggregate were particularly outstanding with respect to water stripping and deterioration, an observation that prompted a further study of the phenomenon and the discovery of soluble iron compounds as adhesion promoters (12).

CONCLUSIONS

Additives can help to alleviate some of the problems associated with the deterioration of asphalt concrete paving materials. It has been shown that, although the incorporation of reclaim rubber into asphalt concrete reduces the brittle temperature by as much as 10°C, it cannot eliminate cracking entirely. Under favorable conditions, the addition of chopped tire cord or other fibrous material to asphalt concrete can increase fracture toughness by a factor of 20. If possible, the process for the recovery of powdered reclaim from discarded tires should be modified to permit the recovery of tire cord in short lengths of about 5 cm.

The incorporation of iron oxide or iron naphthenate into a standard paving mix can help to improve adhesion of the bitumen to the aggregate and perhaps reduce the incidence of stripping, cracking, spalling, ravelling, and swelling due to attack by moisture. Iron oxide of suitable quality is available as a by-product from most steel foundries.

The combined use of rubber, fibers, and adhesion promoters should help to prolong the lifetime of asphalt concrete paving materials and reduce maintenance and thereby justify the additional cost.

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REFERENCES

1. J. Shim Ton, K.A. Kennedy, M.R. Piggott, and R.T. Woodhams. Rubber Chemistry and Technology, Vol. 53, No. 1, 1980, pp. 88-106.
2. M.R. Piggott and R.T. Woodhams. Recycling of Rubber Tires in Asphalt Paving Materials. Environment Canada, Toronto, 1979.
3. J.L.M. Scott. Use of Rubber Asphalt Binder with Graded Aggregate for Seal Coats. Surfacing Branch, Saskatchewan Highways and Transportation, Regina, Canada, 1979.
4. A. French. RA-3 Rubberized Asphalt. Metro Toronto Roads and Traffic Department, Toronto, 1978.
5. J.I. McDougall. Rubberized Asphalt Concrete Memorandum Ref. 3162-2-1-10. Bituminous Section, Ontario Ministry of Transportation and Communications, Downsview, 1979.
6. M.R. Piggott. Methods of Making Composites More Tough. Centre for the Study of Materials, Univ. of Toronto, Canada, 1981.
7. G.C. Hoff and others. Chemical, Polymer, and Fiber Additives for Low Maintenance Highways. Noyes Data Corp., Park Ridge, NJ, 1979.
8. A. Neville and others, eds. Fibre Reinforced Cement and Concrete. Construction Press, Lunesdale House, Hornby, Lancaster, Ontario, 1975.
9. J. Ho and R.T. Woodhams. Fiber Reinforced Sulphur Concrete. Proc., Sulphur 81 Conference, Calgary, Alberta, Canada, May 25-28, 1981.
10. L. Ciplijauskas, M.R. Piggott, and R.T. Woodhams. Industrial and Engineering Chemistry, Vol. 18, 1979, pp. 86-91.
11. J.A.N. Scott. Adhesion and Disbonding Mechanisms of Asphalt Used in Highway Construction and Maintenance. Proc., AAPT, 1978.
12. H.J. Fromm. The Mechanisms of Asphalt Stripping from Aggregate Surfaces. Research and Development Division, Ministry of Transportation and Communication, Downsview, Ontario, Rept. RR 190, 1974.
13. R.J. Good and R.R. Stromberg, eds. Surface and Colloid Science: Experimental Methods. Plenum Press, New York, 1979.
14. S. Varevorakul. Adhesion Promoters in Bitumen. Department of Chemical Engineering and Applied Chemistry, Univ. of Toronto, B.A.Sc. thesis, 1981.
15. D.W. van Krevelen and P.J. Hoftyzer. Properties of Polymers, 2nd ed. Elsevier, New York, 1976.
16. R.D. Deanin and A.M. Crugnola, eds. Toughness and Brittleness of Plastics. American Chemical Society, Washington, DC, Advances in Chemistry Series 154, 1976.
17. Interfaces in Composites. American Society for Testing and Materials. Philadelphia, ASTM Special Tech. Publ. 452, 1968.
18. Quilon and Volan Chromium Complexes. E.I. du Pont de Nemours Tech. Brochure, Wilmington, DE, 1979.

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Evaluation of Filler Effect of Sulfur in Asphalt Binder

AMIR F. BISSADA

The effectiveness of both dissolved and dispersed crystalline parts of sulfur in asphalt binders used in road-paving mixtures was evaluated. The role of sulfur as a binder extender and filler material was compared with that of conventionally used limestone filler. For this purpose, a series of viscosity measurements were performed on sulfur-extended asphalt (SEA) binders and limestone filler-extended asphalt (FEA). Ratios vary up to 50 percent by weight at a temperature range from 25°C to 140°C. Rotational and cone-plate viscometers were used in this study to evaluate the relative viscosity, temperature susceptibility, and shear susceptibility of these binders as a function of the volume concentration. The research indicates that (a) the effect of the dissolved part of sulfur in asphalt cement on the decrease in viscosity is balanced by the filler effect of a certain part of dispersed sulfur particles, which varies in its effective volume concentration according to the temperature of the SEA binder; (b) at temperatures below the softening point of asphalt cement, values of effective volume concentration for both SEA and FEA are found to be similar; and (c) the crystalization process of sulfur in terms of dynamic growth of the sulfur particles seems to be effective if the SEA is mixed with mineral aggregates and represents the major part of stiffness improvement of finished pavement with age. Increase in viscosity of SEA due to increase of effective volume concentration of dispersed sulfur particles represents only a minor part of the stiffening effect.

During the past seven years, sulfur-extended asphalt (SEA) materials have been used to pave experimental road sections in many parts of the United States and in the Persian Gulf states in the Middle East. Investigations of the engineering properties of sulfur asphalt-paving mixtures and their structural response against both fatigue and permanent deformation have suggested that additional comprehensive evaluation of the SEA used and of its role as a binder extender as well as a filler material were warranted (1-4).

SEA is produced by high-shear-rate mixing of liquid sulfur with liquid asphalt. A part of the added sulfur by the dispersion technique ends up as dissolved sulfur. It is believed that the remainder