

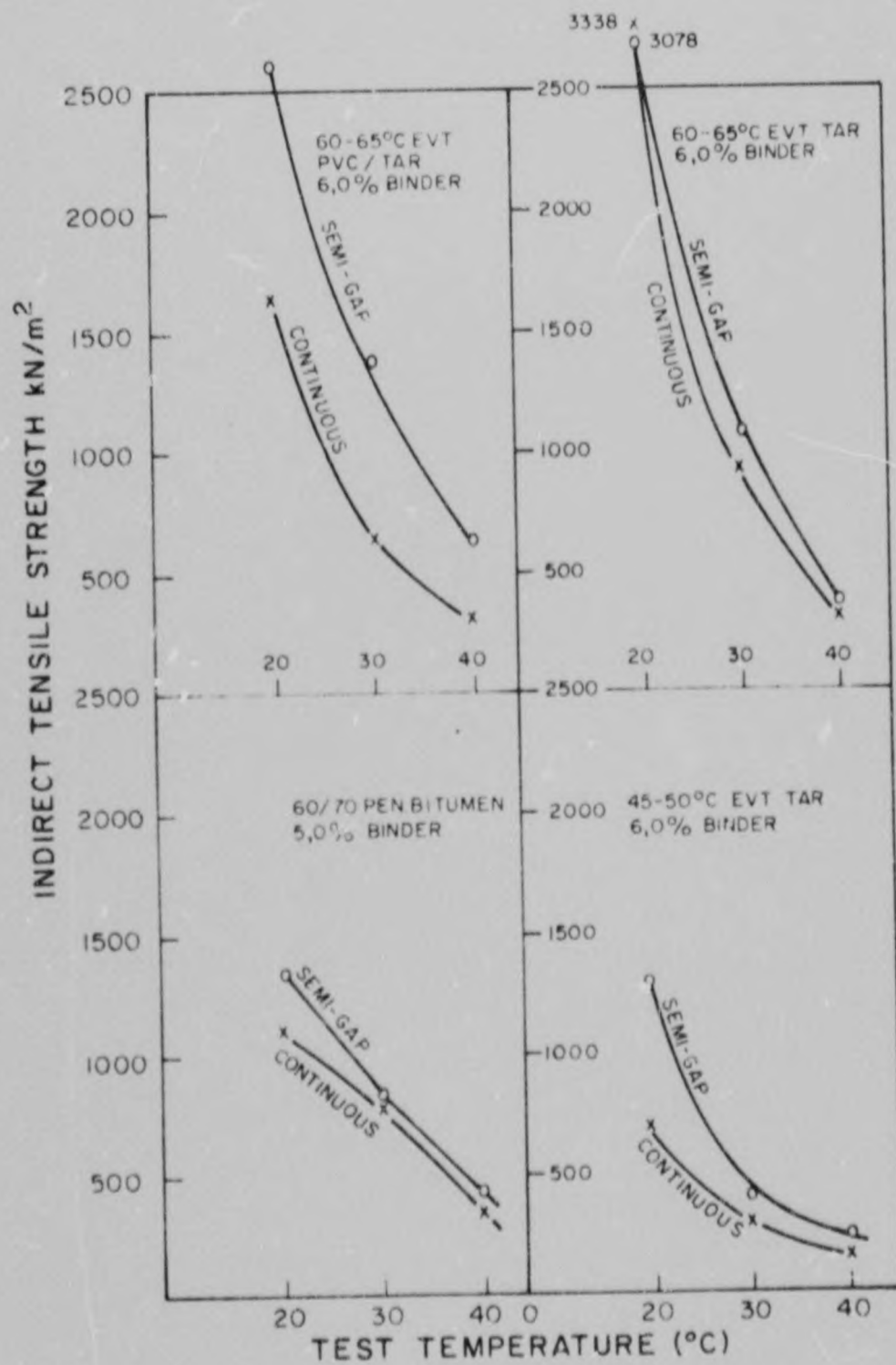


FIGURE 4-9
THE INDIRECT TENSILE STRENGTH OF SEMI-GAP
AND CONTINUOUSLY GRADED MIXES PREPARED
WITH VARIOUS TAR BINDERS AND BITUMEN MIXES
TESTED OVER THE RANGE 20-40°C

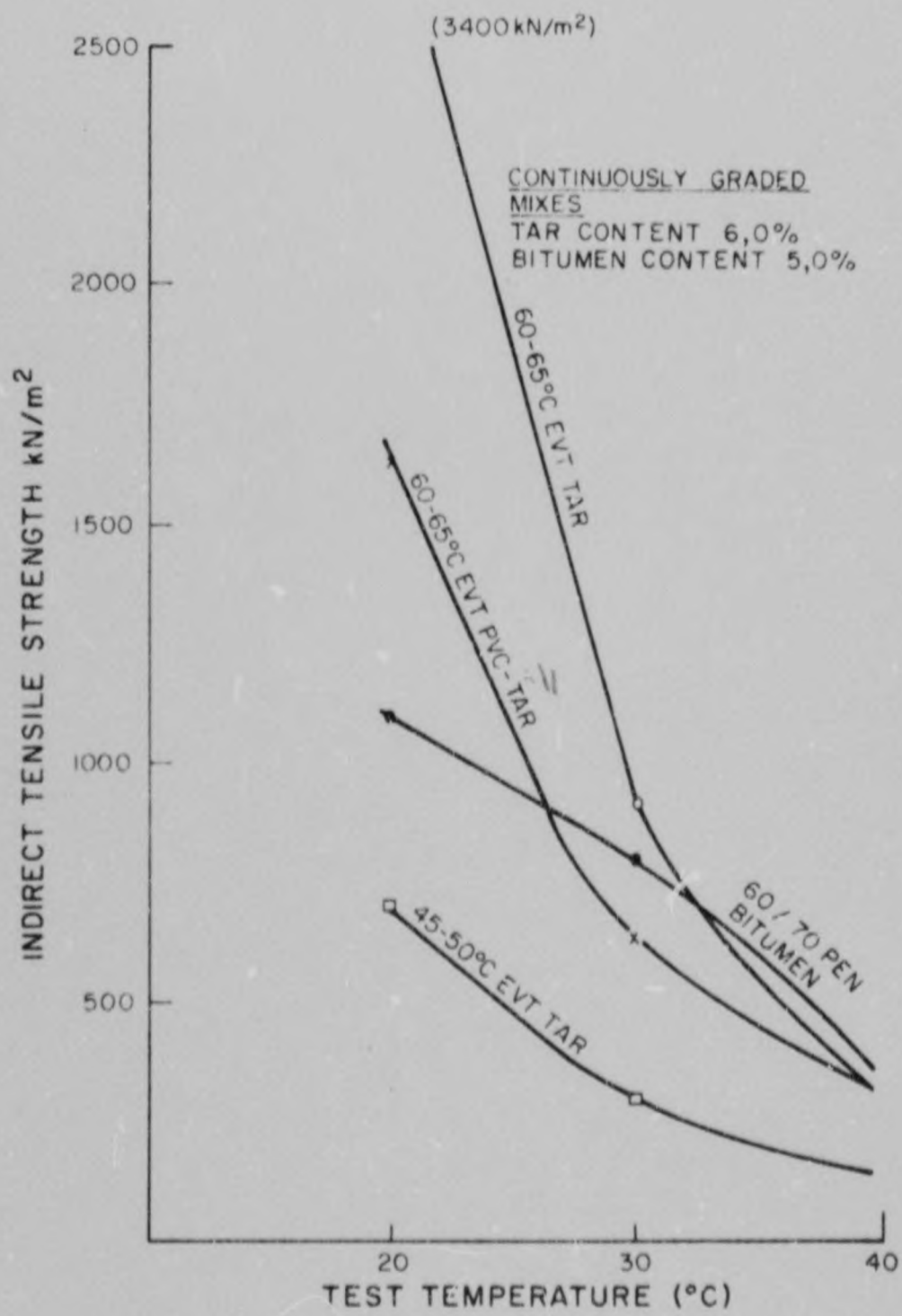


FIGURE 4-10
 STRENGTH (INDIRECT TENSILE) VERSUS TEMPERATURE
 DIAGRAMS FOR CONTINUOUSLY GRADED MIXES PREPARED
 WITH VARIOUS TAR BINDERS AND BITUMEN.

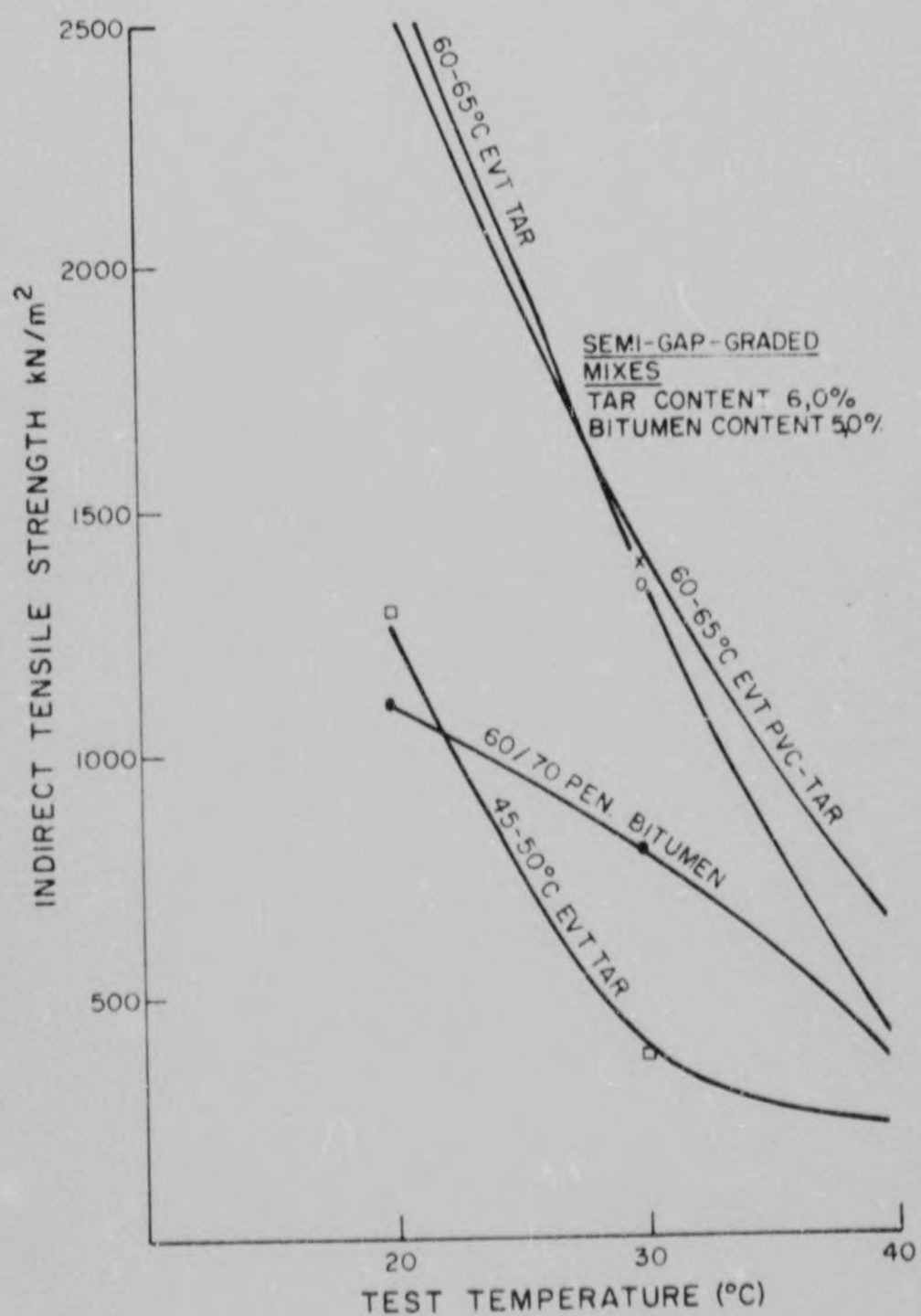


FIGURE 4-II

STRENGTH (INDIRECT TENSILE) VERSUS TEMPERATURE
DIAGRAMS FOR THE SEMI-GAP GRADED MIXES PRE-
PARED WITH VARIOUS TAR BINDERS AND BITUMEN

4.1.5 Elastic modulus of tar mixes

It is generally accepted that the flexing of the surface layer of a pavement less than 50 mm thick under traffic approximates to strain-controlled repeated-load conditions, (Monismith, 1966). Hence mixes with high strain tolerances, i.e. flexible mixes, are required for surface layers and the tensile strength will be a factor of secondary importance. It is suggested that the poor performance of tar-based mixes in surface layers (as compared to bitumen-bound mixes) is partly due to the fact that, as demonstrated by Daines (1972), for similar viscosities, at 25°C, tar has a higher stiffness than bitumen at rates of loading equivalent to fast traffic, and partly to the fact that the stiffness may be greatly increased in service because of the susceptibility of tar to weathering.

Measurements have been made of the elastic moduli of the mixes whose strength properties were described in section 4.1.4. These may be equated to stiffness measurements at short loading times and low temperatures (Monismith, 1969) and hence used to compare the stiffnesses of the mixes and to assess the suitability of the mix types for use in wearing courses.

The moduli measurements were determined on Marshall briquette specimens by an ultrasonic method*, described by Freeme (1974b), at a frequency of 150 kc/s. The measurements were taken in the direction of compaction of the specimen only (the results are influenced by the anisotropy of the material) and the temperatures used were 10, 20, 30 and 40°C as shown in Appendix M (Tables IV and V).

Mixes of the semi-gap-graded type generally had lower stiffnesses than continuously graded mixes. Values given by mixes containing equivalent volumes of binder (5 per cent, by mass, bitumen mixes and 6 per cent tar and PVC/tar mixes are shown in Figure 4 - 12.). This finding is also analagous to the results of studies of the fatigue lives of bitumen-

* The velocities of sonic waves are related to the elastic moduli of the media by: $E = \rho C^2$ (where E = elastic modulus, C = wave velocity, and ρ = density).

The specimen is connected to a receiver situated directly opposite each other on either side of the specimen. An ultrasonic pulse is propagated through the material and simultaneously the time base of an oscilloscope is triggered. By observing, on the screen, the transmitted and received signal in relation to the zero of the time base the travel time of the waves is determined. The velocity is calculated from the travel time and thickness of the specimen.

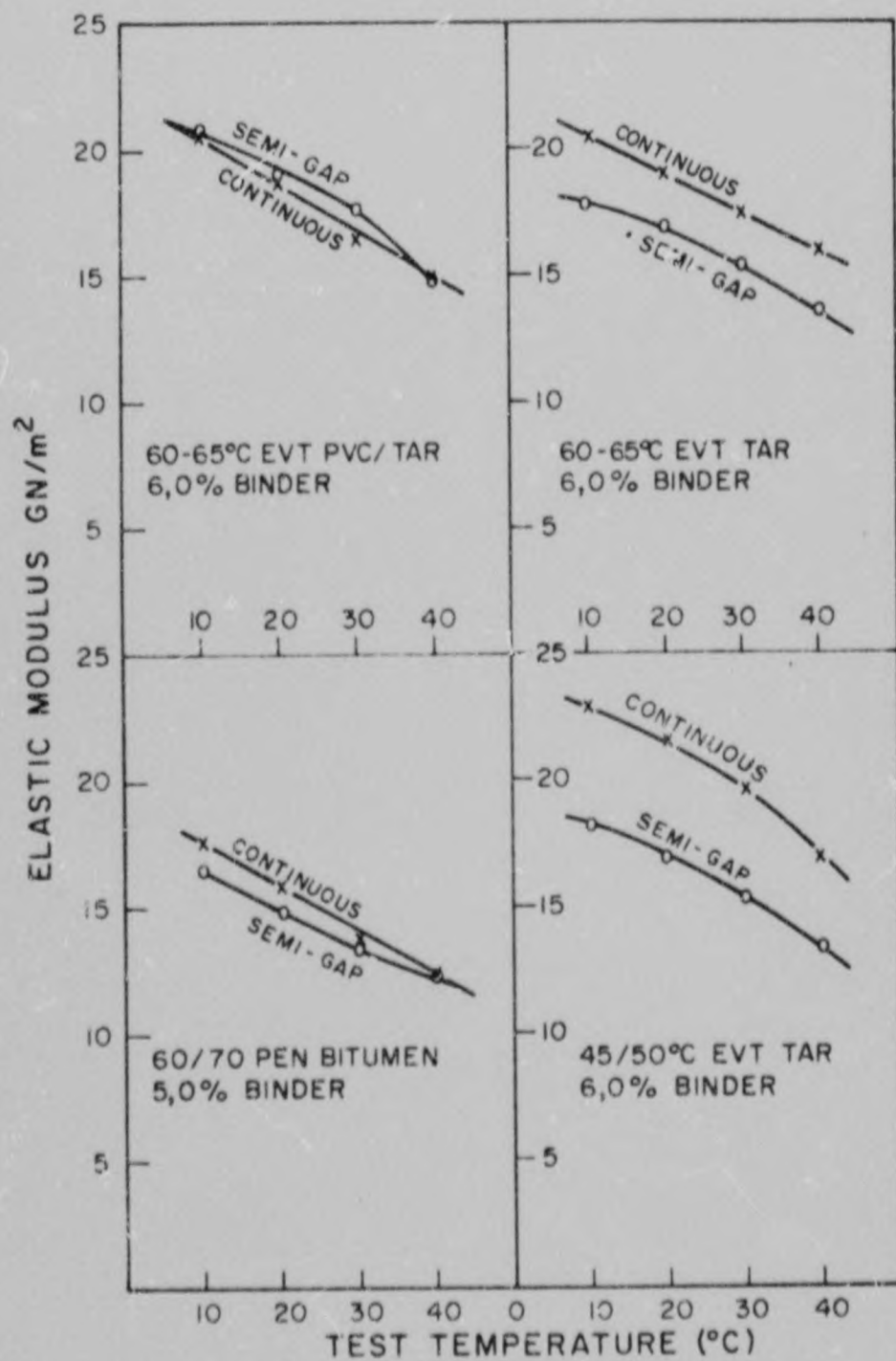


FIGURE 4-12

THE ELASTIC MODULUS OF SEMI-GAP AND CONTINUOUSLY GRADED MIXES PREPARED WITH VARIOUS TAR BINDERS AND BITUMEN MIXES TESTED OVER THE RANGE 10-40°C

bound mixes which showed that under constant strain conditions gap-graded mixes had longer fatigue lives than continuously graded mixes (Freeme, 1971).

The use of the semi-gap-aggregate gradings in tar wearing course mixes may be advantageous in partially offsetting the relatively high stiffnesses of tar mixes. The effect of binder type on the results given by the continuously graded mixes is shown in Figure 4 - 13 and on the results given by the semi-gap-graded mixes is shown in Figure 4 - 14. The bitumen-bound mixes have the lowest moduli in both mix types which confirms that they are likely to have superior fatigue lives in wearing courses. This is in accordance with the results given by Pell and Brown (1972) (Figure 4 - 2).

The stiffness of tar mixes may be reduced by changing the mix constituents. The author found that a continuously graded aggregate with a small maximum stone size (13,2 mm) had stiffnesses^{*} over the temperature range -10°C to +40°C which were virtually independent of the type of binder, as shown in Figure 4 - 15 . (These mixes were used in an experiment at Boksburg (see section 4.1.6).) It would be expected that mixes containing low-viscosity tars would have lower stiffnesses than mixes made with high-viscosity tars, though this is not confirmed by the data in Figures 4 - 13 and 4 - 14. The addition of PVC does not appear to have any effect upon the modulus. Freeme (1974a) has suggested that further reduction in stiffness may be gained by using mixes which are slightly binder-rich.

4.1.6 Weathering properties; the Boksburg experiment

The conclusions drawn from the determinations of the indirect tensile strength and elastic modulus apply only to freshly prepared mixes as these properties will be affected where significant weathering of the binder has occurred.

If weathering can be prevented, as is usually the case in basecourses, the initial properties may be maintained throughout the life of the pavement. However, in surface courses properties will alter if the binder hardens in service and in particular, the stiffness will increase, thus lowering the tensile strain tolerance and the fatigue life. Tar binders are more susceptible to weathering than bitumen and for this reason tar surface mixes should only be used on pavements where deflections and surface strains are low.

It has been mentioned that in South Africa tar surface courses

* Determined on an apparatus described by Freeme and Marais, (1974).

