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**IMPROVEMENTS IN ROLLED ASPHALT SURFACINGS  
BY THE ADDITION OF SULPHUR**

**by**

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# IMPROVEMENTS IN ROLLED ASPHALT SURFACINGS BY THE ADDITION OF SULPHUR

## ABSTRACT

Rolled asphalt is used as the wearing course on most heavily trafficked roads in Britain, and on the majority of these it performs very well. However, some conventional rolled asphalts have insufficient resistance to permanent deformation for roads which carry the heaviest traffic.

In an effort to improve visco-elastic properties and resistance to permanent deformation, experiments have been carried out in which sulphur has been substituted for various proportions of the filler and bitumen components of the rolled asphalt mix. The properties of these modified mixes have been studied in the laboratory using the Marshall Test, the Wheel-tracking Test and measurements of dynamic modulus and fatigue life. These tests showed that resistance to permanent deformation should be improved by adding sulphur, with only a small reduction in laboratory fatigue-life.

Full-scale mixing and laying trials have also been carried out, and these showed that it is possible to manufacture and lay sulphur-modified asphalt providing its processing temperatures are carefully controlled. There are several problems that would need to be resolved, however, before this process could be used more widely.

The nature and structure of sulphur in asphalt has also been investigated and this has improved the understanding of the role of sulphur in the mixture and its likely effects on more practical aspects, such as handling and road performance.

## 1. INTRODUCTION

Rolled asphalt is used as the wearing course on most of the heavily trafficked roads in Britain, and on the majority of these it performs very well. However, on roads carrying the heaviest traffic, some conventional mixtures deform too rapidly. A number of ways of improving resistance to permanent deformation are therefore being investigated at TRRL<sup>1</sup>, including the use of heavy-duty bitumen and the use of organic polymers and sulphur as additives.

This report is concerned with the use of sulphur and its effects on the properties of rolled asphalt. The results of some of this work have been published elsewhere<sup>2</sup>.

## 2. THE ROLE OF SULPHUR IN BITUMINOUS MATERIALS

Sulphur can be incorporated into bituminous mixes so that when the sulphur is molten at mixing and laying temperatures, it improves workability of conventional mixes. When the sulphur solidifies on cooling it increases the stiffness of the compacted material.

The use of sulphur in this way has been the subject of much research in other countries<sup>3</sup>, particularly those where there is a plentiful supply of sulphur. Despite this research, however, many aspects of the use of sulphur have still to be investigated or fully understood. Consequently, the use of sulphur-modified mixes for road construction are, for the most part, still in various stages of evaluation.

In Britain, where the vast majority of sulphur for all uses has to be imported, there has been less interest shown in sulphur until recently, when the need for more durable bituminous materials to cope with the demands of very heavy traffic generated interest in additives of all kinds. It was to meet these demands, particularly with respect to the resistance to permanent deformation of rolled asphalt wearing courses, that a programme of research on additives was begun at TRRL in 1977.

Modification by sulphur is likely to increase the brittleness of a mix. Rolled asphalt, with its high resistance to fatigue cracking was, therefore, potentially very suitable for modification. However, no research to this end had been reported.

Another important factor supporting the inclusion of sulphur in the programme of research on additives was its relatively low price, when compared with that of other additives such as organic polymers. The price of liquid sulphur per tonne is approximately half that of bitumen at present, but as it is twice as dense as bitumen their prices are similar per unit volume. Given that sufficient sulphur would be available for road purposes its use should not, therefore, be uneconomic.

## 2.1 Theoretical background

It is well known that, because the normal binder is visco-elastic, permanent deformation in all bituminous materials occurs more rapidly at high road temperatures. Interparticular friction also plays an important part in resisting permanent deformation; in rolled asphalt the friction between particles in the fine aggregate matrix is of great importance. The addition of sulphur can affect this friction and also the rheological properties of the binder.

The amount of sulphur that will react with bitumen depends on the temperature and composition of the bitumen. It has been shown<sup>4,5</sup> that sulphur reacts predominantly with the so called naphthene-aromatic fraction of bitumen, either by adding to the molecule, or by oxidising it with the extraction of hydrogen as hydrogen sulphide. Between 119.3°C, the melting point of pure monoclinic sulphur, and 150°C, the reaction is mainly one of addition, producing an increase in the polar aromatic fraction and a relatively small change in rheological properties. Above 150°C the competitive oxidation reaction increases sharply, producing an increase in asphaltenes and an effect on the properties of the bitumen similar to that achieved by blowing with air at high temperatures.

The evolution of hydrogen sulphide is obviously undesirable, and in any case in the present work the object has not been to modify the bitumen; all mixing was therefore carried out below 150°C. The aim was to increase interparticular friction and so the quantities of sulphur used exceeded in every case the amount that would react with the bitumen and this ensured the presence of free sulphur in the mix. It was envisaged initially that during compaction this sulphur would partially fill and conform to the voids between sand particles, and at the same time make the hot asphalt more workable because sulphur is a low-viscosity liquid when at temperatures between its melting point and about 160°C. Above 160°C, however, its viscosity increases sharply through the formation of sulphur polymers. On cooling the sulphur was expected to crystallise and thereby increase interparticular friction within the asphalt.

In practice, the stiffening mechanism of the sulphur was not quite as simple as this, and changes in the behaviour and properties of asphalt modified by sulphur made it necessary to look beyond the initial objective of improving resistance to permanent deformation. For this reason the effects of sulphur on the stiffness and laboratory fatigue-life of rolled asphalt have also been measured. Additionally, some study has been made of the stiffening mechanism and structure of the sulphur in asphalt. This has not answered all the questions arising from observations of the behaviour and properties of sulphur-asphalt, but it has contributed information relevant to its manufacture and road performance.

### 3. LABORATORY MEASUREMENTS

#### 3.1 Formulation of mixes

All the mixes studied were based on the aggregate gradings in Table 1. The same aggregates were used throughout, and the angularity of the sand chosen was intermediate in the range available in the UK. A bitumen with a Penetration value of 50 at 25°C and a Ring and Ball Softening Point of 52°C was used in each mix.

**TABLE 1**  
Aggregate gradings for laboratory mixes

Type	Grading	
	Sieve size	% by mass passing
Coarse aggregate (crushed rock)	14 mm	100
	10 mm	50
	6.3 mm	16.7
	3.35 mm	0.0
Fine aggregate (sand)	2.36 mm	100
	600 μm	98
	212 μm	46
	75 μm	0.7
Filler (CaCO <sub>3</sub> )	75 μm	88.5

The first step in formulating these mixes was to substitute systematically sulphur for filler and bitumen, because it was envisaged that the sulphur would contribute to filling the voids between sand particles. The Marshall Test was then used to screen the mixes generated by this process to select the most promising ones for further testing. The compositions of the mixes that formed the starting point for the Marshall tests are given in Table 2.

The optimum composition of the sand-filler-bitumen mixture (Mix A of Table 2) was determined according to the Marshall test procedure of British Standard BS 594<sup>6</sup>. Various proportions of the filler and bitumen components of this mix were then replaced by sulphur on a volumetric basis. Mixes B to F represent volumetric replacements of bitumen of 15, 25, 30, 37.5 and 50 per cent, respectively. Similarly, Mixes G to I represent volumetric replacements of filler of 25, 37.5 and 50 per cent. Mix J represents a 25 per cent by volume replacement of both filler and bitumen.

**TABLE 2**

Composition of mixes in which sulphur was substituted for bitumen and filler

Mix	Per cent by mass of total mix			
	Sand	Filler	Sulphur	Bitumen
A	75.8	13.9	—	10.3
B	75.6	12.6	3.2	8.6
C	74.8	12.5	5.2	7.5
D	74.4	12.4	6.2	7.0
E	73.9	12.3	7.6	6.2
F	72.9	12.1	10.1	4.9
G	77.4	9.7	2.5	10.4
H	77.7	8.1	3.8	10.4
I	78.0	6.5	5.1	10.4
J	75.3	9.5	7.6	7.6

If the sulphur fills the voids between sand particles in a manner similar to bitumen or filler, then by introducing sulphur to Mix A in this way the combined volume of filler, bitumen and sulphur should have been at an optimum level in each of the above mixes. However, to check this hypothesis the bitumen content of each sulphur mix in Table 2 was varied over a narrow range until an optimum was established.

The results of Marshall tests on these mixes were used to formulate new mixes that contained approximately 30 per cent coarse aggregate, for wheel-tracking, modulus and fatigue tests. The compositions of these mixes are given in Section 3.3.

### 3.2 Preparation of mixes

In the preparation of laboratory specimens whose properties are compared in the following Sections, the sulphur was added in the form of a liquid. It was not until a substantial number of specimens had been prepared in this way that it was discovered that the use of solid sulphur would have reduced the time lapse between manufacture and testing (Section 3.3). However, as the final properties of the asphalt were largely independent of the form in which the sulphur was added, and to maintain continuity, liquid sulphur was used throughout.

Before mixing, the aggregate and binder were heated to 150°C and the sulphur to 130°C. All the components were added to the mixing bowl before mixing started, and each sample was mixed at a temperature between 145 and 150°C for four minutes. All samples were compacted at between 120 and 125°C. With these exceptions the preparation of Marshall specimens followed the procedure in BS 594<sup>6</sup>. Samples containing coarse aggregate were cast into slabs approximately 300 x 300 x 50 mm and compacted by rolling in a laboratory roller-compactor. Subsequently, these were cut to the appropriate size for testing.

### 3.3 Engineering properties

The Marshall Test provides a well established method of assessing basic properties such as the stability of a mix, and it is a relatively quick procedure. Marshall stability values are also known to correlate well with resistance to permanent deformation measured in the TRRL Wheel-tracking Test<sup>7</sup>. The Marshall Test was used, therefore, to examine the large number of mixes arising from a systematic replacement of bitumen and filler by sulphur, to identify the more promising ones, before proceeding to wheel-tracking tests and measurements of modulus and fatigue life.

At the beginning of this screening process an important feature of the stiffening mechanism of sulphur was discovered. The first batch of Marshall specimens was mixed and tested on successive days. The results were surprising because the addition of sulphur had reduced Marshall stability rather than increased it. Further investigations confirmed that stability was lowered for the first few days after manufacture. Subsequently it rose, and in the case of an asphalt in which 25 per cent by volume of the bitumen had been replaced by sulphur it reached a value approximately twice that of the unmodified asphalt. The time taken to achieve maximum stability ranged from 7 to 21 days, depending on the form in which the sulphur was added. The changes in Marshall stability with time are illustrated in Figure 1.

As this behaviour, together with the structure of the sulphur in asphalt, could have important effects on the manufacture and performance of sulphur-asphalt, it was investigated further. The results obtained are described in Section 3.4. In the meantime, to take into account this ageing effect in a consistent way, all measurements of engineering properties were made on samples that had been stored for three weeks at room temperature.

**3.3.1 Marshall tests.** The results of the Marshall tests are summarised in Table 3. These tests were made using the procedure in BS 594<sup>6</sup>, except that four samples were tested at each bitumen content instead of two. In this procedure the optimum bitumen content is determined as an average of the bitumen contents corresponding to maxima in stability, mix density and compacted aggregate density. However, as Figure 2 shows, varying the bitumen content of the mixes in Table 2 that contained sulphur did not give rise to a maximum in stability in the range studied. With lower bitumen contents a maximum would probably have been established but these compositions were not tried; such lean mixes would almost certainly lack resistance to abrasion and crack too easily. Therefore, the optimum binder contents for the sulphur mixes, quoted in Table 3, are an average of the bitumen contents corresponding to maxima in mix density and compacted aggregate density only.

When bitumen was replaced by sulphur the optimum bitumen contents determined by the Marshall method were close to those predicted in Table 2. The stabilities of these mixes were much higher than that of the unmodified mix. As the sulphur content was increased, stability increased and flow values decreased. Void content also increased with increasing sulphur contents.

Progressively replacing filler by sulphur resulted in a very small increase in stability and a much lower optimum bitumen content than predicted. The optimum bitumen content was also lower than predicted when both filler and bitumen were partially replaced by sulphur, but this did result in a much higher stability than that of the unmodified asphalt.



**TABLE 3**  
Marshall test results

Mix*	Optimum bitumen content % by mass of mix	Properties at optimum bitumen content					
		Mix density gm/cc	Compacted aggregate density gm/cc	Voids in mix % of total volume	Marshall stability kN	Marshall flow mm	Marshall quotient kN/mm
a	10.3	2.126	1.907	6.7	4.0	4.4	0.9
b	7.5	2.094	1.872	11.0	5.0	3.7	1.4
c	7.4	2.161	1.890	7.6	8.3	3.1	2.7
d	6.4	2.184	1.908	8.0	10.8	2.5	4.3
e	6.0	2.190	1.889	8.1	14.0	2.5	5.6
f	5.1	2.199	1.867	8.1	23.0	3.1	7.4
g	8.6	2.097	1.917	9.7	4.3	3.8	1.1
h	8.0	2.087	1.919	10.5	4.2	3.1	1.4
i	6.9	2.064	1.924	12.0	4.6	2.3	2.0
j	5.9	2.151	1.910	9.7	13.4	2.5	5.4

\* The compositions of these mixes correspond to those of Table 2 except that the bitumen content shown here is the optimum determined by the Marshall procedure.

**3.3.2 Wheel-tracking tests.** The Wheel-tracking Test can be used to predict the permanent deformation of conventional rolled asphalt wearing courses on straight sections of carriageway. It has been established<sup>1,7</sup> that wearing courses are required with tracking rates of 2.0 mm/hour or less at 45°C to cope with the highest traffic levels under normal conditions. It is possible, however, that even better wearing courses may be required where vehicles turn and brake, because these manoeuvres impose shear stresses that are of the same order of magnitude as the vertical loading.

Mixes K, L and M of Table 4 were designed and manufactured on the basis of the Marshall Test results, and in particular the results of Marshall stability. These mixes correspond to Mixes c, d and j of Table 3, respectively, but have been adjusted to contain approximately 30 per cent by mass of coarse aggregate. Mixes e and f of Table 3 were rejected because their high sulphur contents made them too friable for practical purposes.

Wheel-tracking rates were measured at 45°C, over a range of bitumen contents that included those predicted as the optima and given in Table 4. The results are shown in Figure 3, together with those obtained for similar samples without sulphur, based on Mix a of Table 3.