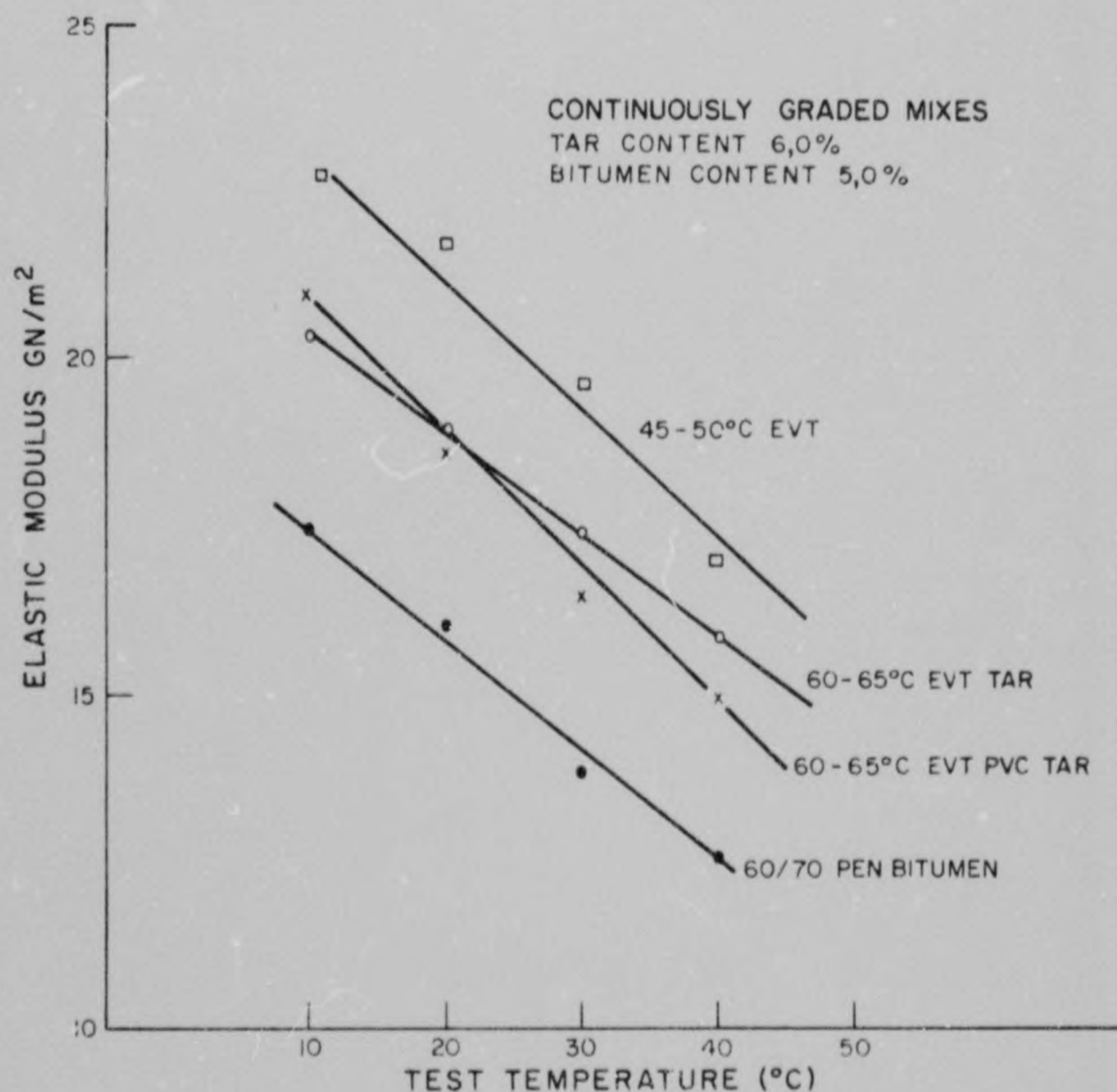


FIGURE 4-13
ELASTIC MODULUS VERSUS TEMPERATURE DIAGRAMS FOR
CONTINUOUSLY GRADED MIXES PREPARED WITH VARIOUS TAR
BINDERS AND BITUMEN

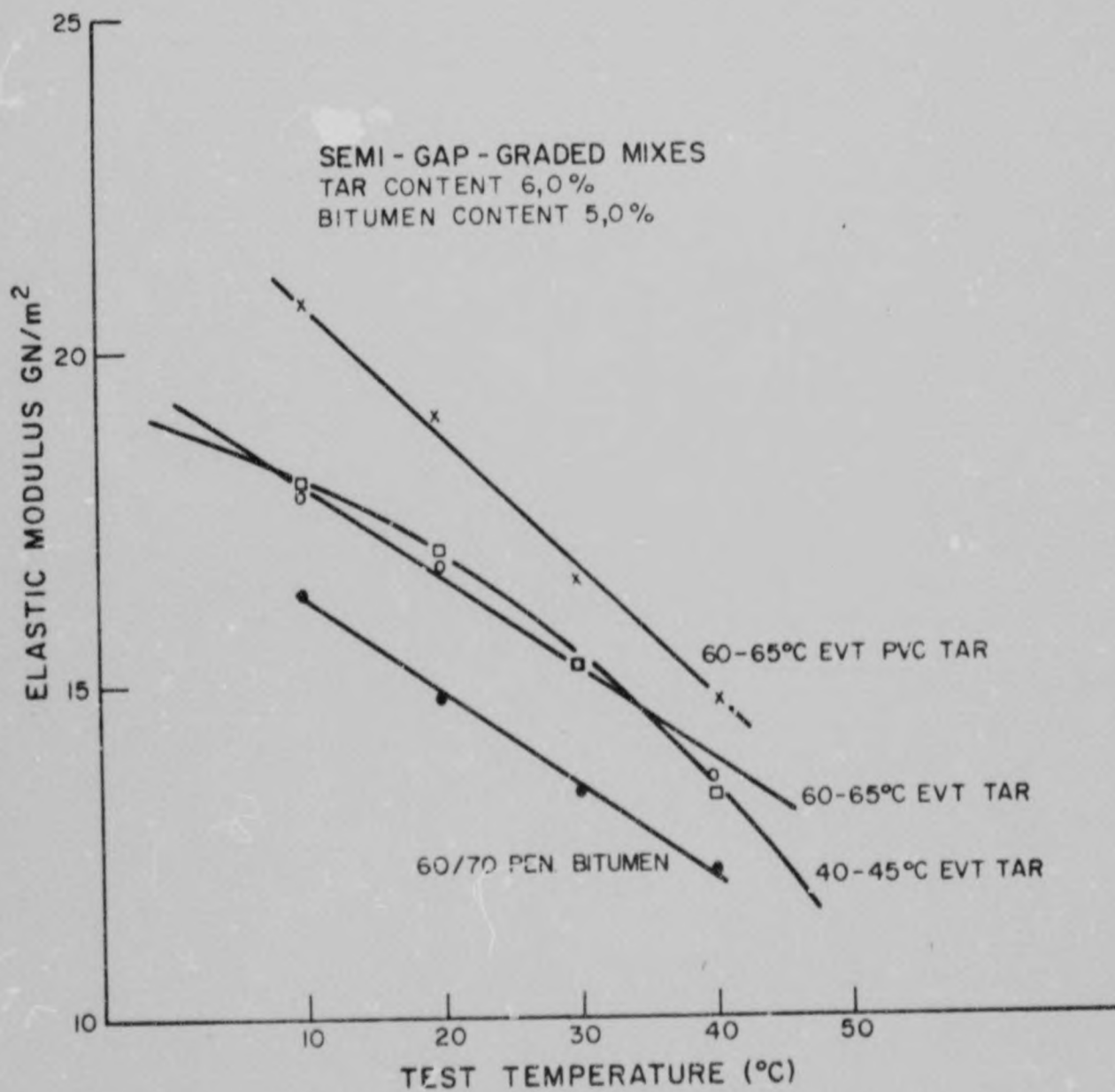


FIGURE 4-14

ELASTIC MODULUS VERSUS TEMPERATURE DIAGRAMS FOR
SEMI-GAP-GRADED MIXES PREPARED WITH VARIOUS TAR
BINDERS AND BITUMEN

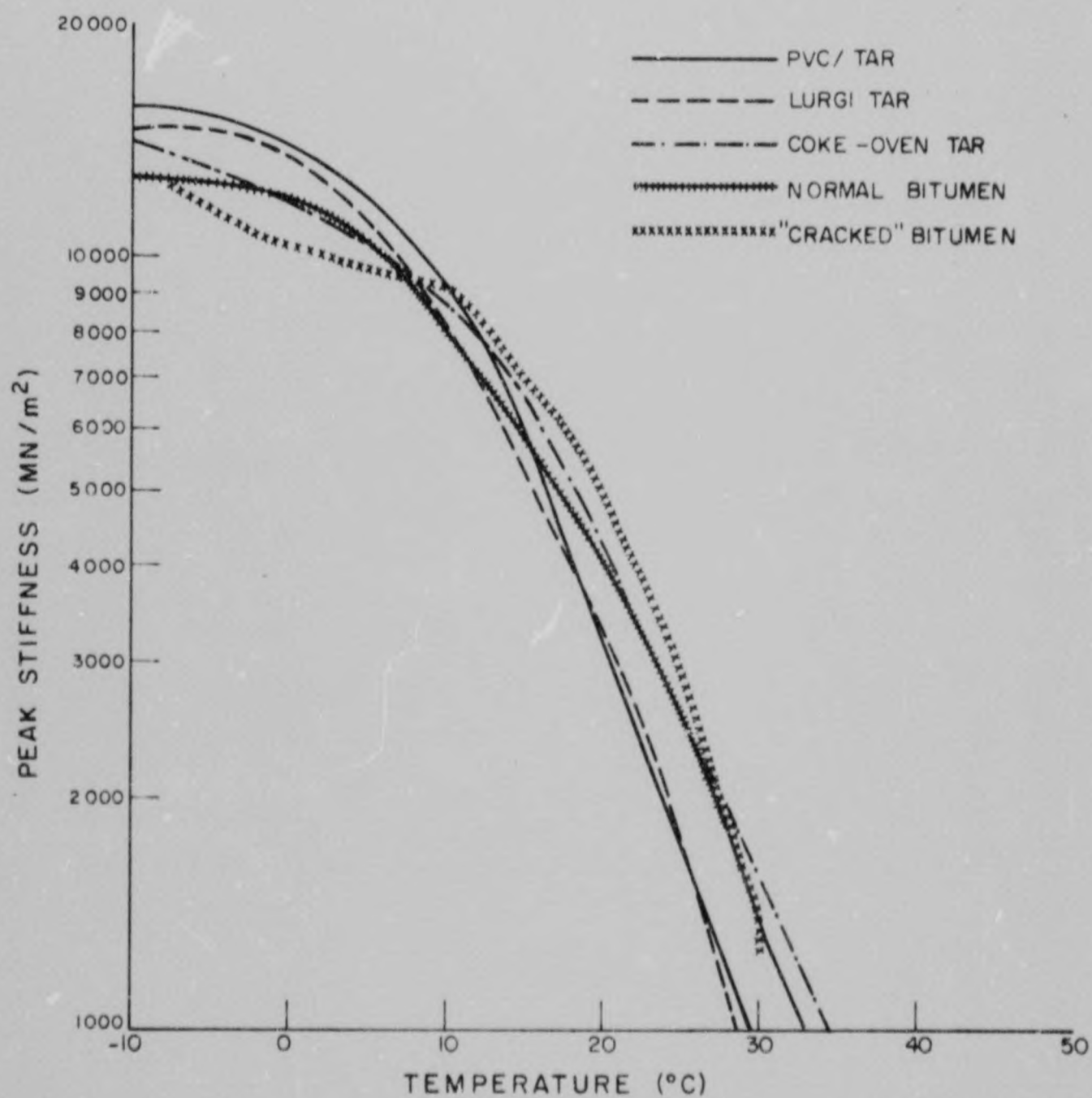


FIGURE 4-15
 VARIATION IN PEAK STIFFNESSES OF BOKSBURG
 MIXTURES (RECOVERED ONE MONTH AFTER LAYING)
 OVER THE TEMPERATURE RANGE -10 TO +40°C

have performed badly. However, in 1968 the Natal Provincial Administration constructed several experimental sections of tar wearing courses on route 12 near Umbumbulu. Some sections where a dense semi-gap-graded tar-mix were used are still in an excellent condition and this indicates that the target "break-even" life of a tar-bound surface mix (12 years) may be achieved if all the conditions are optimised.

To investigate the weathering of tar in a surface course full-scale experimental sections were constructed, at the author's instigation, on Yaldwyn Road, Boksburg, in May-June 1974.

4.1.6.1 Construction of the experimental surfacings

The main object of the experiment was to investigate the durability of various tar and bitumen binders in one aggregate grading type: a continuously graded mix having a maximum stone size of 13,2 mm, and to monitor properties of the mix during service.

Five binders were used: a 55°C Evt PVC-modified coke-oven tar, a 46°C Evt unmodified coke-oven tar, a 55°C Evt Lurgi tar, a 150/200 Pen. bitumen, and a 150/200 Pen. "cracked" bitumen*. For each binder type, mixes containing three binder contents were prepared and these were intended to be such that the voids contents of the mix would approximate to 2 4 and 6 per cent respectively. Hence fifteen experimental sections were constructed (these were 100 m (length) x 8 m (width) approximately). and these are represented graphically in Figure 4 - 16. The surface course had an average thickness of 35 mm. (Full details of the mix type, binder properties, sampling procedures and measurements taken during and subsequent to the construction are given in Appendix K.).

It turned out that the mixes were generally short of filler, as shown in Figure 4 - 17, and were also low in stability. Despite the filler shortage tests on sample cores revealed that the voids contents were low. Cores extracted after one year had lower voids contents than corresponding cores extracted after one month which suggests that because of the low viscosity of the binders the voids had closed up under traffic. Measurements on cores recovered after two years in service revealed no further reduction in voids contents.

* This type of bitumen is a residue from a process in which heavy oils are thermally treated so that large hydrocarbon molecules break down to give small molecules that constitute gasoline fractions.

SECTION No	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	
BINDER TYPE	COKE-OVEN - TAR		LURGI TAR		PVC / TAR		BITUMEN		'CRACKED' BITUMEN							
ACTUAL BINDER CONTENT	5,9	6,6	6,4	6,1	6,4	5,8	5,7	5,8	5,3	5,4	6,2	6,1	5,2	5,9	6,4	

FIGURE 4-16

THE LAYOUT OF THE BOKSBURG EXPERIMENT

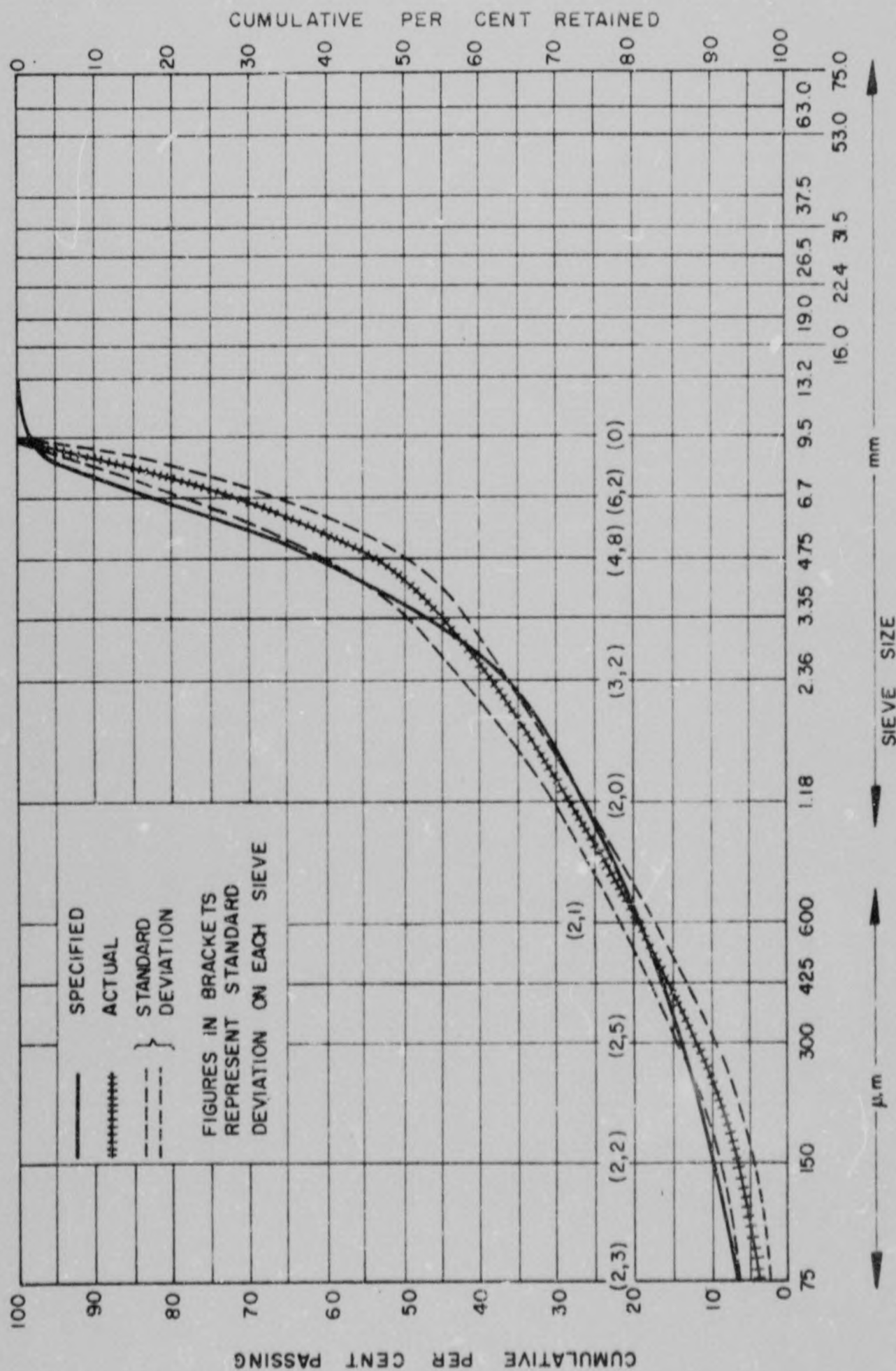


FIGURE 4-17
 MEAN OF ALL AGGREGATE GRADING ANALYSES, ALL MIXES USED IN THE BOKSBURG HOTMIX SURFACING EXPERIMENT.

These results which are shown in Table 4 - 3 are the means of values obtained on eight cores per section.

TABLE 4 - 3

VOIDS CONTENTS OF MIXES USED IN THE BOKSBURG EXPERIMENT
AFTER VARIOUS PERIODS OF SERVICE:
MIXES CONTAINING OPTIMUM BINDER CONTENTS

Section No.	Binder type	Voids content of mixes*		
		1 month	1 year	2 years
2	Coke-oven tar	9,5(0,9)	2,2(0,1)	4,6(0,9)
5	Lurgi tar	4,4(0,4)	4,8(0,6)	4,0(1,5)
8	PVC/tar	5,4(0,6)	3,0(0,2)	4,4(1,5)
11	Bitumen	3,3(0,1)	2,4(0,04)	2,7(0,7)
14	"Cracked" Bitumen	5,8(0,4)	1,4(0,2)	5,5(2,2)

* Figures in parentheses are the standard deviations of results on eight cores.

The coke-oven tar sections became very "fatty" a few months after construction. In these sections, where the binder viscosity was lower than that of the other binders, the tar appeared to have migrated to the surface. Six cores were taken from the surface of section 3 and each was cut into an upper, middle and lower layer with a diamond saw, and voids and binder content determinations were carried out on each layer. These results are shown in Table 4 - 4.

TABLE 4 - 4

RESULTS OF VOIDS AND BINDER CONTENT DETERMINATIONS ON
CORES TAKEN FROM SECTION 3 (UNMODIFIED COKE-OVEN TAR MIX)
BOKSBURG EXPERIMENT

Core No.	1	2	3	4	5	6
Upper layer Voids per cent	2,4	3,5	2,9	2,6	2,8	3,8
Binder content %	9,8	8,4	12,3	9,8	9,9	9,8
Middle layer Voids per cent	6,4	7,4	7,6	9,8	7,7	7,0
Binder content %	5,5	4,8	5,1	5,1	5,2	5,0
Lower layer Voids per cent	6,1	7,5	3,3	12,1	6,3	9,8
Binder content %	3,9	3,8	4,7	4,3	5,1	3,7

The upper layer of each core contained less voids and more binder than the middle and lower layers. Thus an upward migration of the binder under the action of traffic appears to have taken place.

4.1.6.2 Hardening of the binder and change in mix stiffness in service

The condition of the experimental sections has been assessed after four months, one year and two years in service according to the RRL system described in Part 3, Chapter 1. In addition the "fattiness" was assessed on a five point scale. As before the assessments were made by a panel of experienced road engineers and average assessments are given in Table 4 - 5.