**TABLE 4**  
Composition of mixes with coarse aggregate

Mix	Per cent by mass of total mix				
	Coarse aggregate	Sand	Filler	Sulphur	Bitumen
K	27.0	54.0	9.0	4.1	5.9
L	26.9	53.9	9.0	5.0	5.2
M	27.3	54.5	6.8	6.0	5.4

At the predicted optimum bitumen contents (Mixes K, L and M) the mixes with sulphur had much higher resistances to permanent deformation than the mix without sulphur, and their tracking rates were well below 2.0 mm/hour at 45°C.

Having established that a high resistance to permanent deformation can be achieved by introducing sulphur, it was necessary to check the effect of this on other important properties such as load-spreading and fatigue.

**3.3.3 Dynamic stiffness.** Measurements were made in flexure on beams of approximately 375 x 100 x 50 mm, using a sinusoidal loading pattern. The complex modulus, and its storage and loss components, were determined at various temperatures between 0 and 33°C, over a frequency range from 0.1 to 80 Hz. A method of reduced variables<sup>8</sup> was used to determine a relationship between the effects of temperature and frequency on the visco-elastic functions; this enabled the functions to be plotted over a wide range of frequencies at any desired temperature.

So far Mix K and the control mix based on Mix a of Table 4, have been studied in this way. The complex modulus for each mix was reduced to 33°C, and is plotted against test frequency in Figure 4. At frequencies between 5 and 10 Hz, which broadly correspond to the loading times most relevant to road conditions, the stiffness of the sulphur mix is around 2.5 times that of the unmodified mix at this temperature. The divergence of the curves towards the lower frequencies indicated that at higher temperatures the difference in stiffness would be even greater. These initial results show that the introduction of sulphur can produce a significant improvement in load-spreading ability, and further research is in progress to study this effect in more detail.

A comparison of the storage and loss moduli of the same mixes revealed differences similar to those observed between the complex moduli. The difference between the loss moduli, which are measures of the energy dissipated in the material by viscous flow, is supported by the results from the wheel-tracking tests and further confirms the improvement in resistance to permanent deformation.

**3.3.4 Fatigue life.** The addition of sulphur to any bituminous material is likely to make it more brittle and less resistant to fatigue cracking. However, fatigue cracking is rarely observed in rolled asphalt, partly because of its high bitumen content. It was hoped, therefore, that some of this bitumen could be replaced by sulphur without seriously affecting the fatigue life of the material. Because there is so little evidence of failure by fatigue of rolled asphalt wearing courses, an authoritative correlation between laboratory fatigue behaviour and road performance has not been established. However, laboratory results are of value when used for comparative purposes, and it was on this basis that measurements of stiffness and fatigue life were made on Mix K and the unmodified control mix used in the wheel-tracking and dynamic stiffness tests.

Measurements were made on 225 x 50 x 50 mm samples loaded in uniaxial alternating compression and tension at constant temperatures of 25 and 35°C, and over a range of stress amplitudes. The stress amplitude was kept constant throughout each test, and in every case the test frequency was 25 Hz. Stiffness measurements are given in Table 5, in terms of complex modulus, and the characteristic curves of initial strain versus fatigue life are plotted in Figure 5.

**TABLE 5**

Complex moduli measured in the uniaxial fatigue test

Mix	Stiffness (complex modulus) Nm <sup>-2</sup>	
	25°C	35°C
K	2.8 x 10 <sup>9</sup>	1.0 x 10 <sup>9</sup>
Control	1.6 x 10 <sup>9</sup>	4.5 x 10 <sup>8</sup>

At 25°C the sulphur mix was approximately 1.8 times stiffer than the unmodified mix, and 2.5 times stiffer at 35°C. This agreed very well with the complex modulus measurements made in flexure.

Figure 5 shows that for any given initial strain the sulphur asphalt failed after fewer load cycles than the control mix at both 25 and 35°C. However, the reduction in laboratory fatigue-life in these initial tests is not sufficiently great to suggest that the performance of sulphur-asphalt on the road will necessarily be significantly worse than that of conventional mixes. Further evaluation is planned of this aspect of performance.

### 3.4 Crystallisation of sulphur in asphalt

The most likely explanation for the changes in Marshall stability with time, described in Section 3.3, was that the crystallisation of the sulphur was retarded, perhaps by some interaction with the bitumen. When no crystalline sulphur could be seen during the first few days after manufacture, using an optical microscope, it was suspected that the start of crystallisation was coincident with the point at which stability begins to rise, but later experiments showed this to be incorrect.

It was important to try to understand the role of the sulphur more fully, because it affected practical aspects such as the workability of the asphalt during laying and its subsequent road performance. For example, it has been reported<sup>9</sup> that if compaction is carried out after the sulphur crystals begin to form, they are broken and their effect is reduced. Consequently, the choice of compaction temperature will be influenced by the crystallisation of the sulphur, which will in turn depend on the structure of the sulphur and the presence of dissolved impurities that will have a suppressing effect on crystallisation. After compaction the stability of the sulphur structure could also influence the long-term stability of the asphalt.

**3.4.1 Effect of compaction temperature on stability.** Asphalt specimens with compositions similar to Mixes a and c of Table 3 were prepared according to the procedure described in Section 3.2, except that they were compacted at temperatures ranging from 80 to 140°C. The densities and Marshall stabilities of these specimens, measured three weeks after manufacture, are plotted against compaction temperature in Figures 6 and 7, respectively.

The stability of the asphalt without sulphur was largely unaffected by compaction temperature in the range studied, but its density went through a maximum that corresponded to a compaction temperature of 110°C. In contrast with these results, those for asphalts containing sulphur showed that both stability and density were reasonably constant for compaction temperatures of between 105 and 140°C, but below 105°C they were sharply reduced. This sudden change strongly suggests that the sulphur begins

to crystallise at around 105°C, and that to obtain the best results compaction must take place above this temperature. A crystallisation temperature of 105°C would indicate that either the freezing point of the sulphur is depressed substantially or that the sulphur does not crystallise to the  $\beta$ -monoclinic allotrope; on cooling slowly from the melt under normal conditions sulphur crystallises first to the  $\beta$ -monoclinic allotrope at around 115°C and subsequently reverts to the orthorhombic allotrope below about 95°C.

A minimum compaction temperature of 105°C coupled with the slow crystallisation of sulphur suggested by the change in Marshall stability with time does, however, ease the rather strict constraint that would have been placed on compaction by a rapid crystallisation at the melting point of  $\beta$ -monoclinic sulphur.

**3.4.2 X-ray diffraction.** Powder diffraction patterns were obtained using Cu  $k_{\alpha}$  radiation for samples of sand asphalt compacted into small discs. For the samples containing sulphur, diffraction patterns were obtained at 1 hour, 3 weeks and 3 months after manufacture. Comparison of these patterns with those obtained for the samples without sulphur showed that a definite pattern was attributable to the sulphur, showing no sign of changing either with time or with exposure to the X-rays. The spacings between crystal planes (d-spacings) in the sulphur, calculated from the diffraction pattern, are given in Table 6 together with the relative intensities of the X-rays scattered from them.

These results strongly support the evidence from the compaction studies, which indicated that the sulphur begins to crystallise as soon as the asphalt cools. Furthermore, as the X-ray pattern obtained immediately after manufacture did not change with time or exposure to the X-rays, the sulphur appears to be reasonably stable, and therefore, unlikely to give rise to any long-term changes in the properties of the asphalt. To be certain of this, however, measurements would have to be made over a much longer period than 3 months.