The sections containing the optimum binder contents (sections 2, 5, 8, 11 and 15) were selected for a close study of change in mix properties and binder hardness monitoring tests. To date measurements have been taken after one month, one year and two years in service.

The binders were recovered from eight cores taken from each section using the hot centrifuge method. The Evt of the extracted binders was determined by the trough viscometer and a correction was made for the presence of mineral matter in the binder, as described in Appendix N.

In the case of all five binders the change in Evt over the first year of service was much smaller than would have been expected if these binders had been used in single-surface treatments (the change in Evt of the binders in single-surface treatments was estimated from the Accelerated Weathering Test results, Appendix K, Table XXI). This is shown in Figure 4 - 18.

After two years the hardening of the PVC/tar was similar to the hardening of the 150/200 Pen. bitumen. With the exception of the Lurgi tar the viscosity of the binders has not increased greatly over this period. In particular the unmodified and PVC-modified coke-oven tars have shown only a very small increase. After two years the Lurgi tar had hardened considerably and separated into two phases which made accurate viscosity measurements impossible.

After one month in service cores were taken from each of the fifteen sections and Evt measurements were carried out on the recovered binders. The results (Table 4 - 6) show that the Evt of each binder was unaffected by the variations in voids contents incorporated in the mixes.

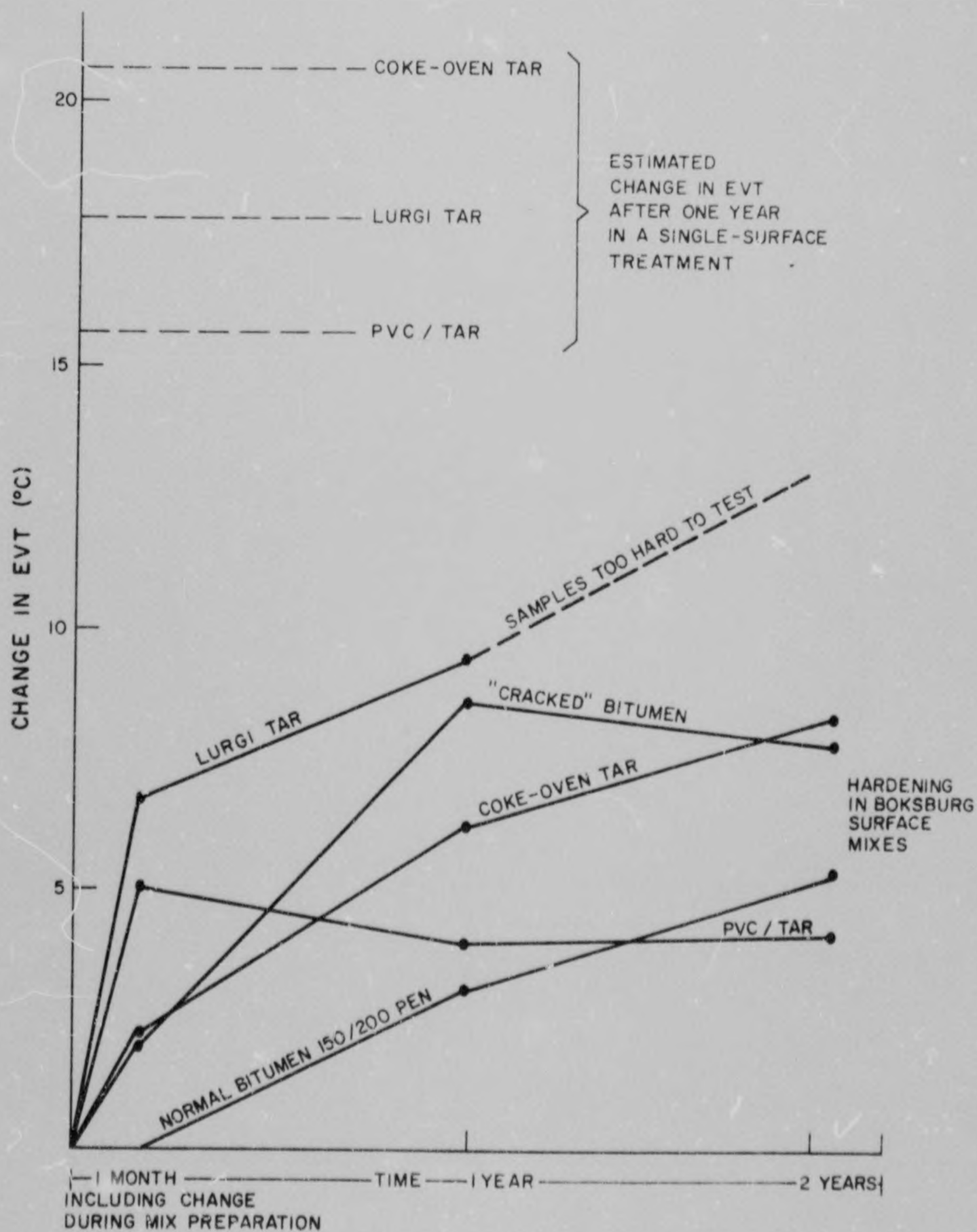


FIGURE 4-18

CHANGE IN EVT OF BINDERS RECOVERED FROM THE ROAD

TABLE 4 - 5
AVERAGE ASSESSMENT OF PERFORMANCE AFTER 4 MONTHS,
ONE YEAR AND TWO YEARS IN SERVICE

Sect. No.	Binder type	Binder content	Assessment rating**			
			4 months	1 year	2 years	
1	Coke-oven tar	5,9	G ₊	F ₊	EB FG ₊ (4)	WB G ₊ (3)
2	Coke-oven tar	6,6	P ₊ /F ₊	P ₊	G ₊ (5)	G ₊ (4)
3	Coke-oven tar	6,4	P ₊	P ₊	G ₊ (4)	G ₊ (4)
4	Lurgi tar	6,1	G ₋	G _{-v}	FG _v /G ₊ v(2)	G ₊ (1)
5	Lurgi tar	6,4	VG	VG	G ₊ (2)	G ₊ (1)
6	Lurgi tar	5,8	VG	VG	G ₊ (2)	G ₊ (1)
7	PVC/tar	5,7	VG	EB — G ₊ (1)	WB — VG	G ₊ (1) VG
8	PVC/tar	5,8	VG	FG ₊ (2)	VG	G ₊ (2) VG
9	PVC/tar	5,3	VG	G ₊ (1)	VG	G ₊ (1) VG
10	Bitumen	5,4	VG	G ₊ (1)	VG	G ₊ (1) VG
11	Bitumen	6,2	VG	FG ₊ (2)	G ₊ (1)	G ₊ (2) VG
12	Bitumen	6,1	VG	G ₊ (1)	G ₊ (1)	G ₊ (2) G ₊ (1)
13	"Cracked" Bitumen	5,2	VG	G ₊ (1)	VG	G ₊ (2) VG
14	"Cracked" Bitumen	5,9	VG	G ₊ (1)	VG	G ₊ (2) VG
15	"Cracked" Bitumen	6,4	VG	FG ₊ (3)	G ₊ (1)	G ₊ (3) G ₊ (1)

* For an explanation of the symbols see Part 3 Chapter 1

+ Figures in parentheses indicate the degree of fattening on a five point scale; (1) slight to (5) very bad

TABLE 4 - 6

EVT OF BINDERS RECOVERED FROM EACH SECTION OF THE
BOKSBURG EXPERIMENT AFTER ONE MONTH'S SERVICE

Section No.	Binder type	Actual binder content of mix %	No. of cores	Mean Evt of recovered binder
1	Coke-oven tar	5,9	2	52,9
2	" " "	6,6	8	50,8
3	" " "	6,4	2	50,2
4	Lurgi tar	6,1	2	67,0
5	" "	6,4	8	63,6
6	" "	5,8	2	64,5
7	PVC/tar	5,7	2	64,6
8	" "	5,8	8	62,9
9	" "	5,3	2	64,0
10	Bitumen	5,4	2	71,0
11	"	6,2	8	69,4
12	"	6,1	2	69,0
13	"Cracked" bitumen	5,2	2	62,3
14	" "	5,9	8	62,7
15	" "	6,4	2	62,0

The low increase in Evt of the tar binders in the first year of service may be ascribed partly to their low-volatile characteristics. However, the results of an investigation on other tar surfacings constructed in the same area (Field Street, Boksburg) suggest that the weathering of the binders may have been retarded by the impermeable nature (as indicated by the low voids content) of the mix. In contrast to the initial performance of the tar binders in the Boksburg surfacings experiment, the sections on Field Street, in which a Lurgi tar-bound mix was used, were severely ravelled and cracked after one year in service while adjacent sections

constructed with bitumen-bound and " cracked " bitumen-bound mixes are still in good condition. The structural condition of the pavement on Field Street was assessed by La Croix Deflectograph (Leger, 1969) measurements. Four cores were taken from each of the tar, bitumen and " cracked " bitumen sections on Field Street. The voids content of the cores was determined after which the binder was recovered and the Evt determined.

It was found that the average deflections on all three Field Street sections were similar but were higher than those on the Boksburg sections (1,4 mm and 0,4 mm respectively). The voids contents of the cores taken from the tar mix on Field Street were higher than those in the adjacent bitumen and " cracked " bitumen mixes. The results in Table 4 - 7 show that the tar mixes had voids ranging from 6,5 to 12,7 per cent (whereas the voids in the bitumen and " cracked " bitumen mixes ranged from 3,3 to 7,2 per cent. (The Lurgi tar-bound mix had a higher voids content than the mix used in the Boksburg experiment, shown in Table 4 - 3.)

TABLE 4 - 7

VOIDS IN CORES TAKEN FROM FIELD STREET, BOKSBURG

Binder type	Core No.	Percentage voids content	Percentage binder content
Bitumen	A1	5,5	4,9
Bitumen	A2	5,6	5,7
Bitumen	A3	5,8	5,0
Bitumen	A4	4,8	5,5
Lurgi Tar	B1	6,5	4,9
Lurgi Tar	B2	9,3	4,7
Lurgi Tar	B3	11,1	4,6
Lurgi Tar	B4	10,4	4,6
Lurgi Tar	B5	12,7	-
"Cracked" Bitumen	C1	3,3	5,9
"Cracked" Bitumen	C2	4,7	5,9
"Cracked" Bitumen	C3	5,8	5,3
"Cracked" Bitumen	C4	7,2	5,5

The Evt of the Lurgi tar recovered from the cores taken from Field street was much higher than that of the Lurgi tars recovered from the Boksburg sections after a similar period in service. The initial Evt was also greater (62,5°C as compared to 55,6°C) and the Evt had increased by a greater amount after a similar period in service as shown in Figure 4 - 19.

It is clear that the permeability of the mix, as assessed here by the voids contents measurements, has a considerable influence on the hardening of tar binders in surfacing mixes and hence on the service performance of these mixes. The results of the determinations of voids and change in Evt in service in the Boksburg experiment augurs well for the service performance of these mixes.

Stiffness measurements were made on specimens of the mix recovered from the pavement after one year and two years in service. In order to allow for the recovery of surfacing samples of sufficient thickness (40 mm minimum) for stiffness tests, the thickness of the base layer was reduced in certain areas. These sampling areas were situated on the sections where the mix with the optimum binder content was used so that comparisons could be made between changes in mix stiffness and binder viscosity. Stiffness measurements after one year and two years in service were determined using the stiffness tester developed at the National Institute for Transport and Road Research (Freeme and Marais, 1974). These results were determined at 20°C and at a rate of loading equivalent to a vehicle speed of 90 km/hour and are given in Table 4 - 8.

TABLE 4 - 8
STIFFNESS (20°C AND 90 km/h) OF MIXES USED IN THE
BOKSBURG EXPERIMENT AFTER ONE YEAR AND TWO YEAR'S SERVICE

Section No.	Binder type	Stiffness (GN/m ²)	
		1 year	2 year's
2	Coke-oven tar	1,34	1,22
5	Lurgi tar	3,09	5,05
8	PVC/tar	1,04	1,49
11	"Cracked" Bitumen	1,51	1,81
14	Bitumen	1,06	1,74

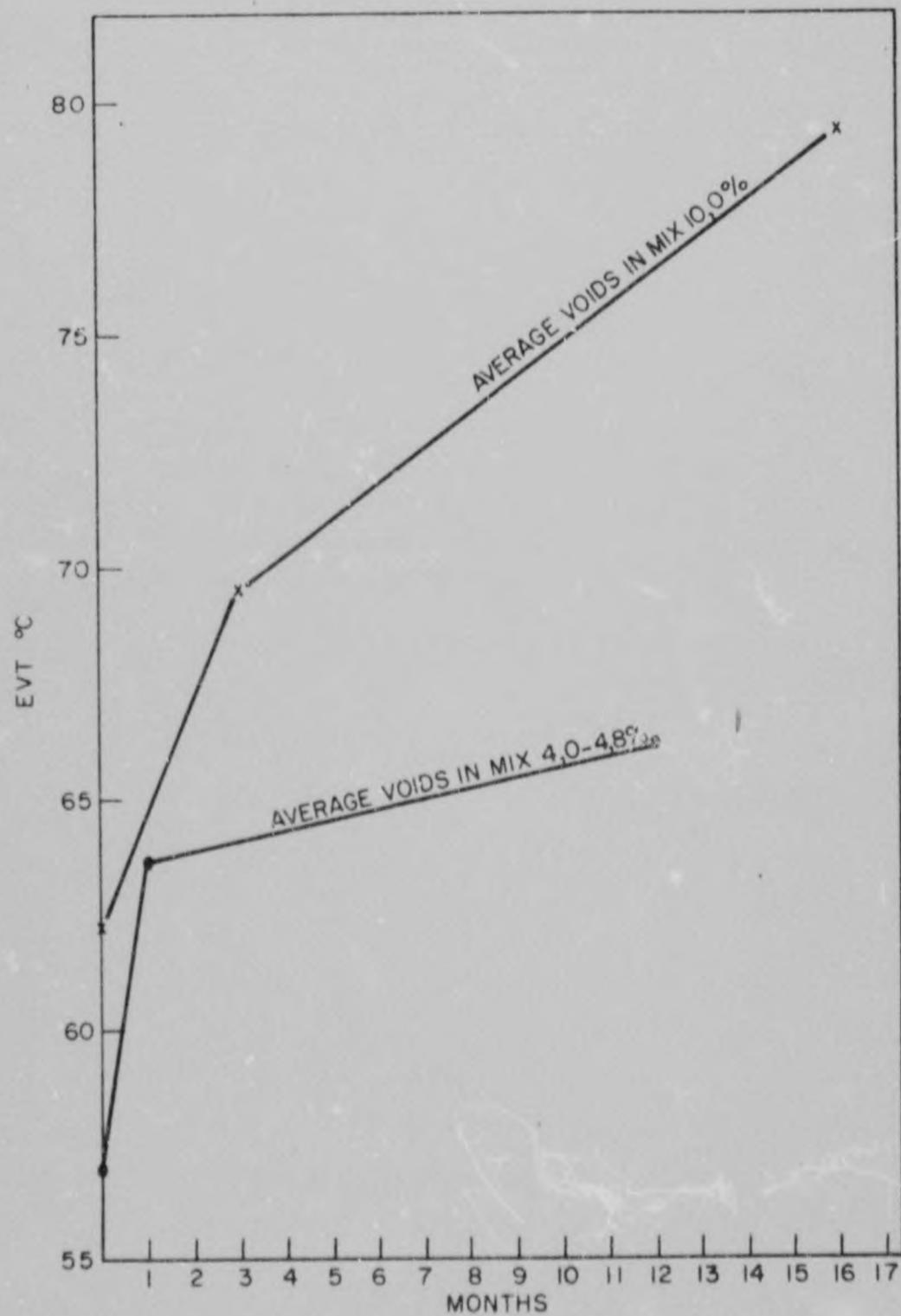


FIGURE 4-19
 EVT OF THE LURGI TARS USED IN THE FIELD S7
 AND BOKSBURG EXPERIMENTS

(The results of initial stiffness measurements on unweathered specimens have not been included due to doubts about their accuracy.) The stiffness of each mix type has increased over the one to two year period. However, after two years the coke-oven tar and PVC/tar bound mixes have stiffnesses similar to the bitumen. The stiffness of the Lurgi tar-bound mix is higher than that of the other mixes and this indicated that this mix may be less satisfactory in fatigue.

Deflection and radius of curvature measurements indicate that the horizontal strains in the pavement are low. Benkelman beam, radius of curvature and La Croix deflectograph measurements were taken shortly after construction and after various periods in service. Initial Benkelman beam and radius of curvature measurements are shown in Figure 4 - 20. These show that only on sections 3 and 7 were deflections greater than 500 μm recorded and in no case was the radius of curvature less than 100 metres. Deflectograph results obtained after four months and two years are shown in Figure 4 - 21. and excluding sections 2 and 3, in which measurements may have been erroneous due to the very low stability of the surface layer, only section 13 showed a deflection of more than 500 μm .

The most recent stiffness results show a moderate increase in stiffness over results obtained after one year. However, the present level of stiffness is low and this fact, together with the low horizontal strain levels given by the deflection and radii of curvature results suggests that the unmodified coke-oven tar and PVC modified coke-oven tar-bound mixes are unlikely to suffer fatigue failure at this stage. Although it is too soon to say whether the target life (12 years) will be achieved, this appears likely.

The effect of the weathering of the binder on the skid-resistance of the surfacings is a further factor that has been studied. The National Institute for Transport and Road Research recently acquired a machine for taking continuous readings of the sideways force coefficient^{*} of surfacings. This machine, the Sideways Force Coefficient Routine Investigation Machine (SCRIM) was developed at the Transport and Road Research Laboratory in the United Kingdom (Hosking and Woodford,

* The sideways force coefficient is the ratio of the force at right angles to the plane of a wheel, which is free to rotate but which has sideways as well as forward motion, to the load on the wheel

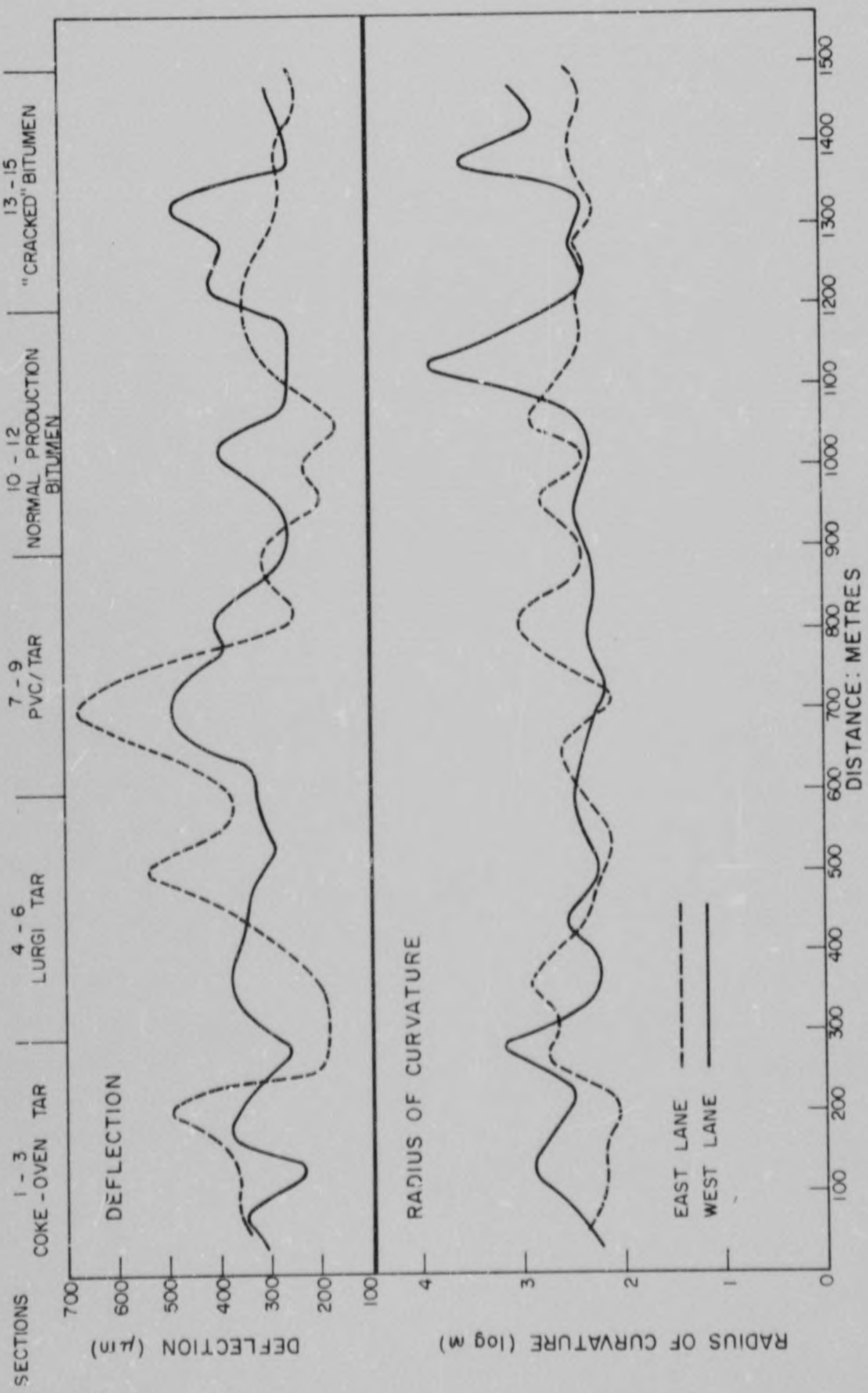


FIGURE 4-20
 RESULTS OF DEFLECTION (BENKELMAN BEAM) AND RADIUS OF CURVATURE MEASUREMENTS 3 MONTHS AFTER CONSTRUCTION.

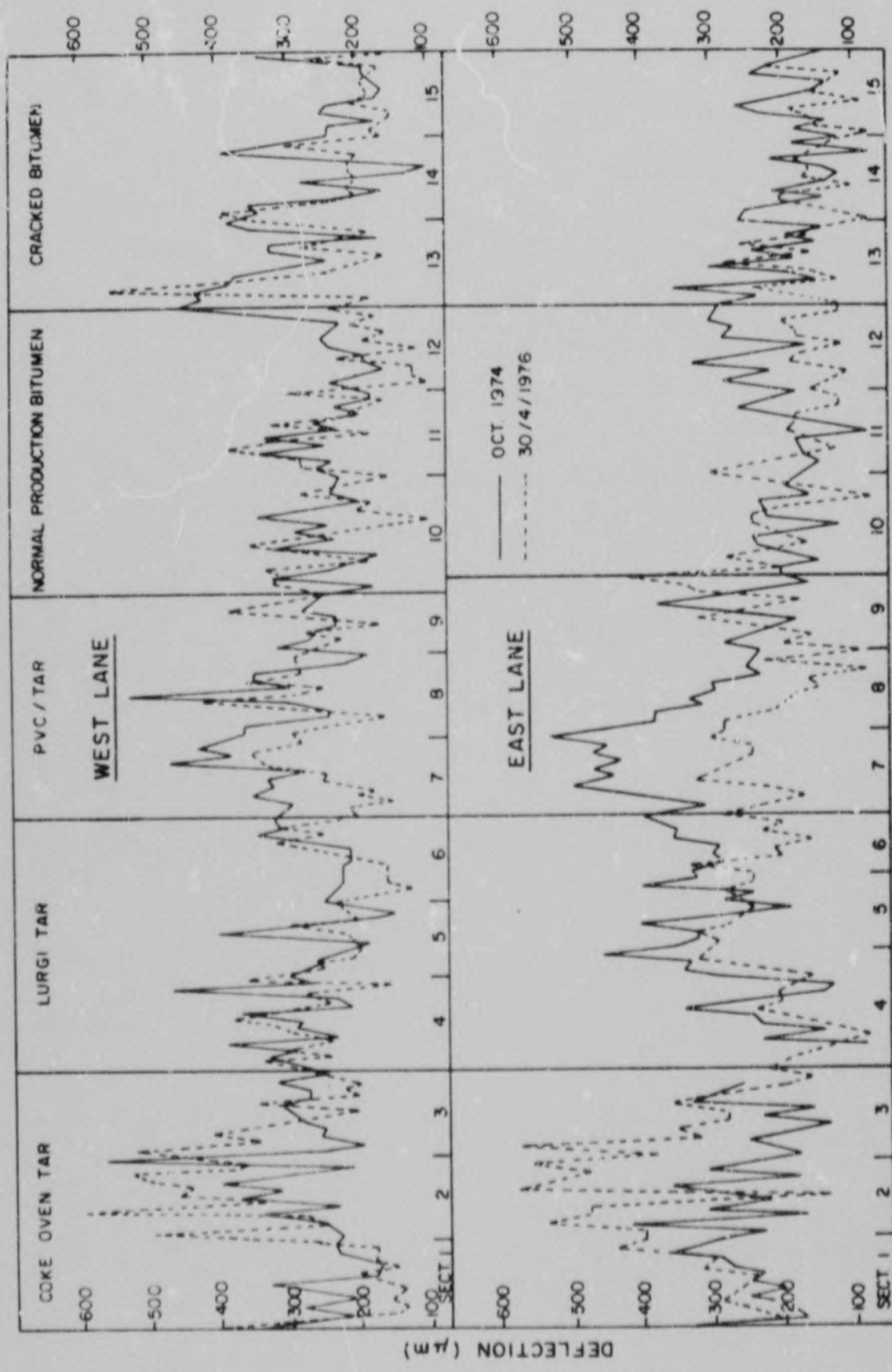


FIGURE 4-21
DEFLECTION MEASUREMENTS TAKEN BY LACROIX DEFLECTOGRAPH ON THE
BOKSBURG EXPERIMENTAL PAVEMENT AFTER 4 MONTHS AND 2 YEARS
IN SERVICE

1976 a,b,c,) and its application in South Africa has previously been described by Visser (1974).

The skid resistance of the experimental sections was measured at two speeds, 50 km/h and 80 km/h, shortly after construction and again after one year and two years in service. The change in skid resistance between one month and one year is shown in Figure 4 - 22, either as an improvement or deterioration in skid resistance. Due to the very "binder-rich" nature of the unmodified coke-oven tar sections, these showed a large drop in sideways force coefficient in the first year. They dropped to levels (0,10 - 0,25) well below minimum acceptable level (0,45). The sideways force coefficient of the surfacings after two years is shown in Figure 4 - 23. In Figure 4 - 22 and 4 - 23 there is a differentiation between values obtained on the eastbound and westbound traffic lanes, and axle mass analyser (Basson et al. 1976) measurements, which are given in Appendix K, showed that the number of equivalent 80 kN standard axles was greater on the former lane than on the latter by a factor of about 3.

Initial skid resistance measurements (given in Appendix K) were generally satisfactory, except for the eastbound lanes of the bitumen and "cracked" bitumen-bound mix sections which tended to be low. Figure 4-22 shows that after one year these sections and indeed all the sections had improved slightly. Of the tar-bound mixes the Lurgi tar mix had improved most and had the highest skid resistance of all. This binder had hardened most of all as discussed previously.

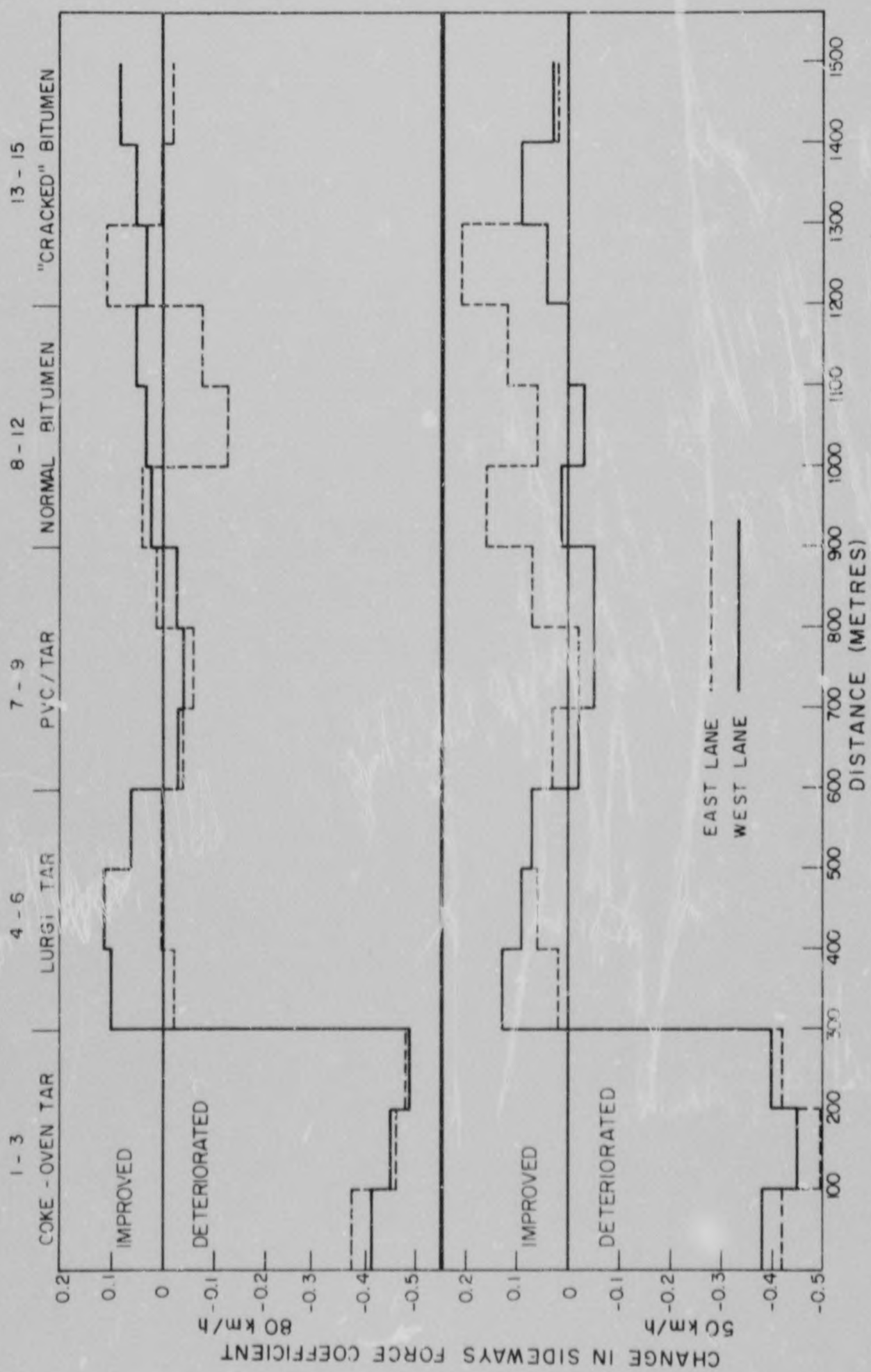


FIGURE 4-22

CHANGE IN SKID RESISTANCE (SIDEWAYS FORCE COEFFICIENT) OF THE SECTIONS 1 MONTH - 1 YEAR.

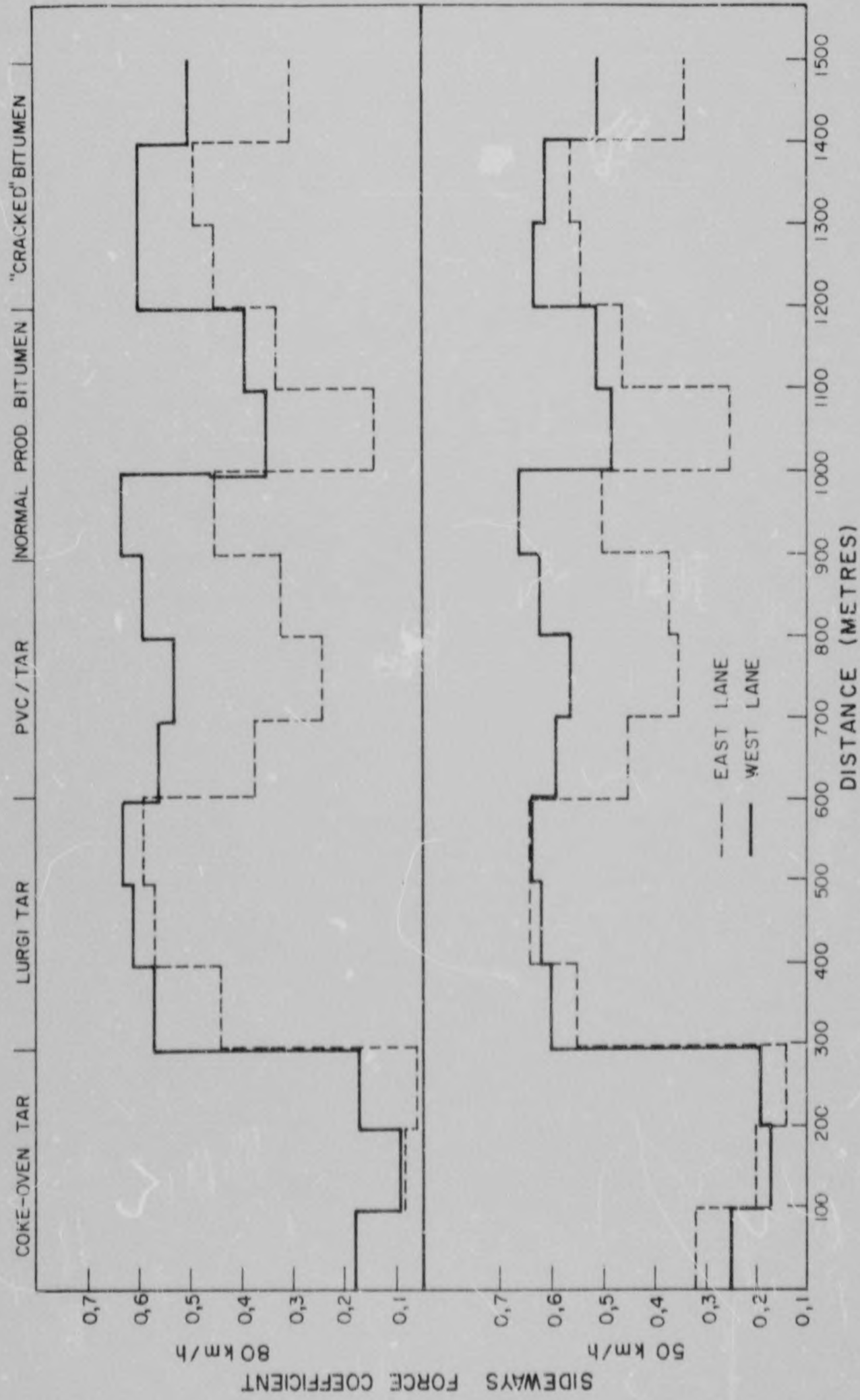


FIGURE 4-23
 THE SKID RESISTANCE (SIDEWAYS FORCE COEFFICIENT) OF THE BOKSBURG SECTIONS AFTER
 TWO YEARS IN SERVICE

4.1.7 Use of PVC in tar mixes

The use of tar-bound mixes is advantageous in areas where there is considerable spillage of petroleum fuel and oil and of hydraulic fluids. Bitumen-bound mixes may be severely affected by such spillage and, particularly where the pavement is required to take heavy static load, severe damage can occur as shown in Figure 4 - 24.

Tar mixes have been used for this purpose in many countries though in South Africa they have not yet been employed to any significant extent. Larcombe (1972) has described the construction of a pavement varying in thickness from 50 - 450 mm with such a tar mix at the White Bay container terminal in Australia. Marshall specimens of this mix showed no loss in stability after soaking in diesel oil for seven days. The distillation test results suggest that the tar would comply with the proposed specifications for low-volatile tar (see Appendix L). The performance of this pavement which is subjected to heavy wheel loads at high surface temperatures (70°C) after five years has recently been described by McGovern (1976). This author has reported no "rutting", cracking, softening, or signs of failure."

High-viscosity tars are required to give adequate stability in areas subjected to heavy loads and the use of additives is often employed to increase the viscosity of the tar. While studies with coal-modified tars in the USA showed that the durability of these binders were inadequate (Rhodes, 1962, Halstead et al. 1962 and Drake, 1962), considerable success has been achieved with polymer modified tars. A French company has established a special process involving a styrene-modified tar, and PVC-modified tars have been used for a number of such surfacings.

A 62 - 64°C Evt tar containing 1,5 per cent PVC was used in a mix covering a 250 000 m² area at the container terminal at Bremerhaven (Deters, 1971) and a 68°C Evt tar containing 4 per cent PVC was used in a surfacing mix laid on aircraft standing bays at Madrid airport, ("Road Tar", 1972). In Britain PVC/tar-bound surface mixes have been applied at RAF stations at Upper Heyford and Brize Norton using a low - viscosity tar (54°C Evt) containing 1,5 per cent PVC. (Holden and Williams, 1975).

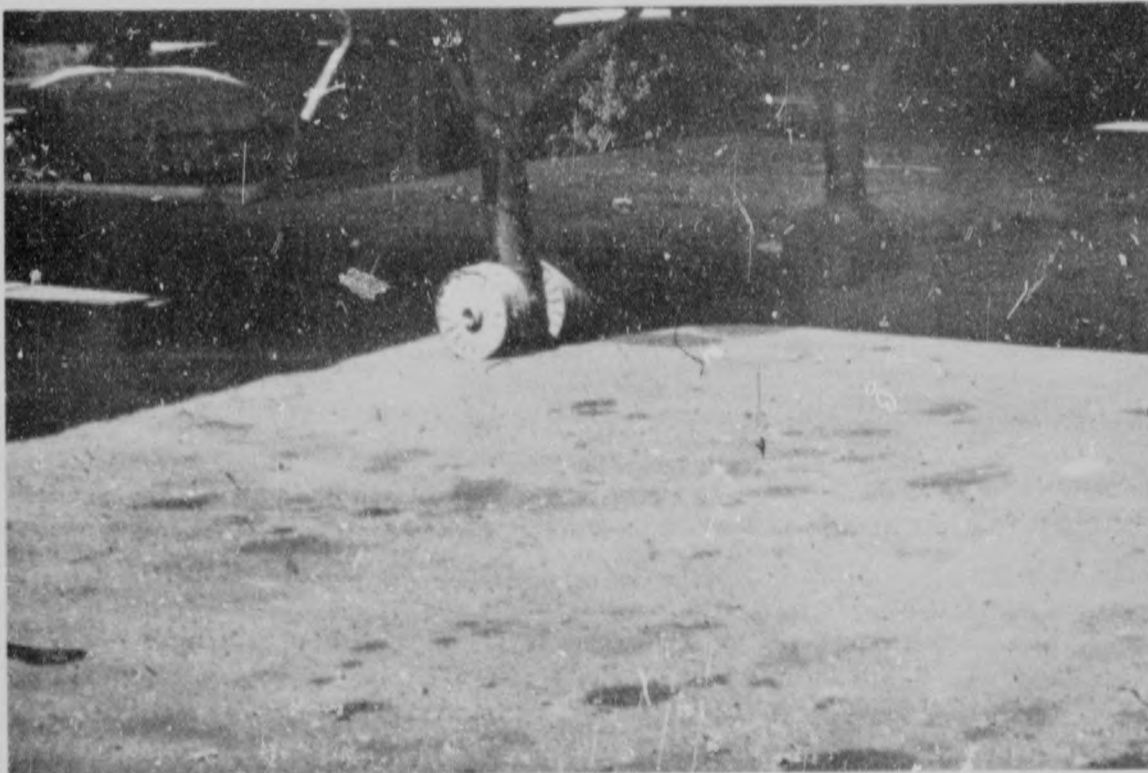


FIGURE 4 - 24

Effect of oil spillage and point loading on a bitumen macadam surfacing at the freight terminal at Le Bourget Airport in Paris. The point loading and oil spillage is shown in the upper photograph. The lower photograph is a close-up of the 'pitted' area. The 'pitting' occurred within a period of three years after construction.

From these reports it is uncertain whether the addition of PVC renders additional benefits to the mechanical properties of the mix apart from the increase in stability due to the increase in viscosity of the tar. Ansart, (1971) has referred to the "greater stability and resistance to fatigue of these mixtures". In section 4.1.4 it was shown that tar-bound mixes have a greater indirect tensile strength at moderate (20°C) temperatures than bitumen-bound mixes and this may be interpreted as a superior fatigue resistance under constant stress load repetitions, according to the Moore-Kennedy correlation. These measurements also showed that the change in strength with temperature of the continuously graded and semi-gap-graded mixes was less in the case of mixes made with PVC/tar than with mixes made with unmodified tar, Figures 4 - 10 and 4 - 11, and this is consistent with the lower temperature susceptibility of viscosity of the PVC/tars.

A study was carried out to determine whether the stability of the mixes incorporating PVC/tar binders is affected by the PVC content, the Evt of all the binders being the same, and whether the change in stability with temperature is affected by the PVC content. The PVC contents ranged from 0 - 5 per cent and three types of mix were used.