**TABLE 6**

X-ray diffraction data for sulphur in asphalt

$2\theta^*$	d (Å)	Approximate relative intensity
14.6	6.06	20
16.4	5.40	15
18.2	4.87	15
19.3	4.59	80
19.7	4.50	40
22.0	4.04	10
23.2	3.83	80
23.6	3.77	60
24.0	3.70	80
24.5	3.63	50
25.1	3.54	80
25.9	3.44	100
27.5	3.24	30
28.2	3.16	60
30.0	2.98	10

\*  $\theta$  = angle between crystal planes and incident X-ray beam.

An alternative way of gaining information on the properties of sulphur in asphalt, including its stability, was to identify its structure as that of one of the known allotropes of sulphur. Identification by indexing the d-spacings calculated from the X-ray powder data was limited to the few allotropes of sulphur for which structures are known; most allotropes of sulphur are too unstable to withstand structural examination. However, given its apparent stability sulphur in asphalt was expected to be the orthorhombic allotrope with the Space Group  $D_{2h}^{24}$ , which is the only stable one under normal conditions at STP.

It was surprising, therefore, to find that the d-spacings for sulphur in asphalt could not be indexed according to this structure, or indeed according to any of those known for the other allotropes of sulphur. One possible reason for this could be the presence in the sulphur of impurities from other components of the asphalt. Alternatively, if the sulphur in asphalt is not orthorhombic its apparent stability could be the result of its being embedded in the asphalt. For example,  $\gamma$ -monoclinic sulphur is known to exist for years when embedded in a rubber solution, and it can withstand 40 hours of X-ray examination<sup>10</sup>. Furthermore, this allotrope melts at 106.8°C and its crystals grow in thin sheets that glitter brightly<sup>11</sup>, like those of sulphur in rolled asphalt. Despite these similarities an attempt to index the d-spacings for sulphur in asphalt according to the latest structure reported for  $\gamma$ -monoclinic sulphur<sup>12</sup> failed.

Some doubt remains, therefore, about the nature of sulphur in asphalt and although it is slight, the possibility of a change in the structure of the sulphur affecting the long-term stability of the asphalt should not be overlooked.

#### 4. FULL-SCALE MIXING AND LAYING

Experiments were carried out to determine the effects of sulphur on the mixing and laying characteristics of rolled asphalt, to monitor levels of hydrogen sulphide during manufacture of the asphalt, and to determine whether the improvements in the properties of asphalt manufactured in the laboratory could be reproduced on a large scale.

Sixty tonnes of sulphur-asphalt were manufactured and laid within the precincts of a contractor's premises, together with 50 tonnes of conventional asphalt as a control. The control asphalt was designed using the procedure in BS 594<sup>6</sup>. The sulphur asphalt was identical to the control except that 25 per cent by volume of the bitumen was replaced by sulphur. Full details of both types of asphalt are given in Table 7. Precoated 20 mm Craig-yr-Hesg chippings were applied to the surfaces of the asphalts to provide surface texture. The rates of spread of these chippings were not measured directly but estimated after laying by counting chippings over several square metres of the finished surfacings.

The control asphalt was mixed at about 180°C and compacted in a thickness of about 50 mm, with the temperature at the centre of the asphalt carpet between 140 and 150°C at the start of rolling. The contractor had considerable experience of mixing and laying asphalt of this composition, and advised that these temperatures would allow good compaction to be achieved together with sufficient embedment of the precoated chippings.

The sulphur asphalt was mixed at temperatures between 140 and 150°C, using a mixing time of approximately one minute, which was a few seconds less than the time used for the conventional asphalt, although this difference was not significant. The sulphur was added in the form of a coarse powder, directly to the mixing chamber in polythene bags. The use of pre-weighed bags, each containing 10 kg, enabled the

**TABLE 7**

Details of asphalts laid in full-scale experiments

	Composition % by mass					Quantity mixed (tonnes)	Mixing Temp. °C	Compaction Temp. °C	Approximate max. rate of spread of precoated chippings kg/m <sup>2</sup>
	Aggregate			Sulphur	Binder*				
	Coarse	Fine	Passing 75 µm sieve						
Unmodified asphalt	30	51.6	8.6	—	7.7	50	~180	140–150	8.5
Sulphur-asphalt	Limestone (Chipping Sodbury)	Hilton pit (Bridgenorth)	Limestone (Chipping Sodbury)	4.0	5.8	60	140–150	85–105	12

\* Bitumen (Penetration 48 at 25°C, Ring and Ball Softening Point 55°C).

correct amount of sulphur to be added quickly, and by putting in the bags as well the escape of sulphur dust into the atmosphere was eliminated (sulphur dust can flash-fire under some circumstances).

As long as the mixing temperature did not rise much above 150°C, the levels of hydrogen sulphide recorded a few feet from the open mixing chamber were about 1 ppm. However, when one batch of asphalt was mixed at 160°C the level of hydrogen sulphide rose sharply to between 10 and 12 ppm. This agrees closely with results from laboratory experiments using different mixing temperatures, and demonstrates that strict control of the mixing temperature is essential. A concentration above 10 ppm would, if continuous throughout the working day, exceed current Health and Safety Executive regulations<sup>13</sup>.

After mixing, the concentration of hydrogen sulphide close to the surface of the asphalt was barely measurable and never rose above about 1 ppm throughout the laying operation. Sulphur vapour was present, however, and this did cause some irritation of the eyes.

The handling and laying characteristics of the sulphur asphalt were good. It was laid in a thickness of approximately 50 mm, and rolling started with the temperature at the centre of the asphalt carpet at about 105°C. There was no discernible hardening of the asphalt as it cooled and it could still be worked and compacted at 85°C, with satisfactory embedment of the precoated chippings applied to the surface. This agreed with the results obtained in the laboratory that showed that crystallisation of the sulphur is a slow process although to get the best properties the asphalt should be compacted at 105°C or above. It was possible to start compacting at 105°C in these small trials because there was no delay between mixing and laying but special attention would have to be paid to achieving this when working under more normal conditions where asphalt is hauled longer distances to the laying site.

Tests on samples of asphalt taken from the laid surfacings showed that the sulphur had improved resistance to permanent deformation by a factor of approximately 2. There was, however, considerably more variability in the results from this full-scale experiment than in those from the earlier laboratory tests; variability should however be reduced as more experience of mixing and laying techniques is gained.

Obviously, the use of sulphur to modify asphalt is not without problems, but despite these the experiments described here have shown that the benefits from using sulphur asphalt would justify its further investigation in an experiment on a public highway.

## 5. CONCLUSIONS

1. The resistance to permanent deformation of rolled asphalt should be improved substantially by the addition of sulphur. Mixes in which 25 to 30 per cent of the bitumen was replaced by sulphur produced about a five-fold improvement in wheel-tracking-rate.
2. These sulphur-asphalts also had higher complex moduli than the equivalent unmodified asphalt. At 33°C and a test frequency of 10 Hz, the sulphur asphalt was approximately 2 to 3 times stiffer than the unmodified asphalt, indicating that load spreading ability could also be improved by adding sulphur.
3. Initial measurements of laboratory fatigue-life indicate that the improvements in load spreading ability and resistance to permanent deformation are accompanied by some reduction in resistance to fatigue cracking. The significance of this reduction requires further evaluation.