- (i) A continuously graded mix recommended for basecourses and conforming to clause 810 of the Ministry (British) of Transport Specifications for roads and bridges. 60-65°C Evt tars were used with this mix and an 80/100 Pen. bitumen was used for comparison purposes.
- (ii) A continuously graded surface course mix type with a maximum stone size of 13,2 mm. This is similar to the mix used in the Boksburg experiment and is known locally as a TPA* medium mix. 50-55°C Evt tars were used with this mix and an 80/100 Pen. bitumen was used for comparison purposes.
- (iii) A semi-gap-graded surface mix of the type recommended by Freeme (1974a) and shown in Figure 4 - 6. 50-55°C Evt tars and an 80/100 Pen. bitumen were the binders used with this mix.

In a preliminary investigation the optimum bitumen content of

* Transvaal Provincial Administration

each mix type was determined using the Marshall method. From this equivalent tar contents were selected so that a comparison could be made between mixes with similar voids contents. The binder contents for each mix type were as follows:

	Binder content %	
	Bitumen	Tar
Continuously graded mix	4,8	5,8
TPA medium mix	5,5	6,6
Semi-gap-graded mix	5,7	6,75;

The Marshall specimens were prepared in the normal manner, the tar-bound mixes (with both 50-55°C Evt and 60-65°C Evt tars) being prepared at 120°C and compacted at 105°C and the bitumen mixes being prepared at 140°C and compacted at 124°C. The voids contents of the Marshall specimens are shown in Table 4 - 9.

TABLE 4 - 9
VOIDS CONTENTS OF THE MARSHALL SPECIMENS USED
IN THE STABILITY VERSUS PVC CONTENT INVESTIGATION

Binder type	Test temperature (°C)				
	35	40	45	50	60
<u>Continuously graded basecourse mix</u>					
80/100 Pen. Bit.	2,3	2,3	2,3	2,3	3,1
Tar + 0% PVC	2,6	2,7	2,3	1,7	3,4
Tar + 0,75% PVC	-	-	-	-	2,2
Tar + 1,5% PVC	3,5	3,8	3,1	3,2	3,3
Tar + 2,25% PVC	-	-	-	-	2,7
Tar + 3,0% PVC	3,9	4,2	4,1	4,1	3,2
Tar + 4,0% PVC	2,5	2,9	2,8	3,0	3,3
Tar + 5,0% PVC	2,3	2,6	2,4	3,0	3,0
<u>Continuously graded TPA medium surface course mix</u>					
80/100 Pen. Bit.	-	-	-	-	-
Tar + 0% PVC	-	3,5	3,8	3,8	3,7
Tar + 1,5% PVC	-	3,7	4,3	4,4	4,8
Tar + 3,0% PVC	-	3,7	4,1	4,7	3,9
Tar + 4,0% PVC	-	2,9	3,0	2,8	2,9
Tar + 5,0% PVC	-	2,3	2,5	2,5	2,9
<u>Semi-gap-graded surface course mix</u>					
80/100 Pen. Bit.	-	2,3	2,4	3,5	3,3
Tar + 0% PVC	-	3,8	4,3	4,6	4,4
Tar + 0,75% PVC	-	-	-	-	4,1
Tar + 1,5% PVC	-	4,1	4,4	4,8	4,0
Tar + 2,25% PVC	-	-	-	-	3,6
Tar + 3,0% PVC	-	4,0	4,3	4,5	3,8
Tar + 4,0% PVC	-	2,4	2,5	2,6	3,1
Tar + 5,0% PVC	-	2,7	2,7	2,9	2,7

The stability of each mix was measured at 35, 40, 45, 50 and 60°C and these results are given in full in Appendix M. The effect of PVC content on the stability of each type of mix is shown in Figure 4 - 25. Provided the viscosity of the binder is unaltered the PVC has no influence on the stability of the mixes. This was also true when the stabilities of the various mixes were compared at 35, 40, 45 and 50°C.

From the results it was possible to investigate the effect of change in temperature on the stability of the various mixes. Linear regression analyses of the log stability/log temperature (°C) data were carried out for each of the three types of mix. Values of the slope intercept and regression coefficient are given in Table 4 - 10.

TABLE 4 - 10
LOG STABILITY - LOG TEMPERATURE (°C) REGRESSION DATA

	Bitumen	Tar				
		0% PVC	1,5% PVC	3,0% PVC	4,0% PVC	5,0% PVC
		<u>Continuously graded basecourse mix</u>				
Slope	-1,47	-1,92	-2,10	-2,28	-1,86	-1,59
Intercept.	3,51	4,38	4,70	4,93	4,32	3,77
Regression coeff.	0,96	0,96	0,97	0,94	0,98	0,97
		<u>Continuously graded TPA medium surface course mix</u>				
Slope	-1,10	-1,37	-1,72	-1,38	-1,65	-1,41
Intercept	2.85	3,28	3,92	3,37	3,79	3,37
Regression coeff.	0,96	0,99	0,00	0,92	0,00	0,99
		<u>Semi-gap-graded surface course mix</u>				
Slope	-1,58	-1,80	-2,42	-1,57	-2,72	-2,00
Intercept	3,69	3,98	5,09	3,66	5,64	4,42
Regression coeff.	0,96	0,91	0,99	0,97	0,99	0,99

The addition of PVC to the tar did not appear to have any effect upon the temperature susceptibility of stability of the mixes despite the fact that the polymer improved the temperature susceptibility of viscosity of the tar (Part 3, Table 3 - 3 and Jamieson, 1973a)

Although the use of high (up to 5 per cent) PVC contents has not

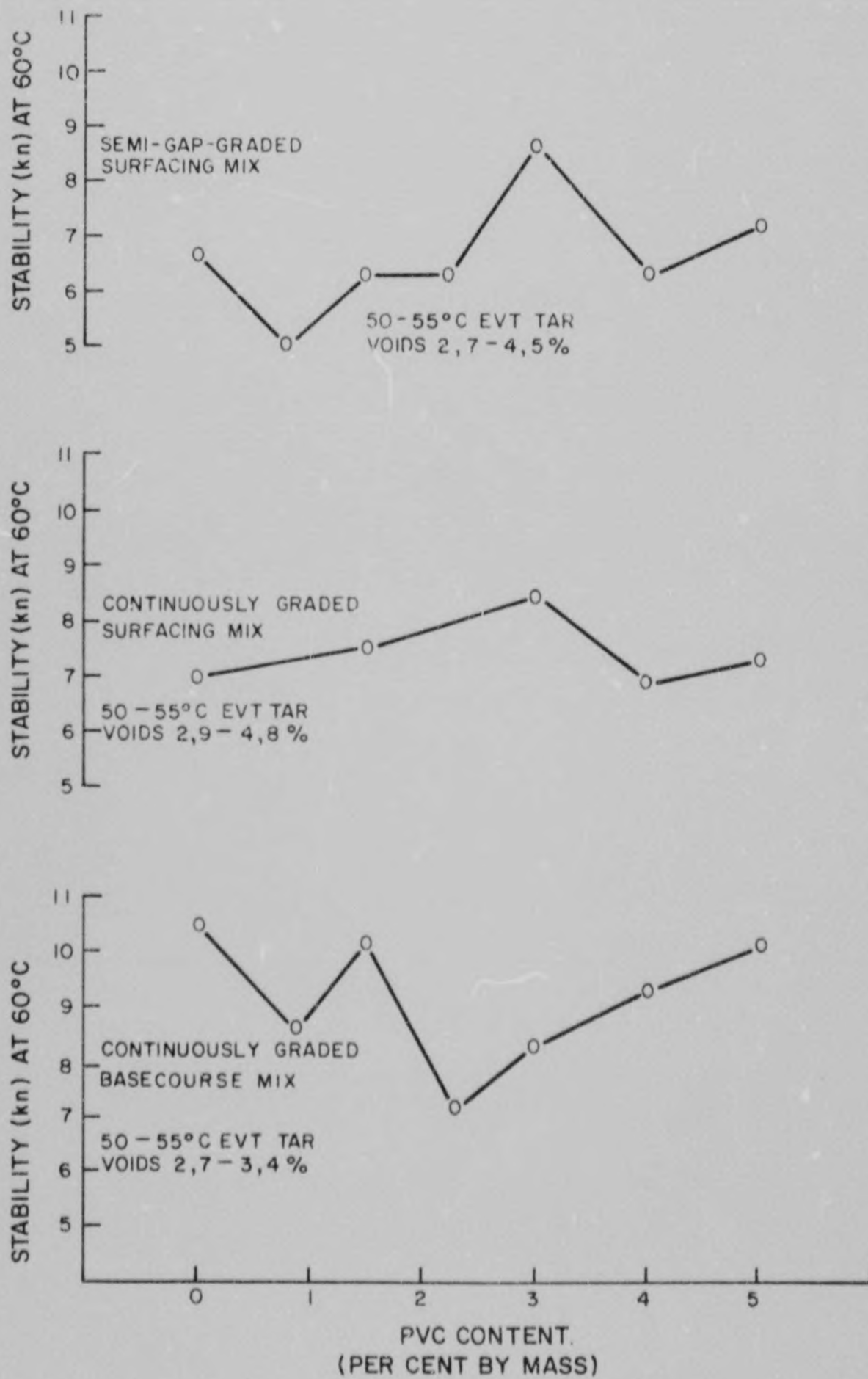


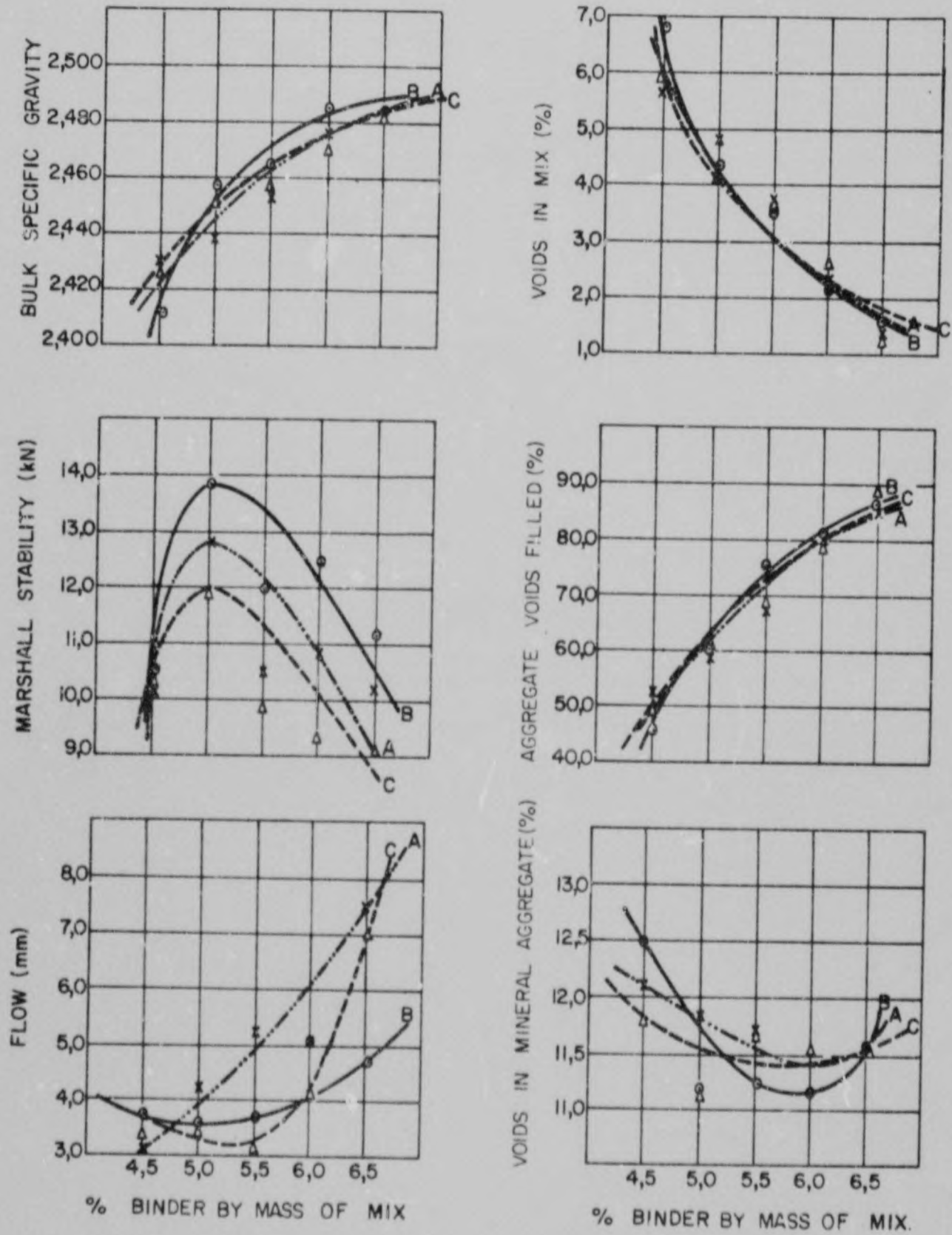
FIGURE 4-25
THE EFFECT OF PVC CONTENT OF THE BINDER ON THE
STABILITY AT 60°C OF THE THREE TYPES OF MIX.

improved the stability characteristics of mixes its inclusion does make it possible to improve the durability of the tar. In the preparation of these blends a very heavy oil, described as having more than 90 per cent of constituents boiling above 340°C was used. The more PVC contained in the binder the more of this oil was required and as described in Part 3, (Chapter 1.4.3) this would increase the proportion of plasticising-oil to volatile-oil in the blend and would enhance the durability of the binder. Mixes with tars containing 5 per cent PVC would be unsuitable for normal road surfacings, apart from their high cost, as the binder would tend to be brittle at rapid rates of loading (see ductility-tenacity results Part 3, Figure 3 - 8). The composition of these blends is shown in Appendix M, Table 1X.

It has been suggested by workers in Europe (Jamieson, 1973b) that it may be possible to prepare PVC/tar-bound mixes by incorporating the PVC powder in the fine aggregate proportion instead of first preparing the binder in the conventional manner. This would have certain advantages in avoiding the problems occasionally resulting from storage of the binder for prolonged periods at high temperatures. An investigation was carried out to examine the properties of mixes prepared in this manner.

Two series of mixes (each series consisting of binder contents varying from 4,5 to 6,5 per cent) were prepared, one in which the 60-65°C Evt tar contained 1,5 per cent PVC and one in which the calculated amount of PVC powder was added to the fine aggregate fraction and a lower Evt (55°C) unmodified tar was used to allow for the normal increase in Evt which results from addition of the polymer to the tar. It was thought that in the three minute mixing period the digestion of the PVC in the tar might be incomplete which would effectively increase the filler content with possible effect on mix properties. Hence a third series of mixes was prepared in which the amount of fine aggregate was reduced by the quantity of PVC powder added, assuming that none of the PVC powder was fully digested.

Marshall test specimens were prepared from each mix and the normal type of Marshall mix design tests were performed. These results are shown in Figure 4 - 26. Although the stability of all the mixes was adequate a number of factors indicate that in the mixes where the PVC powder was incorporated in the fine aggregate fraction it had not been adequately digested in the tar. The stability of the mixes in which the PVC was added via the fine aggregate was slightly below that of the mixes



LEGEND

- x PVC REPLACING PART OF AGGREGATE
- o PVC ADDED TO BINDER
- Δ PVC ADDED TO AGGREGATE

NO OF BLOWS: 2 x 75
 TYPE OF AGGREGATE: REEF QUARTZITE
 SOURCE AND GRADE OF BINDER: LB 326
 (No. 3 DUNDEE TAR)

BINDER CONTENTS

MAXIMUM UNIT WEIGHT: 6.5
 MAXIMUM STABILITY: 5.0
 4% VOIDS IN MIX: 5.0
 OPTIMUM: 5.5

FIGURE 4-26
 INFLUENCE OF BINDER CONTENT ON MARSHALL
 TEST VALUES

in which the PVC/tar was pre-prepared and the former mixes had high flow values at the highest binder contents.

The binders were extracted from the mixes and subjected to elastic recovery and PVC content determinations. These results are given in Table 4 - 11. The elastic recovery and PVC content data jointly may be considered as an indication of the effective solution of the PVC in the tar. It has been shown (Part 3, Chapter 1.1.

Table 3 - 5) that a coke-oven tar containing 1,5 per cent PVC would have an elastic recovery value in the range 170 to 240 per cent, though usually they are above 200 per cent (Part 3 Tables 3 - 9 and 3 - 11) The results given in Table 4 - 11 are at the low end of this range.

TABLE 4 - 11

EVT, ELASTIC RECOVERY AND PVC CONTENT OF PVC/TARS RECOVERED
FROM MIXES IN WHICH THE PVC WAS ADDED VIA THE
FINE AGGREGATE FRACTION

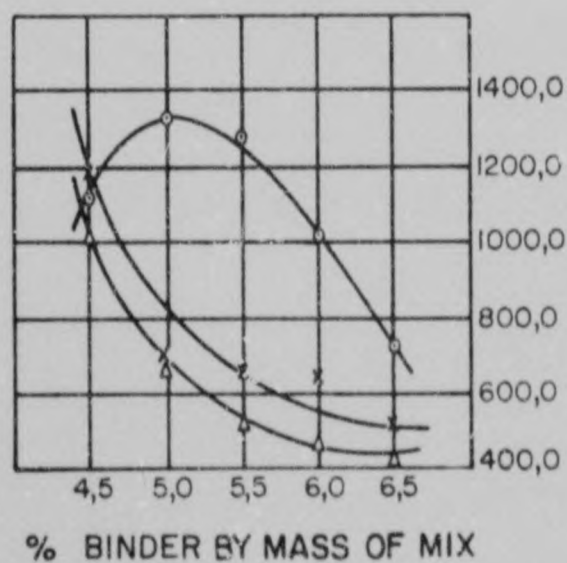
Sample Ref. No.	Mix type	Evt (°C)	Elastic Recovery % of film thickness	PVC % by mass of binder
92	PVC Powder	77,2	122	1,1
93	replacing	76,0	158	1,1
94	part of the	76,2	172	1,4
95	fine aggreg-	77,6	173	1,5
96	ate	75,6	148	1,4
97		76,5	136	1,3
98	PVC Powder	77,9	179	1,5
99	additional	75,2	155	1,4
100	to the fine	76,8	171	1,3
101	aggregate	78,1	175	1,6
102		81,1	158	1,5
103		78,2	175	1,2

While low elastic recovery figures could also be explained by degradation of the polymer, in this case such degradation is unlikely to have occurred. The highest temperature to which the binder was subjected was 120°C for three minutes and results (Part 3 Figure 3 - 13) showed that no decrease in elastic recovery occurred after storage of PVC/tar at 110°C for eight days.

The mixes in which the PVC powder was inadequately dispersed in the tar had unusual indirect tensile strength and elastic modulus characteristics. These properties were measured (at 40°C) according to procedures mentioned in Part 4, Chapters 1.4 and 1.5 respectively, and the results are shown in Figure 4 - 27. The mix in which the PVC/tar was pre-prepared had a maximum inflexion on the indirect tensile strength versus binder content curve at a binder content of 5.0 per cent, whereas the mixes in which the PVC was added via the aggregate had a completely different plot with decreasing strength as the binder content increased. The mixes in which the PVC was thought to be inadequately digested also showed an increase in elastic modulus with increasing binder content in contrast to the mix in which the PVC was fully digested in the tar.

This investigation has shown that in preparing PVC/tar-bound mixes the PVC/tar binder must be pre-prepared. The alternative of adding the polymer via the fine aggregate fraction does not permit thorough digestion of the PVC in the tar. This in turn means that not only are the full benefits of the PVC upon the mix properties not realised, but that such mixes may have high flow values, low stability and low tensile strength. These problems would be particularly disadvantageous where the mix is required to have high deformation resistance, i.e. in basecourses layers or in surfacings required to support heavy static loads.