4. The mixing and laying temperatures for sulphur-asphalt must be controlled carefully, to avoid any emission of hydrogen sulphide and to achieve optimum properties on compaction. Full-scale mixing and laying trials showed that this can be done, at least well enough to carry out a full-scale road experiment, in the near future.
5. Although there is little doubt that sulphur can be used to improve the engineering properties of rolled asphalt, the practicability of its use under more normal contractual conditions has still to be established. The decision whether or not to invest the effort needed to do this will depend largely on the results from the planned road experiment.

## 6. ACKNOWLEDGEMENTS

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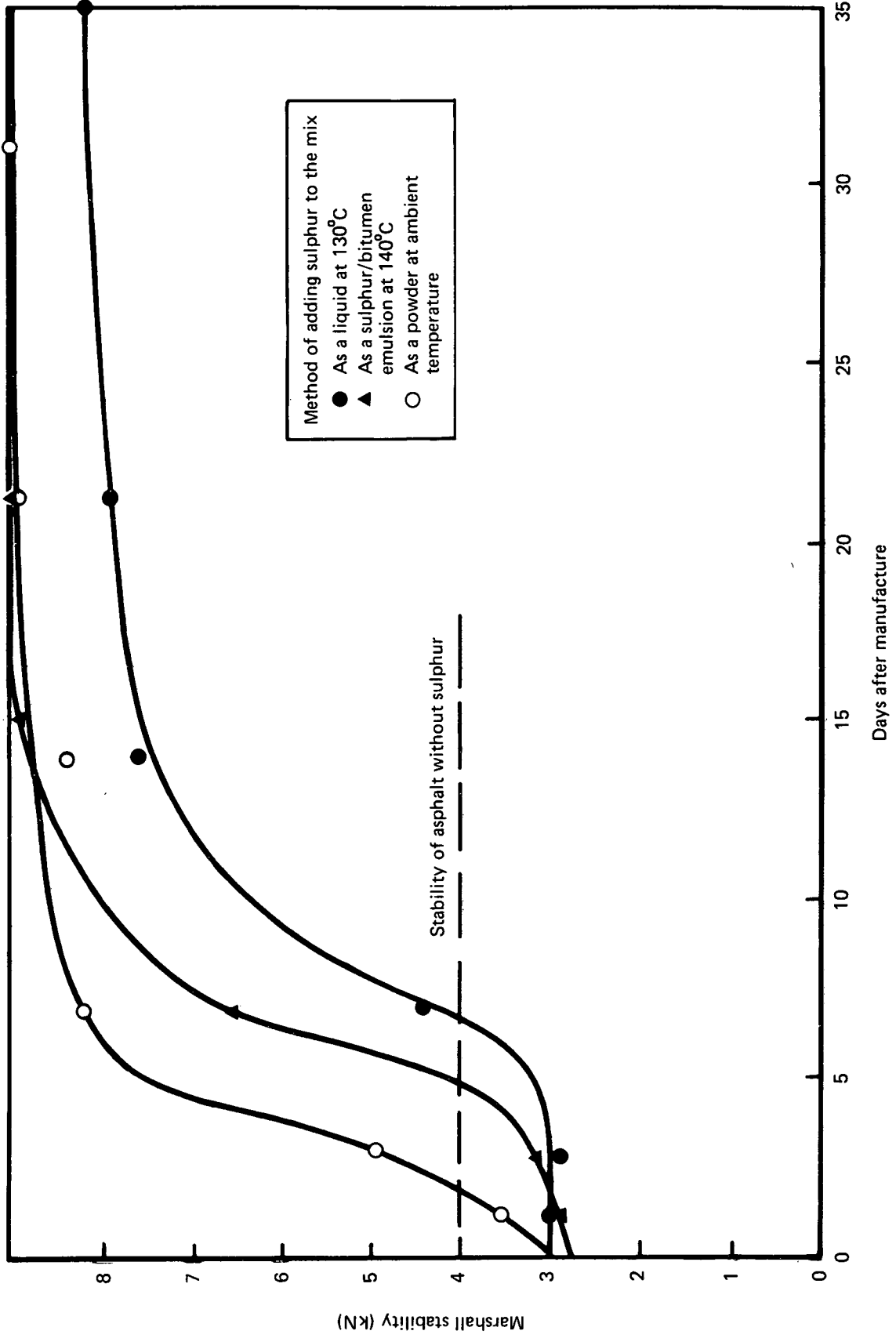
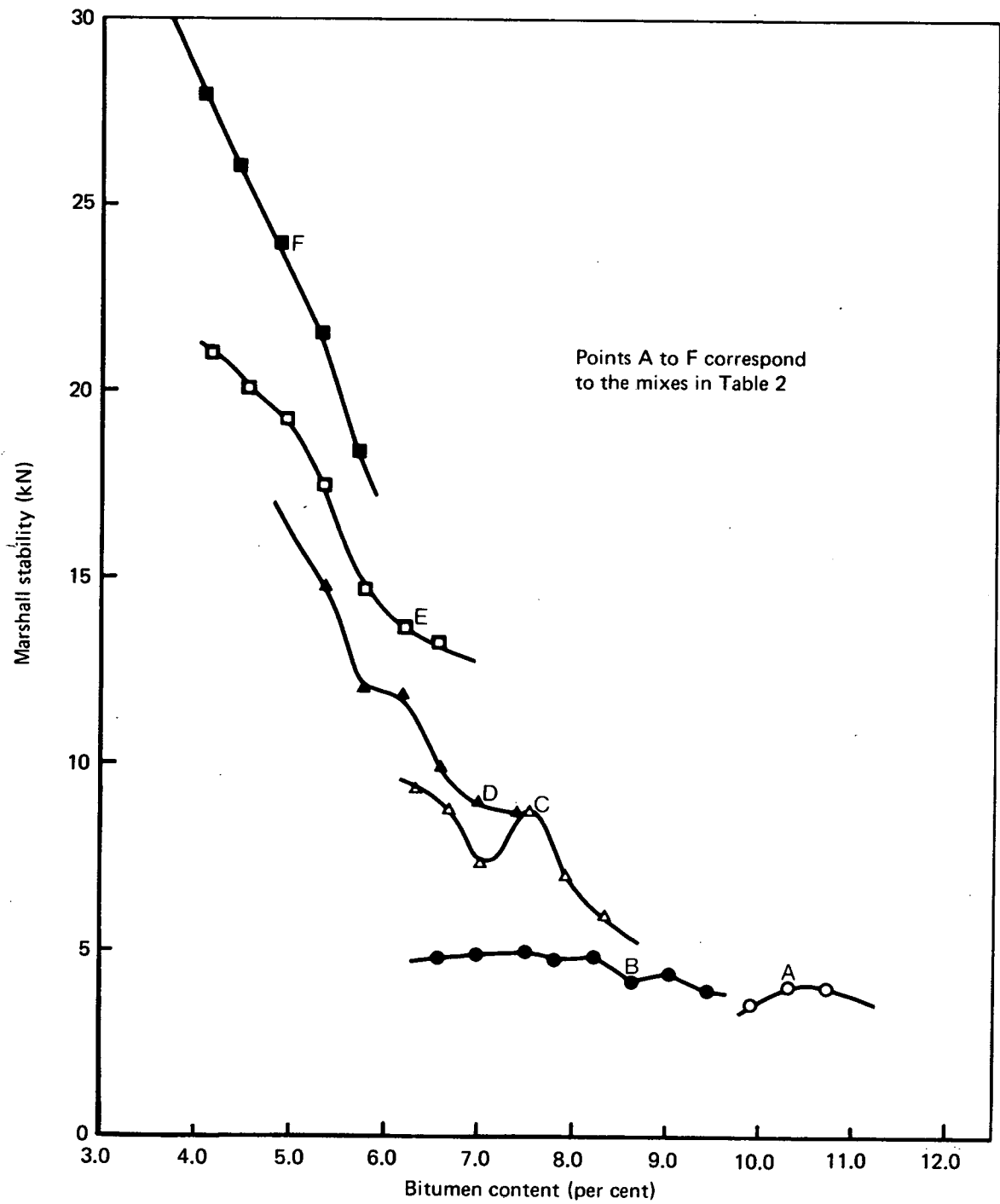


Fig. 1 CHANGE IN MARSHALL STABILITY OF A SULPHUR-ASPALT WITH TIME



**Fig. 2 RELATIONSHIP BETWEEN BITUMEN CONTENT AND MARSHALL STABILITY FOR SULPHUR-ASPHALTS**

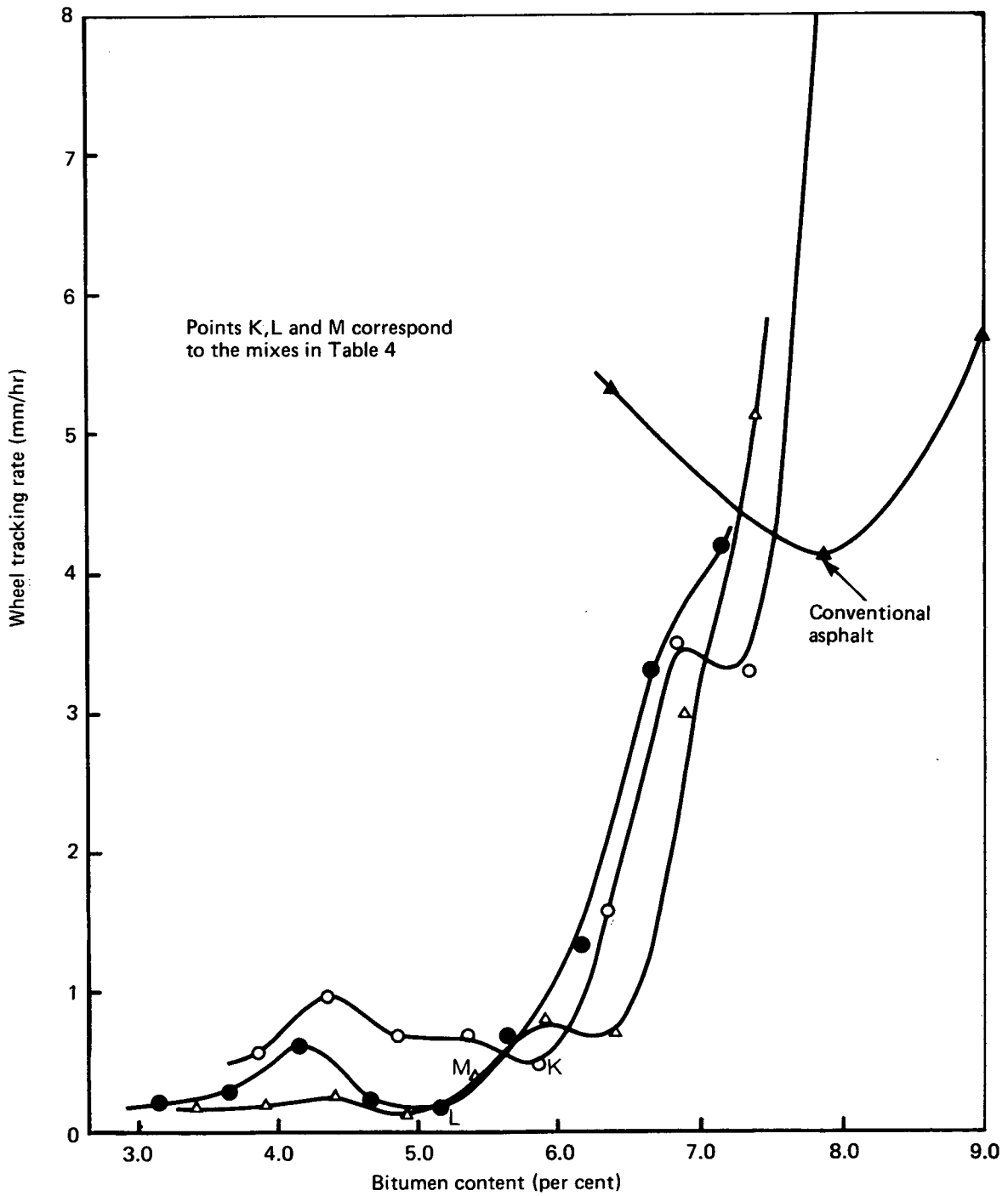
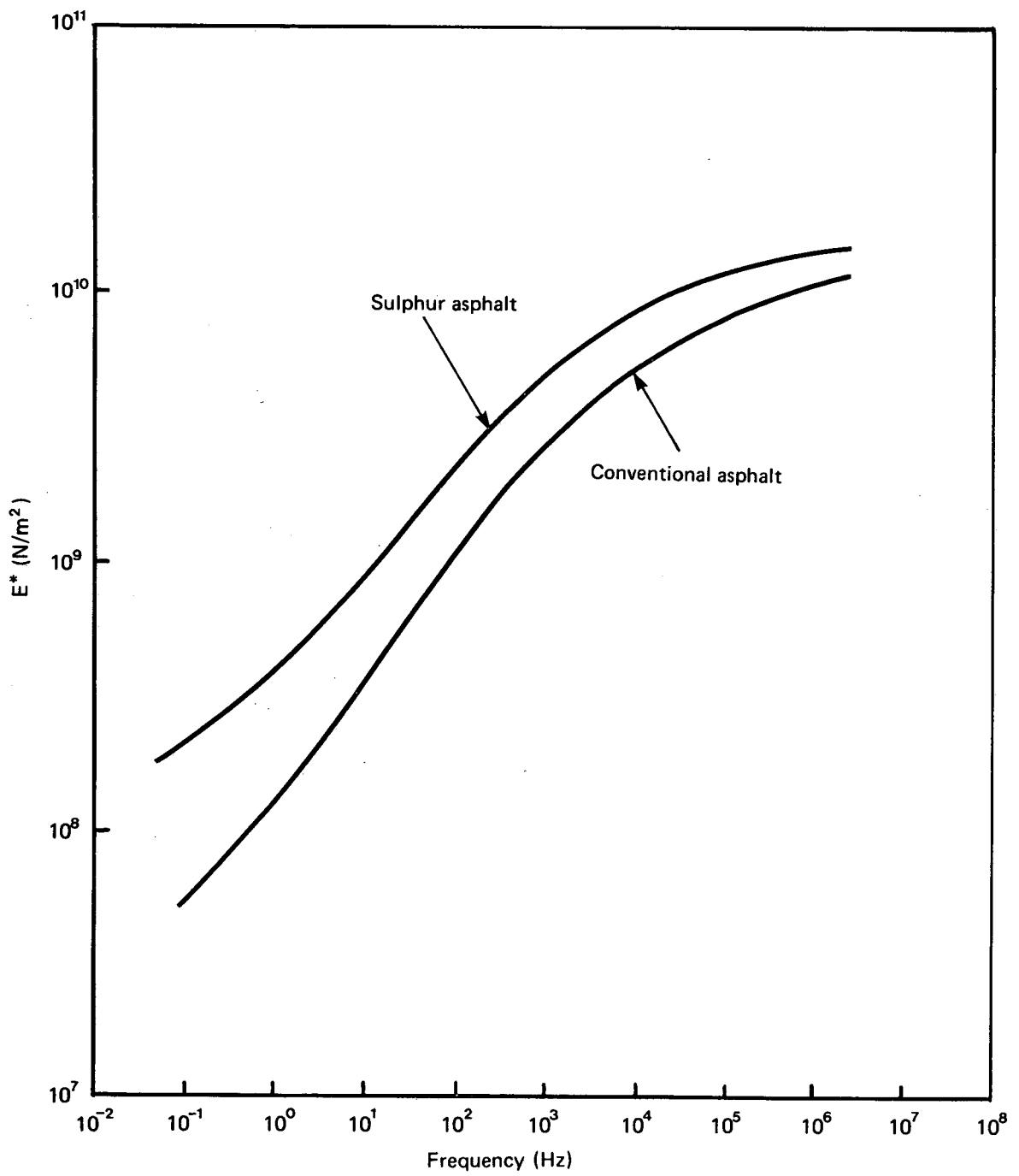
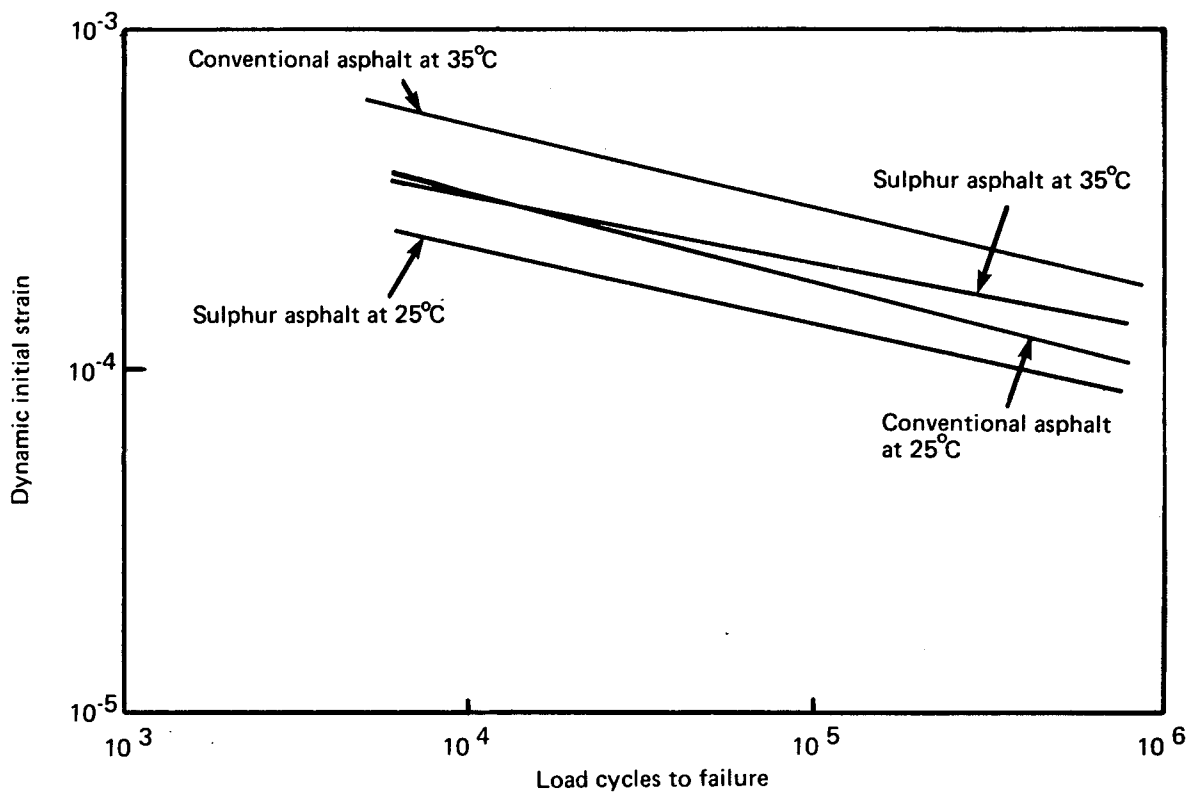