INDIRECT TENSILE STRENGTH (kN/m²)



LEGEND

- x PVC REPLACING PART OF AGGREGATE
- o PVC ADDED TO BINDER
- Δ PVC ADDED TO AGGREGATE

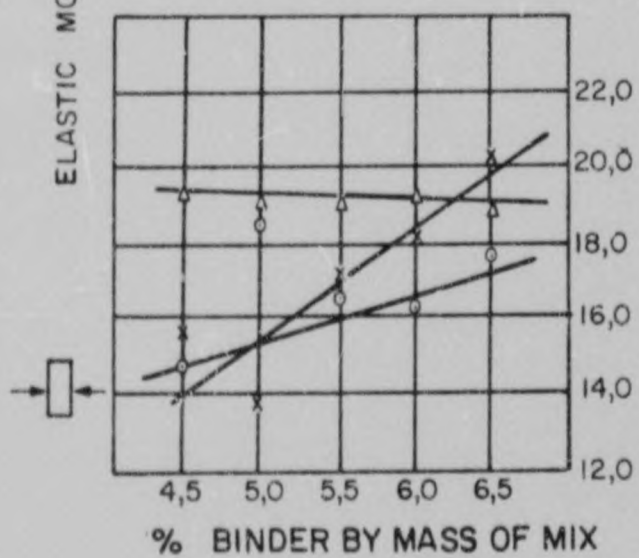
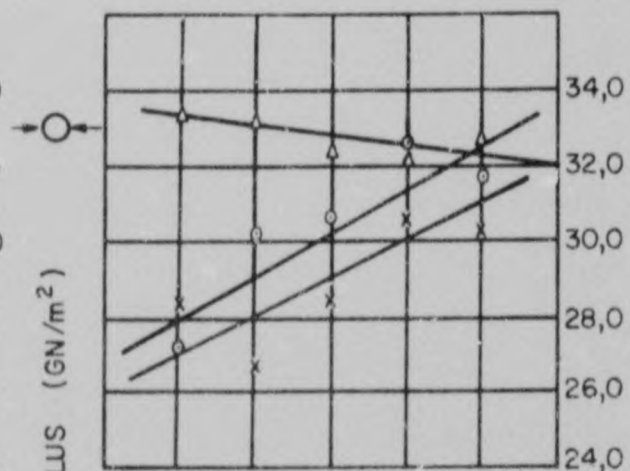


FIGURE 4-27

INFLUENCE OF BINDER CONTENT ON INDIRECT TENSILE STRENGTH AND ELASTIC MODULUS

4.2 EFFECT OF MIX PREPARATION AND HANDLING ON THE VISCOSITY
OF TARS

Resume'

Care must be taken to prevent overheating of tar mixes. The effect of film thickness, temperature and storage time at high temperature on the change in Evt of coke-oven tars has been studied. These investigations suggest that tar may be more susceptible to change in viscosity when used in semi-gap-graded mixes than in continuously graded mixes. Where the coating of large particles in semi-gap-graded mixes is insufficient it may be preferable to increase the binder temperature to slightly above the maximum permitted mix temperature, and other ways of improving coating e.g. the use of a lower viscosity grade of tar, or of a higher mix temperature, are undesirable.

A study of the hardening at various stages during the mix preparation and paving has shown that under normal operating conditions, changes in viscosity occur mainly during mixing and changes at subsequent stages are minimal. The change in viscosity of tar during mixing may be less than occurs in bitumens because of the higher mixing temperatures employed with the latter.

The change in viscosity of Lurgi tar during mix preparation exceeded that of coke-oven tar and bitumen binders.

Because of the nature of coal tars great care must be taken to avoid overheating them particularly while they are exposed in thin films on aggregate surfaces during mix preparation. Because of the infrequent use of tar mixes in South Africa, damage to the mix from overheating is common as operators are used to controlling mixing plants at the higher temperatures normally used with penetration grade bitumens. Overheating leads to a large increase in the viscosity of the tars which may affect the compaction of the mix and cause increased mix stiffness in service. Thermal degradation of the hydrocarbon constituents may take place in severe cases and the tar may lose its binding properties and the mix may be without adequate cohesive strength.

Coal tar hydrocarbons are lighter than those comprising bitumen (Part 2, Chapter 2) and they are more susceptible to thermal degradation.

It may be considered that apart from the composition of the tar itself, factors affecting the change in viscosity of tar during mix preparation are: binder film thickness, temperature and the time during which the mix is held at a high temperature in the uncompacted state. A number of experiments have been carried out in the laboratory and at full-scale mixing plants to investigate these factors.

4.2.1 Effect of film thickness

The effect of film thickness upon the change in viscosity during mixing was investigated by preparing a series of mixes of one aggregate grading and various binder contents. After the mix preparation the binders were recovered and changes in Evt were determined. The continuously graded basecourse aggregate grading was used for these investigations and the binder contents (60-65°C Evt coke-oven tar) were: 4,5, 5,0, 5,5, 6,0 and 6,5 per cent which was equivalent to film thicknesses of 8, 9 10 11 and 12 μm respectively. Each mix was prepared at 120°C and mixing lasted for exactly 3 minutes after which briquettes were prepared in the Marshall mould in the usual manner. After being stored for 24 hours at room temperature the specimens were broken up and the tar extracted using the hot centrifuge. The Evt was measured, allowance being made for the mineral matter in the tar as described in Appendix N. The effect of film thickness on change of Evt under these conditions is quite marked as shown by the results given in Figure 4 - 28.

4.2.2 Effect of mixing temperature

The effect of varying the mix temperatures on the change in Evt of the 60 - 65°C Evt coke-oven tar was measured in full-scale trials at the mixing plant of the National Institute for Transport and Road Research and also at the Johannesburg City Council plant at Ophirton Johannesburg.

In a preliminary investigation the relationship between the temperature of the various mix components and the final mix temperature was determined for both the continuously graded basecourse mix and the semi-gap-graded mix described previously. The temperatures of the fine, medium and coarse aggregate fractions and the binder

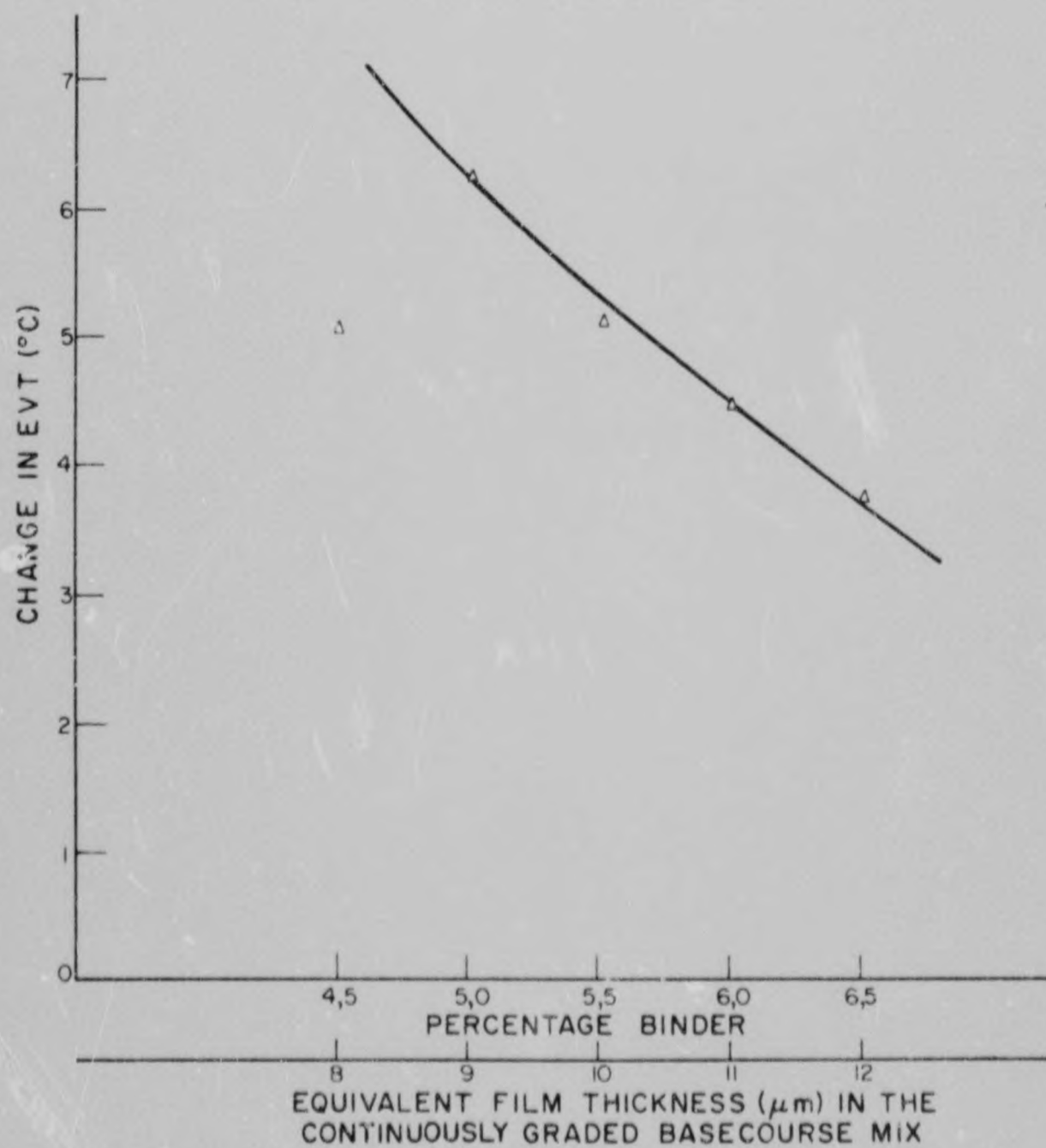


FIGURE 4-28

*THE CHANGE IN EVT OF TAR IN PREPARING MIXES
MADE WITH VARIOUS BINDER CONTENTS (HENCE
VARYING BINDER FILM THICKNESS)*

and resultant mix temperatures were taken during the preparation of a number of mix batches. These results are given in Figures 4 - 29 and 4 - 30.

In both mix types the final mix temperature was more closely related to the temperature of the fine aggregate than the temperature of any other component of the mix; this seems logical as this particular fraction has a large proportion of the total stone mass and a large surface area.

The investigation of change in viscosity as a function of mixing temperature shows that the Evt change increases with increasing temperature and that this is affected by the mix type. During the full-scale preparation of mixes of the continuously graded and semi-gap-graded types the final mix temperature was varied between 100 and 160°C. The binder content of each mix in this investigation was 5.5 per cent which gave a similar voids (Marshall) content and from previous data on laboratory prepared mixes the film thickness of each mix type would be expected to be about 5 μ m. In practice there was some uncertainty as to the constancy of all the other mix variables. The Evt of the binder after mixing was determined in the normal manner and plots of change in Evt versus mix temperature for both mix types are given in Figure 4 - 31. In the preparation of the semi-gap-graded mix type the Evt of the tar increased to a greater extent than when this tar was used in the preparation of the continuously graded mix. These results suggest that tar may be more susceptible to overheating during the preparation of semi-gap-graded mixes than in the preparation of continuously graded mixes.

In the case of the continuously graded mix type the effect of increasing the mixing temperature by a large amount was to cause only a moderate increase in the Evt. A difference of 50°C in mixing temperature (110 - 160°C) caused an Evt increase of only 8°C (this is equivalent to a change of 40 - 50 on the penetration scale).

The use of high mix temperatures in preparing semi-gap-graded mixes improved the coating of the larger stone sizes. The degree of coating in the mixes was assessed by the Ross count method (Ward and Warden, 1959). In this procedure the + 9.75 mm material is separated from a sample of the mix and examined to determine the number of particles having some portion uncoated by the binder. Such particles are classified as "uncoated" and the number of "uncoated" particles per 100 particles of + 9.75 mm stone is used to express (numerically) the degree

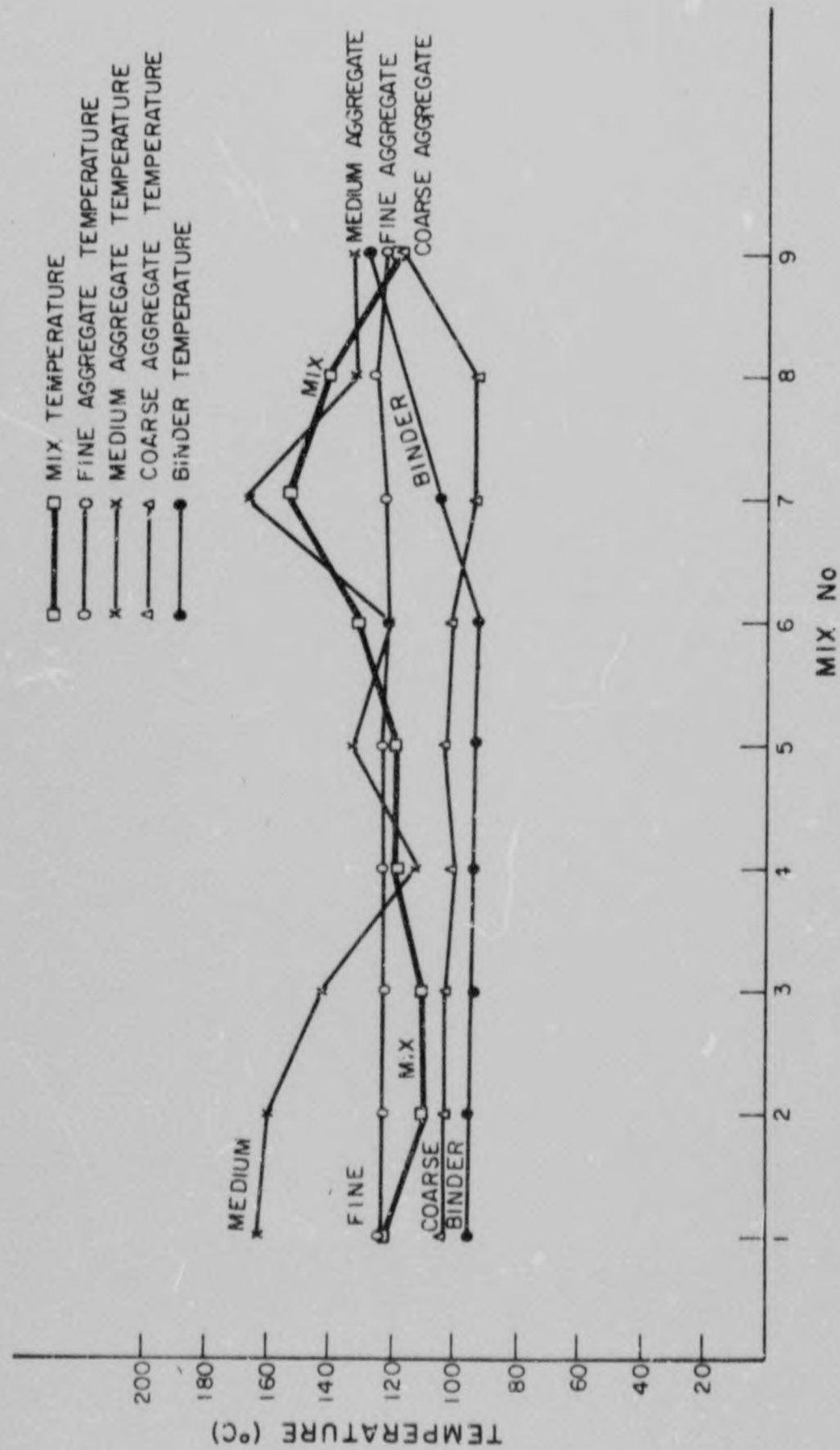


FIGURE 4-29

TEMPERATURES OF THE VARIOUS AGGREGATE FRACTIONS THE BINDER AND THE MIX, RECORDED DURING THE PREPARATION OF THE CONTINUOUSLY GRADED MIXES.

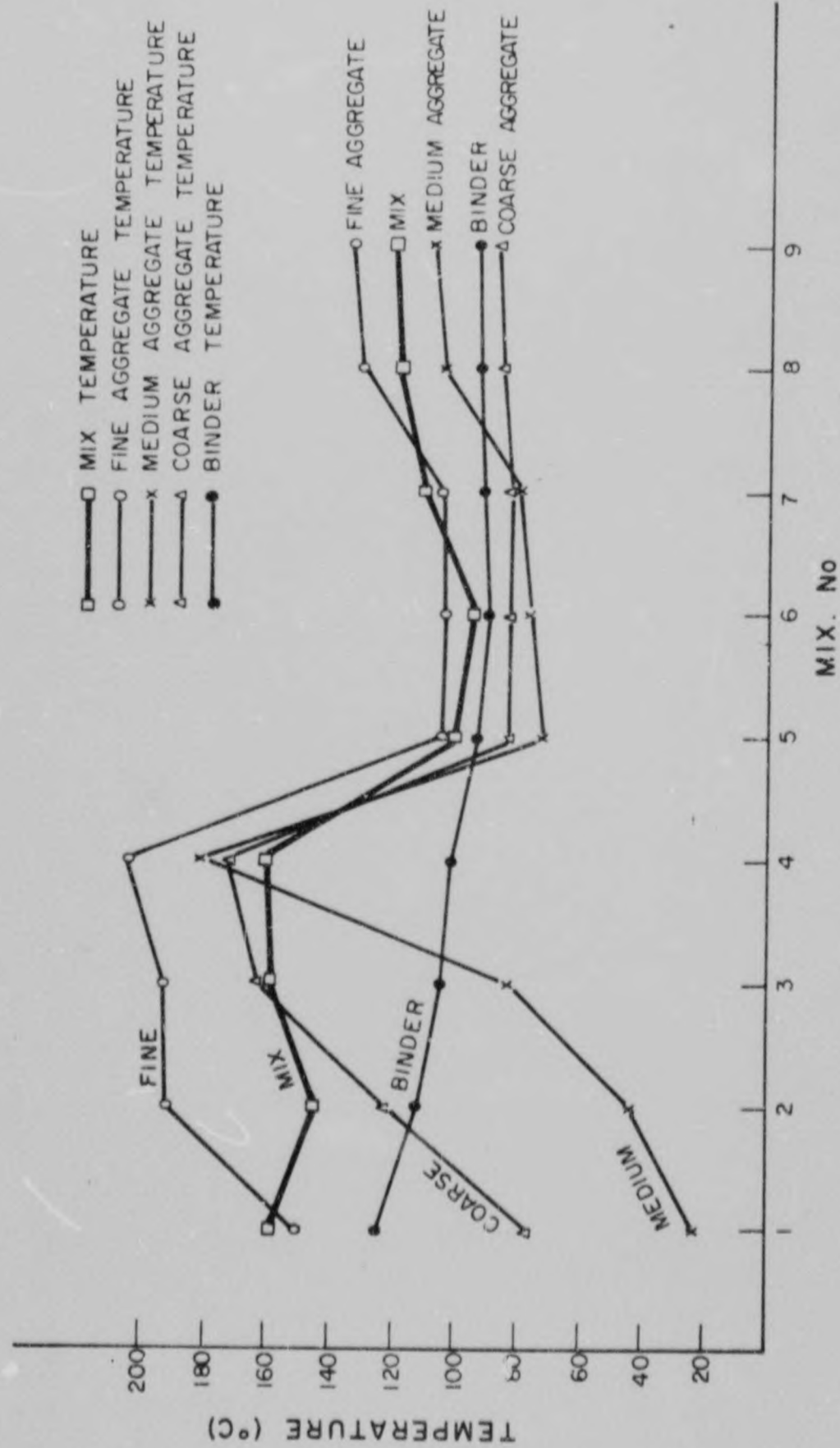


FIGURE 4-30

TEMPERATURES OF THE VARIOUS AGGREGATE FRACTIONS, THE BINDER AND THE MIX, RECORDED DURING THE PREPARATION OF THE SEMI-GAP GRADED MIXES.