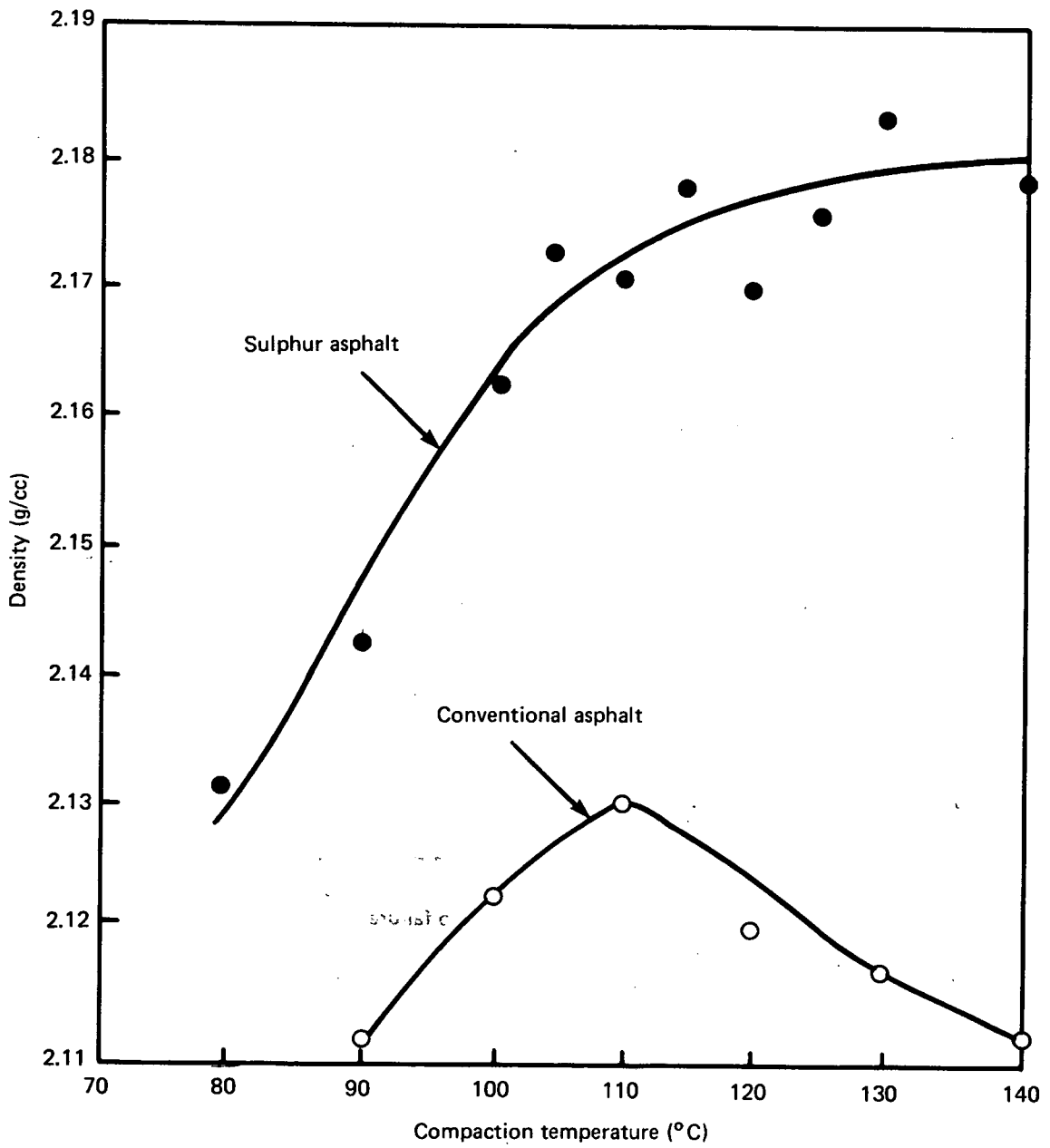
Fig.3 RELATIONSHIP BETWEEN BITUMEN CONTENT AND TRACKING RATE FOR SULPHUR-ASPHALTS AT 45°C