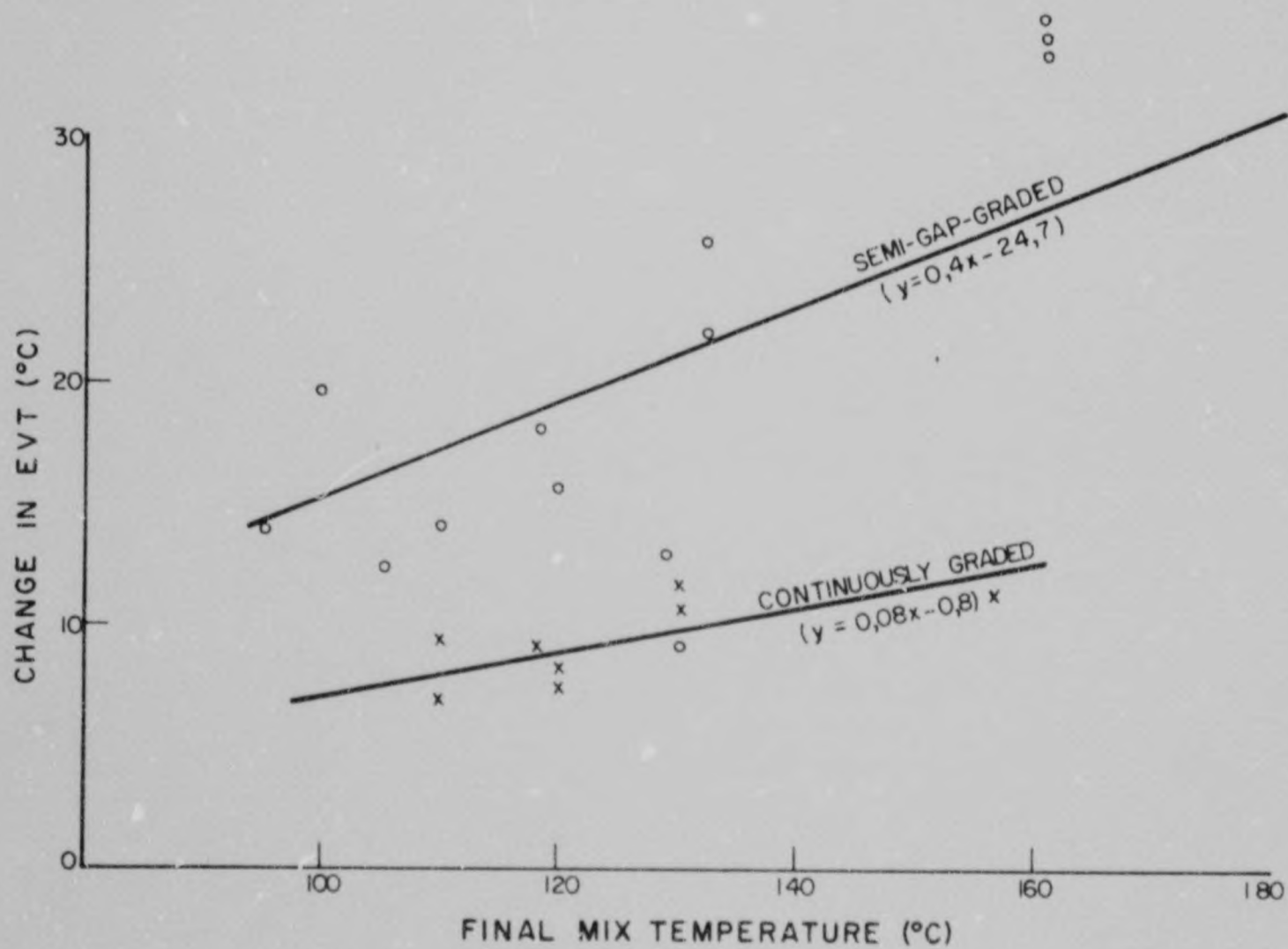


FIGURE 4-31

EFFECT OF MIX TEMPERATURE ON CHANGE OF EVT OF THE TAR

of coating. In the continuously graded mixes all the large particles were fully coated but in the case of the semi-gap-graded mixes there were some "uncoated" particles. Some results are given in Table 4-12.

TABLE 4 - 12

EFFECT OF MIX VARIABLES ON THE COATING,
(ROSS COUNT), OF SEMI-GAP-GRADED TAR MIXES

Mix No.	Binder content (%)	Mixing time (min)	Final Mix temp. (°C)	% Stones uncoated
1	6,0	3	120	6,3
2	5,0	3	118	2,0
5	6,0	5	100	8,0
6	6,0	3	105	4,3
9	6,0	3	160	0,3
10	6,0	1	125	9,0
11	6,0	1	124	22,0

These results imply that, as might be expected, good coating is associated with high mix temperatures and long mixing times. However, the results take on a particular significance when it is realised that mixing times of one minute are normal in most commercial mixing plants. (In fact the mixing time is a very important factor affecting the profitability of an asphalt road surfacing contractor) For this reason it is obviously desirable to use as high a temperature as possible during the preparation of these mixes.

It is apparent that in preparing semi-gap-graded mixes there is a conflict between the need to maintain temperatures as low as possible, to minimise the change in viscosity in mixing (and the ultimate stiffness of the mix), and the need to use high temperatures to ensure adequate coating of the larger aggregate particles. In order to avoid the use of high temperatures the employment of lower viscosity tar grades may be advantageous in certain cases but this would be inappropriate where the mix is to be used to support heavy loads. In Figures 4 - 29 and 4 - 30 it was shown that the binder temperature appeared to have less of an influence on the final mix temperature than did the temperature of the fine and medium aggregate fractions. However, an

increase in binder temperature for a short period may reduce the binder viscosity sufficiently to improve binder coating. For this reason it has been suggested (Appendix L) that in specifications the maximum binder temperature during mixing may be permitted to exceed the recommended maximum mix temperature by a small amount, e.g. in preparing mixes with 60-65°C Evt tars a maximum binder temperature of 135°C is suggested provided that the mix temperature on discharge does not exceed 125°C. This measure may also help to alleviate difficulties in preparing tar mixes with damp or wet aggregate.

4.2.3 Effect of the time factor in storage and handling

It has been mentioned that further changes in viscosity of the tars may occur if the mix is held in the uncompacted state at high temperatures for long periods. This aspect has been studied using an adaptation of the Beckton tray test, (The Beckton tray test, which simulates the change in viscosity of tars during mixing, is described in detail in Appendix O and its use in tar specifications is discussed in Appendix L). The test consists of placing a container holding a film of tar of specified thickness in an oven maintained at a prescribed temperature, which is usually the appropriate mixing temperature, for a stipulated period after which the change in Evt of the tar is measured.

In this study 0,2 mm and 1,25 mm thick films of 60 - 65°C Evt coke-oven tar were exposed for periods varying up to 84 minutes at 120°C. In each case the change in Evt was determined and the results are given in Figure 4 - 32.

The results indicate that the Evt is likely to increase on storage of tar mixes and this increase will be greatest with the thin films associated with mixes (5 μ m) . It may be inferred from these results that tar mixes should be paved as soon after mix preparation as possible and that they should not be stored for lengthy periods.

However, in a separate study of the change in Evt of coke-oven tars after the various stages of mix preparation, handling and application it appears that under normal circumstances the change in Evt of the binder mainly occurs during the actual mixing with the aggregate and no further significant increase occurs during transportation and handling. Investigations were carried out on two tar mixes: a continuously graded basecourse mix incorporating a 60°C Evt tar and a semi-gap-graded surface mix with a 54°C Evt tar. Tar samples were taken prior to mixing and mix samples were taken from the mixer, from the mix loaded into the lorry, the mix

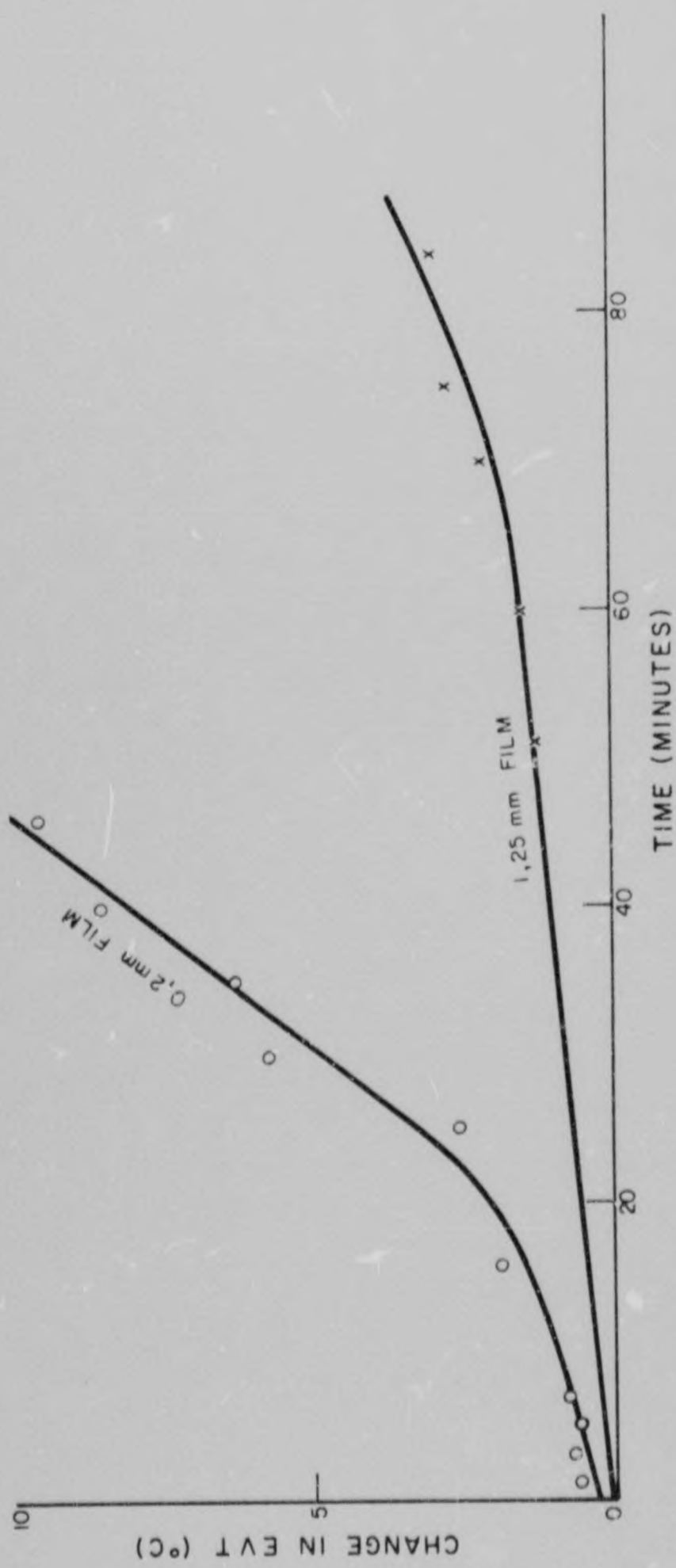


FIGURE 4-32

THE EFFECT OF STORAGE TIME ON THE CHANGE IN EVT OF 0.2 mm AND 1.25 mm FILMS OF COKE-OVEN TAR (60-65°C EVT) AT A TEMPERATURE OF 120°C

charged to the paving machine and the mix from the road immediately after paving. The tar was recovered from the mix samples and the Evt determined in the manner previously described. In order that, if necessary, the significance of Evt changes at each successive stage of the production could be established by the null hypothesis procedure, eight samples of the materials were taken at each stage. Samples were taken at five points and their purpose was as follows:

- | | |
|---|---|
| (1) Eight samples of tar drawn from the tank during the preparation of one lorry load of mix. | - To establish the variance of error in determining the Evt of the tar. |
| (2) Eight samples of mix from one skip i.e. one mix batch | - To estimate the variance in recovering the tar and determining the Evt, and by comparison with 1 to determine whether a significant change in Evt occurred during mixing. |
| (3) Eight samples of mix from one lorry | - To estimate the variance in the Evt of the tar in the lorry load of tar mix |
| (4) Eight samples of mix from the paver | - To estimate by comparison with (3) whether a significant change in Evt occurred in transportation and in loading the mix into the paver |
| (5) Eight samples of mix from the road | - To estimate by comparison with (4) whether a significant change in Evt occurred during the paving of the mix |

The Evt of the tar at different stages in the preparation and paving of the mixes is shown in Figure 4 - 33. The greatest change in Evt, in both cases, occurred during the mixing stage and no significant further Evt increase occurred once the mix was in the lorry. In the case of the basecourse mix it appeared that a significant increase in Evt also occurred during the period when the mix batches were being accumulated in the storage hopper prior to filling up the lorry.

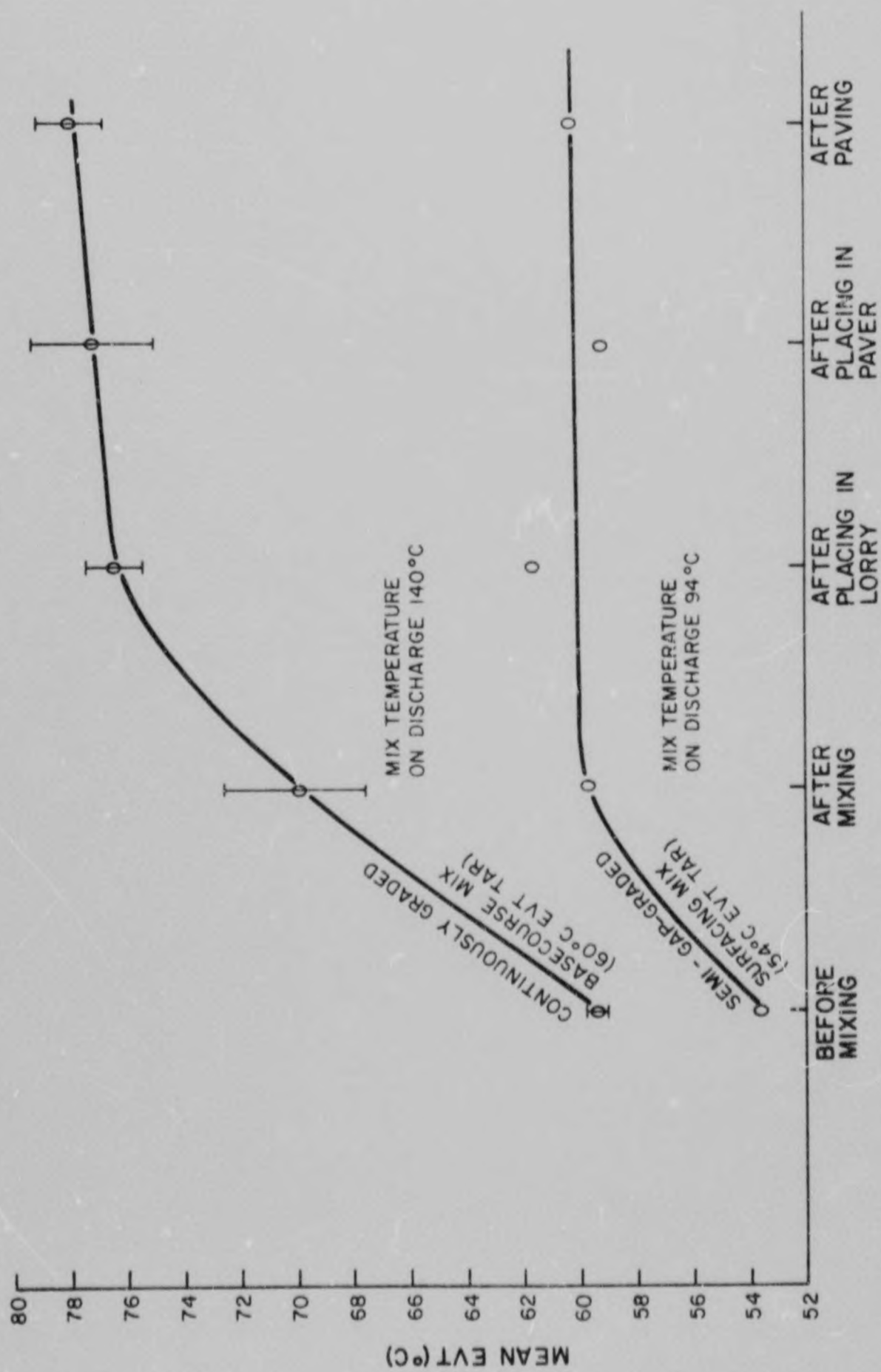


FIGURE 4-33
THE EVT OF COKE - OVEN TAR AT VARIOUS STAGES OF MIX PREPARATION AND PAVEMENT CONSTRUCTION.

During mix preparation and paving in the construction of the Bokslurg experimental sections samples were taken to enable the susceptibility to change in viscosity of the various binders to be measured. (The standard test properties of the binders are given in Appendix K, Tables XXIV and XXV). Exactly the same procedure was followed as previously; eight samples being taken at the various stages of preparation and paving of the mix (TPA medium type). The change in Evt of the five binders at each stage is shown in Figure 4 - 34.

The change in viscosity of the unmodified coke-oven tar and PVC-modified coke-oven tars was less than that which occurred with the 150/200 Pen. normal and "cracked" bitumens. This result, which may seem surprising is partly due to the fact that higher temperatures are used for the mixing of bitumen mixes than in preparing tar-bound mixes. Details of the actual temperatures at various stages of production and handling of these mixes are given in Table 4 - 13.

TABLE 4 - 13
AVERAGE MIX TEMPERATURES DURING CONSTRUCTION

Sec. No.	Binder type	Temperatures (°C)						Time from mixing to paving (mins.)
		Stone	Binder	Mix on disch.	Mix at site	Mix in paver	Mix on road	
2	C.O. tar	115	85	105	102	-	94	65
5	Lurgi tar	115	102	120	104	-	94	60
8	PVC/tar	115	92	105	104	96	94	55
11	Bitumen	135	111	130	146	-	92	55
14	"Cracked" bitumen	147	135	149	-	128	106	55

Thus if coke-oven tar-bound mixes are prepared at temperatures within the specified ranges changes in the viscosity of the tar may be of the same order or less than occurs in bitumen in preparing bitumen-bound mixes (provided these are also prepared at recommended temperatures). This finding may be affected to some extent by the type of mix in which the binder is used.

The Lurgi tar was most susceptible to change during the mix preparation phase and in view of the more rapid weathering of this binder it is unlikely to perform as well in surface mixtures as coke-oven tar or bitumen binders.

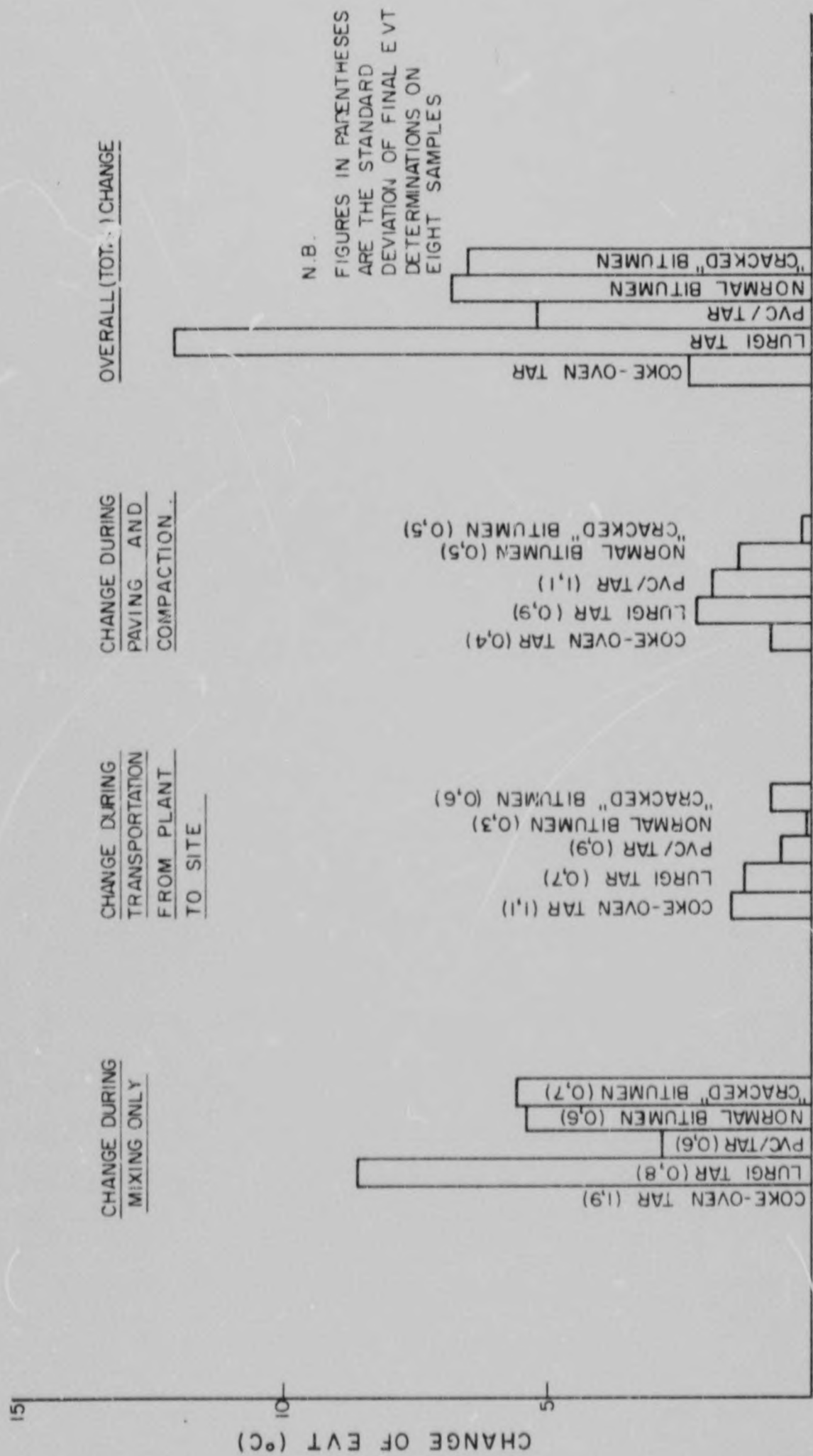


FIGURE 4-34

CHANGE IN EVT OF THE VARIOUS BINDERS USED IN THE BOKSBURG EXPERIMENT AT DIFFERENT STAGES IN THE PREPARATION AND CONSTRUCTION OF THE SURFACE.