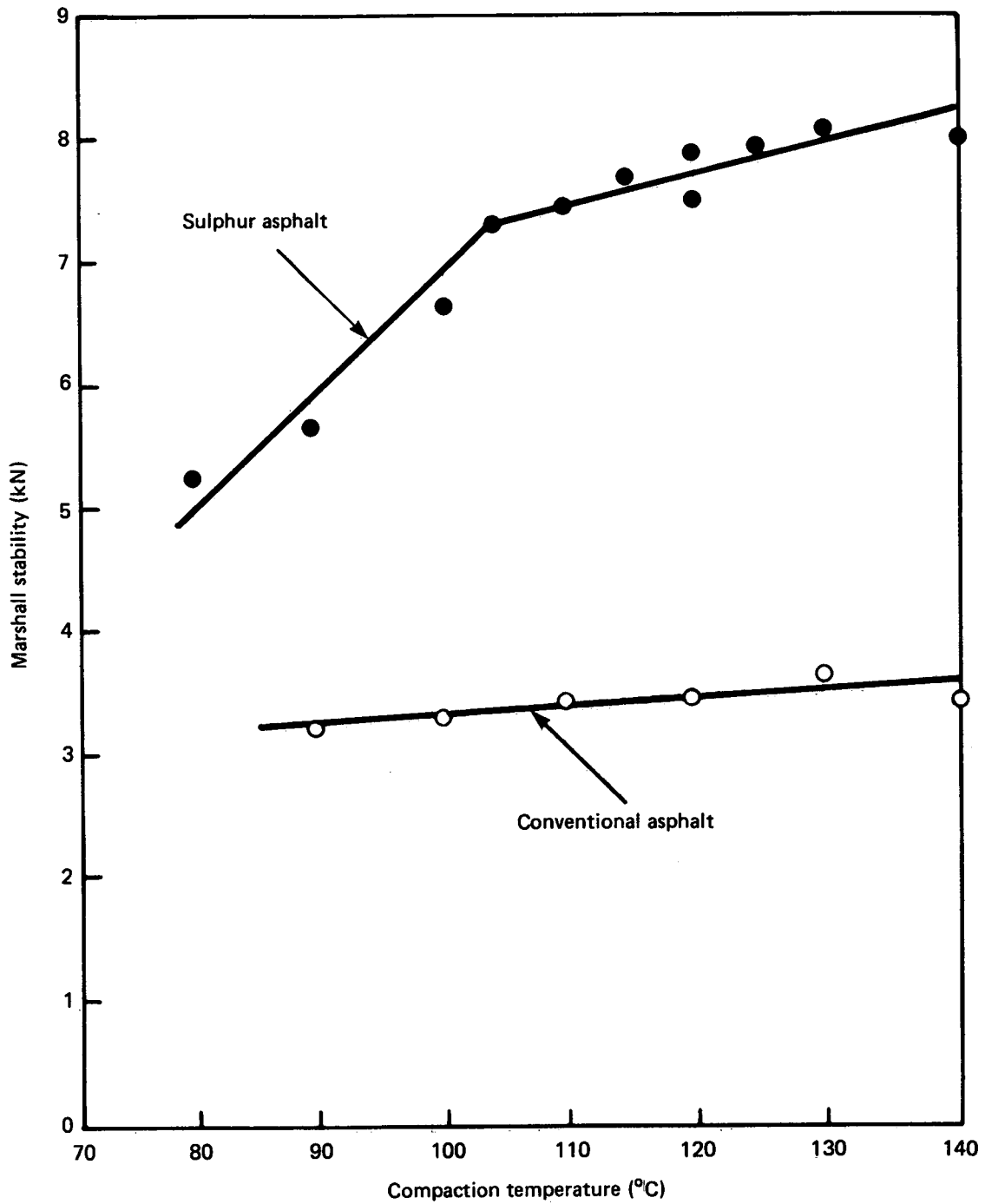
**Fig. 4 RELATIONSHIP BETWEEN COMPLEX MODULUS AND TEST FREQUENCY FOR A SULPHUR-ASPHALT AND A CONVENTIONAL ASPHALT**



**Fig.5 RELATIONSHIP BETWEEN LABORATORY FATIGUE LIFE AND DYNAMIC INITIAL STRAIN FOR A SULPHUR-ASPALT AND A CONVENTIONAL ASPHALT**



**Fig. 6 RELATIONSHIP BETWEEN MIX DENSITY AND COMPACTION TEMPERATURE**



**Fig. 7 RELATIONSHIP BETWEEN MARSHALL STABILITY AND COMPACTION TEMPERATURE**



## ABSTRACT

**Improvements in rolled asphalt surfacings by the addition of sulphur:** J H DENNING BSc PhD CChem MRSC and J CARSWELL BSc: Department of the Environment Department of Transport, TRRL Laboratory Report 963: Crowthorne, 1981 (Transport and Road Research Laboratory). Rolled asphalt is used as the wearing course on most heavily trafficked roads in Britain, and on the majority of these it performs very well. However, some conventional rolled asphalts have insufficient resistance to permanent deformation for roads which carry the heaviest traffic.

In an effort to improve visco-elastic properties and resistance to permanent deformation, experiments have been carried out in which sulphur has been substituted for various proportions of the filler and bitumen components of the rolled asphalt mix. The properties of these modified mixes have been studied in the laboratory using the Marshall Test, the Wheel-tracking Test and measurements of dynamic modulus and fatigue life. These tests showed that resistance to permanent deformation should be improved by adding sulphur, with only a small reduction in laboratory fatigue-life.

Full-scale mixing and laying trials have also been carried out, and these showed that it is possible to manufacture and lay sulphur-modified asphalt providing its processing temperatures are carefully controlled. There are several problems that would need to be resolved, however, before this process could be used more widely.

The nature and structure of sulphur in asphalt has also been investigated and this has improved the understanding of the role of sulphur in the mixture and its likely effects on more practical aspects, such as handling and road performance.

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