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PART 5

Toxic hazards in using road tars

INDEX TO PART 5

	<u>PAGE</u>
5.1 COAL-TAR CONSTITUENTS AND CANCER.....	213
5.2 THE OCCURRENCE OF LUNG CANCER AMONG WORKERS EXPOSED TO POLYNUCLEAR AROMATIC HYDROCARBONS.....	217
5.3 HYDROCARBON POLLUTION DURING THE PAVING OF TAR MIXES.....	219
5.3.1 Method for trapping volatile hydrocarbons in tar fumes.....	220
5.3.2 Analyses of hydrocarbons.....	220
5.3.3 The composition of tar volatiles emitted during the paving of tar mixes.....	221
5.3.4 Threshold Limit Values for tar fumes.....	223
5.4 REFERENCES.....	223

TOXIC HAZARDS IN USING ROAD TARs

Resume'

The history of the association of coal-tar constituents with cancer is outlined. Coal-tar hydrocarbons vary in their carcinogenic potency and polynuclear aromatic hydrocarbons are most potent. The benz(a) pyrene content of vapours has been accepted as an index of their carcinogenic activity although more recently epidemiological evidence has been conflicting. Furthermore, heavy dosages of benz(a)pyrene are necessary to produce skin tumours whereas relatively low concentrations are emitted during paving operations. The main causes of irritation to the eyes and respiratory organs of paving workers are phenol and cresols which have low carcinogenic potency, and measurements taken during a paving operation show that these emissions are much greater than benz(a) pyrene concentrations reported by other workers. Threshold Limit Values for "coal-tar-pitch volatiles" are unrealistically low and a value of 25 mg/m^3 is suggested for the volatile hydrocarbons, as distinct from the particulate polynuclear aromatic hydrocarbons ($200 \text{ } \mu\text{g/m}^3$).

It is true that in road construction and maintenance operations, particularly paving, tar is less pleasant to work with than bitumen. Furthermore, because tar contains an abundance of polynuclear aromatic hydrocarbons, and as these have been found to be carcinogenic, it has been inferred that whenever tar is used operators are exposed to conditions in which there is a high risk of cancer occurring. Such arguments have been used in discussions on the merits of tar and bitumen as road binders (Crawford, 1974). However, a close examination of the evidence, in particular the degree of exposure involved, shows that the health risks are not as great as may be thought.

5.1 COAL-TAR CONSTITUENTS AND CANCER

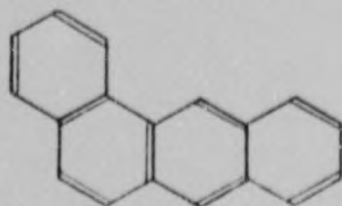
A detailed review tracing the occurrence of cancer among workers handling materials comprised of polynuclear aromatic hydrocarbons has been given in a monograph sponsored by the World Health Organisation

(International Agency for Research on Cancer, 1973).

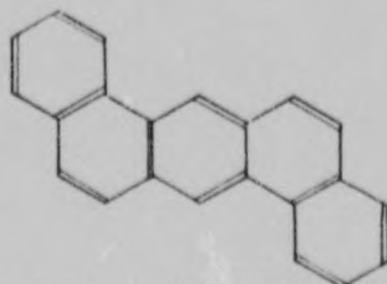
At the beginning of the twentieth century it was realised that individuals engaged in specific occupations in the carbonising and coal-tar by-product industries showed an abnormally high incidence of skin cancer. In Britain the Workman's Compensation Act of 1907 gave official recognition to the fact that pitch or tarry substances could cause ulcers of the skin. Such skin cancers developed only several years after the commencement of the period of exposure and the first recorded description of them appears to have been made by Butlin, (1892).

Yamegiwa and Ichikawa (1915) proved that skin cancer can be caused by contact with tar when they produced tumours on the ears of rabbits upon prolonged application of a tar-oil fraction. Kennaway (1925) confirmed this using high-boiling oil-fractions, and skin cancers produced from tar, pitch and oils (described as creosote oil and anthracene oil) were recorded during the period 1920 - 1945. Later Henry (1946 and 1947) found that the composition and type of tar product being handled was related to the frequency of skin cancer among workers and previously Passey (1922) had found that the most potent extracts of soot were those that were free of "acidic and phenolic compounds".

The first step towards the identification of specific constituents responsible for the occurrence of cancer was made by Hieger (1930) who found that carcinogenic tars had characteristic fluorescence spectra with bands at 400, 418 and 440 m μ m. A hydrocarbon with spectra similar to that of carcinogenic tars is benz(a)anthracene and a derivative, dibenz(ah)anthracene, produced skin tumours and became the first chemical recognised as having carcinogenic potency (Maddow, 1959).

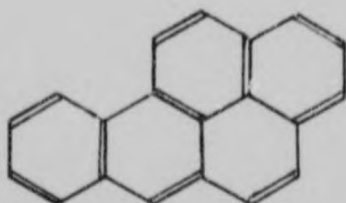


Benz(a)anthracene



Dibenz(ah)anthracene

Following this work Cook and Hewett (1933) isolated the constituent responsible for the characteristic three-banded spectrum of the tar; the hitherto unknown compound which proved to be benz(a)pyrene.



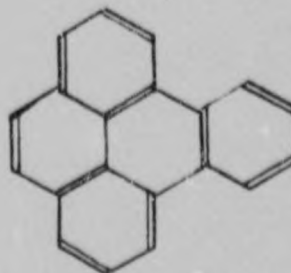
Benz(a)pyrene

Since this time hundreds of carcinogenic hydrocarbons have been identified (Milner, 1954) of which the polynuclear aromatic hydrocarbons are only one class. Benz(a)pyrene is emitted from a number of industrial and domestic processes and is regarded as the most important of this type. Hangebranck et al. (1964) have listed typical concentrations in various emissions:

Gas-fired power station	0,1 $\mu\text{g}/\text{m}^3$
Coal-fired power station	0,3 $\mu\text{g}/\text{m}^3$
Diesel exhaust	5 $\mu\text{g}/\text{m}^3$
Refuse burning	11 $\mu\text{g}/\text{m}^3$
Emissions from coke-oven plants	35 $\mu\text{g}/\text{m}^3$
Household coal-fired furnace	100 $\mu\text{g}/\text{m}^3$

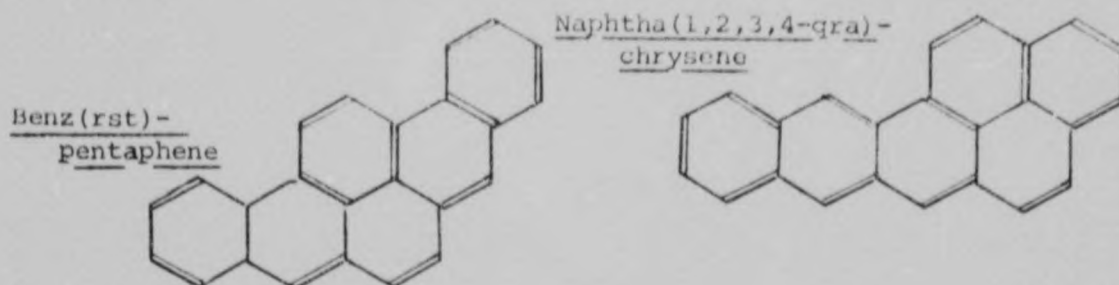
As a result, in examinations of city atmospheres and emissions from various processes the benz(a)pyrene content has been measured more often than the content of any other polynuclear aromatic hydrocarbon and as such is regarded as an index of the carcinogenic risk on exposure to these emissions.

Although the International Agency for Research on Cancer (1973) has described the carcinogenic nature of these compounds as "well established" it must be emphasised that they are by no means all carcinogenic. In fact not all isomers of a single aromatic group are carcinogenic (Haddow, 1959) and for example whereas benz(a)pyrene is a potent carcinogen, benz(e)pyrene has only weak carcinogenic properties.

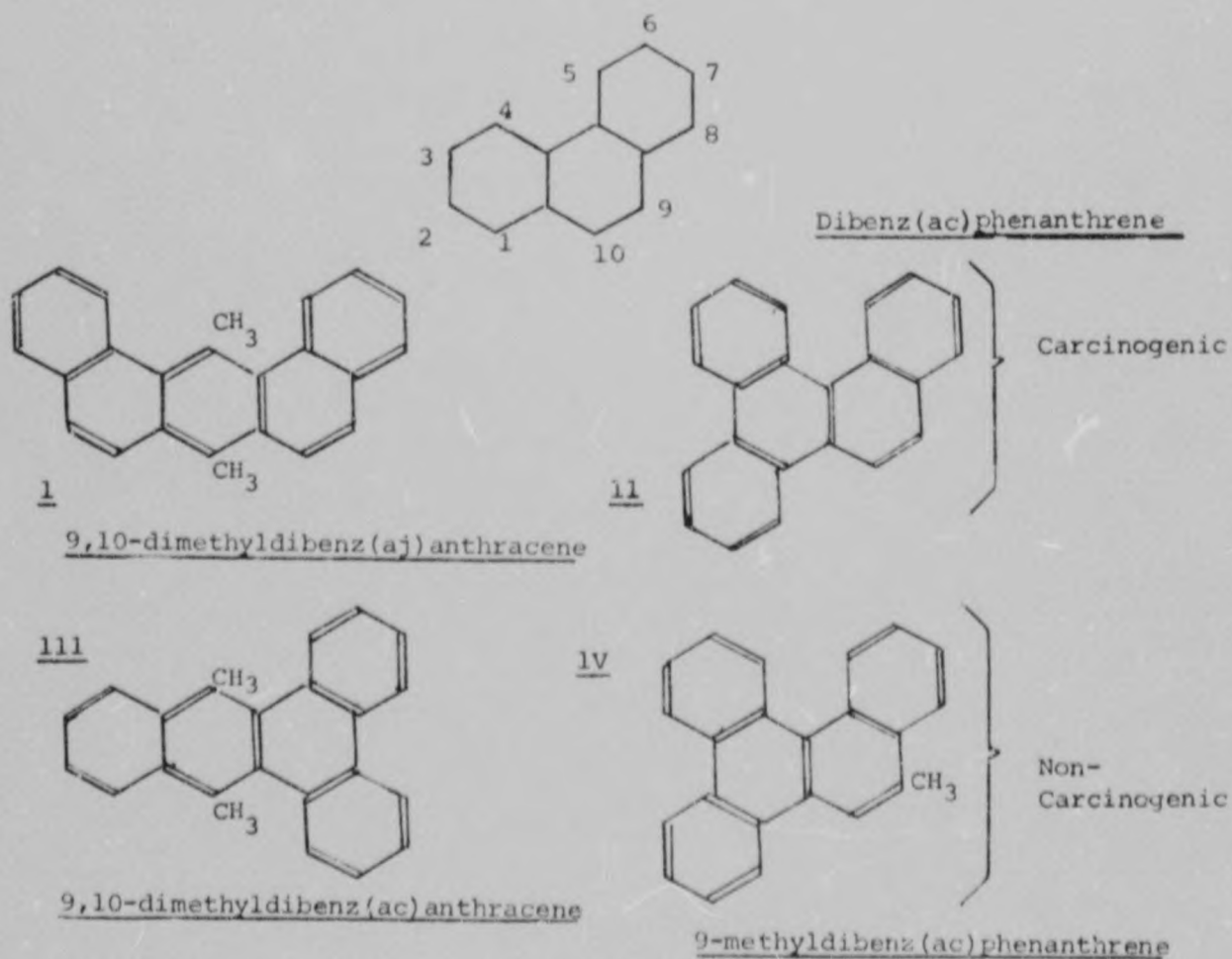


Benz(e)pyrene

Similarly benz(rst)pentaphene is a powerful carcinogen whereas naphtha(1,2,3,4-qr)chrysene has only mild carcinogenic properties (Pierce, and Katz, 1975)



The relationship between the chemical constitution and carcinogenic activity of hydrocarbons has been reviewed by Haddow, (1959). Earlier work showed that highly carcinogenic hydrocarbons tended to have flat planar molecules, and phenanthrene types substituted in the 1,2,3, or 4 positions were potent provided there was no deactivating substituent in the 9 or 10 positions. Thus substances I and II are strongly carcinogenic whereas types III and IV are not.*



This led to the concept of a minimal electron density required for carcinogenic activity (Schmidt, 1938 and 1939) and a threshold activity

* The fact that benz(a)pyrene has strong carcinogenic properties and benz(e)pyrene has low potency, is consistent with this rule.

of π electrons (Pullman and Pullman, 1955). It will be apparent that on this basis few of the low-boiling hydrocarbons among coal-tar constituents could be expected to be carcinogenic.

In view of the concern about the carcinogenic potency of benz(a)pyrene the American Conference of Government and Industrial Hygienists (ACGIH) have assigned a Threshold Limit Value* of $200 \mu\text{g}/\text{m}^3$ to the hydrocarbon and the same concentration has been accepted by the Occupational Safety and Health Association (OSHA) as a maximum air-contaminant standard.

5.2 THE OCCURRENCE OF LUNG CANCER AMONG WORKERS EXPOSED TO POLYNUCLEAR AROMATIC HYDROCARBONS

From results of experiments in which relatively large amounts of coal-tar hydrocarbons have been painted on the skin or injected subcutaneously it has been implied that exposure to tar fumes involves a high risk of cancer of the internal organs. However, the criterion of industrial toxicology should be not whether a substance is capable of harming workmen, but whether "bearing in mind protective measures that are used" actual harm occurs. This view has been given by Gadian (1975) who points out that many highly toxic chemicals such as cyanide are used extensively in industry and ways of protecting workers have been devised.

Benz(a)pyrene concentrations in a number of "tarring" processes have been measured. Values of 0,2 - 1,4 per cent were recorded during the treatment of walls with a tar/pulverised-cork mixture (Bonnet, 1962). In British gas-retort houses average values of $1,4 - 4,8 \mu\text{g}/\text{m}^3$ have been reported with peak values of up to $2,3 \text{ mg}/\text{m}^3$ by Lawther et al. (1965) and in South Africa Brandt and Nicholls (1968) recorded $23,1 \mu\text{g}/\text{m}^3$. These levels

* To provide guidance in choosing concentration levels above which a danger to health may exist, bodies such as the ACGIH have defined Threshold Limit Values (TLV's) for the concentration in air of many substances encountered in industry. The basis on which values are established may differ from substance to substance; protection against the impairment of health may be the guiding factor for some, whereas reasonable freedom from irritation, narcosis, nuisance or other forms of stress may be the basis for others.

In selecting the values, it is assumed that air is inhaled at a normal rate for eight hours and no definite statement is made regarding the extent to which transient concentrations above the TLV may be allowed. Where it is necessary to work for periods in excess of eight hours the dosage would increase and hence a lower TLV would be advisable. A similar principle would apply to increase in working temperature and increase in the amount of heavy work as these result in a greater depth of respiration and the absorption of the toxic substances would correspondingly increase.

are higher than the mean annual values recorded in city atmospheres, (Louw, 1965).

Epidemiological studies on workers exposed to relatively high concentrations of polynuclear aromatic hydrocarbons have given conflicting results. A high incidence of lung cancer among workers exposed to coal-gas and tar was reported by Kennaway and Kennaway, (1947). Doll (1952) found that of 2 071 male pensioners of a London gas company 25 deaths occurred due to lung cancer as compared with 0,67% for the male inhabitants of London of the same age. In a further study Doll et al. (1972) found a significant excess of deaths from lung cancer among gas workers. Similar findings have been reported in Japan (Kurela and Kawahata, 1936; Kawai et al 1967), and in America a study by Lloyd (1971) of almost 60 000 steel works employees showed that the death rate from lung cancer of coke-oven plant operators was twice that of the steel workers as a whole.

However, a study by Hammond et al. (1969) showed that of 6 000 workers in pitch roofing occupations 43 deaths were due to lung cancer as compared with the 36,4 deaths expected (using the total male population of the USA as a control), and this was found to be not statistically different by the null hypothesis that employment in the pitch roofing trade has no influence on death rates. This study also showed that the number of deaths due to coronary heart disease was six times the number of deaths due to lung cancer. Hueper et al (1962) found no correlation between the benz(a)pyrene content in the urban and rural environments of eight American cities and the results of an experimental bio-assay thus refuting contentions that urban dwellers were more prone to lung cancer because of higher benz(a)pyrene concentrations.

From the results of their investigations, Hammond et al. concluded that the effect of benz(a)pyrene on lung cancer is trivial compared with the increase in lung cancer deaths associated with heavy cigarette smoking, and pointed out that the amount of benz(a)pyrene inhaled by the cigarette smoker and also the average city dweller is low in comparison with that required to induce lung cancer in mice. In fact the methods required to induce lung carcinoma in laboratory animals have been described as drastic (Jackson et al., 1974; Saffiotti et al; 1964 and Pybus, 1964). Furthermore, where epidemiological evidence has shown a significantly higher incidence of lung cancer among workers exposed to polynuclear aromatic hydrocarbons, the number of deaths has been a very small percentage (about 1 per cent) of the total (Doll, 1952).

It would seem that although there has been a world-wide increase in mortality from lung cancer, the toxicological mechanisms of polynuclear aromatic hydrocarbons, (and particularly benz(a)pyrene) are mainly undetermined.

5.3 HYDROCARBON POLLUTION DURING THE PAVING OF TAR MIXES

Although concern over the carcinogenic properties of polynuclear aromatic hydrocarbons may be out of proportion to the death and suffering that has actually occurred as a result of exposure to emissions containing these constituents, it is obviously necessary to continue to investigate this subject. The compositions of emissions from asphalt mixing plants have been determined recently by The Asphalt Institute, (1975) in a comprehensive study. The concentrations of carbon monoxide, sulphur dioxide, phenolic substances and C_2-C_{14} gaseous substances were determined as well as solid particulates of polynuclear aromatic hydrocarbons. The multi-disciplinary research team, found that no serious public or employee health hazard existed and the emissions fell below the Threshold Limit Value for asphalt fumes (5 mg/m^3) (ACGIH, 1974).

Road construction workers are more likely to be exposed to high concentrations of toxic substances during the paving operation than workers employed in mixing plants, because the mixing process is usually controlled by remote methods. A number of workers are required in close proximity to the paving machine and bitumen and tar fumes may well be a health hazard at this stage. In the author's knowledge no comprehensive study has been made of emissions during paving. Benz(a)-pyrene concentrations have been determined by Sawicki et al. (1962) in America and by Louw (1965) who found a concentration of $1.1 \text{ } \mu\text{g/m}^3$ in the air surrounding the driver operator in a "tarring" operation in South Africa. This is lower than concentrations ($14 \text{ } \mu\text{g/m}^3$) endured by pitch workers in the study by Hammond et al. (1969) described previously.

Little has been published regarding the broad spectrum of concentrations of organic pollutants during paving operations, and it should be realised that the immediate discomfort in these fumes, to the worker, is not the polynuclear aromatic hydrocarbons but the irritation to the eyes, mucous membrane and respiratory system caused by volatile constituents,* notably phenol and creosols and other acidic types.

* McNeil (1966) has described the characteristic odour of tar as a combination of the smell of naphthalene and phenol modified by the admixture of small amounts of pyridine and thiophenol.

"Pitch burns" have been described by Milner (1954) and Pierre et al. (1965) and can be prevented by the use of barrier creams (Centre D'Etude et D'Applications Dermatologiques, 1975) but operators usually complain that it is inconvenient to use masks and goggles to protect the eyes, nose and mouth.

In view of these circumstances the concentration of volatiles in the air surrounding the driver of the paving machine has been measured during the paving of a tar-aggregate mixture. The method consisted of first trapping the volatiles and subsequently recovering them in the laboratory where they were identified and quantified.

5.3.1 Method for trapping volatile hydrocarbons in tar fumes

The procedure used was first described by Grob and Grob (1971) and an adaptation used by the Air Pollution Group of the CSIR was followed. Grob and Grob trapped the C_6-C_{20} hydrocarbon range and identified 108 different compounds.

Full details of the trapping device and the application are given in Appendix P. The particulate matter was first removed from the air stream on a filter and the volatile hydrocarbons adsorbed on a bed of activated charcoal. The trap was set up on a paving machine in such a position that the air drawn through the apparatus was representative of the air surrounding the driver-operator.

Samples were taken during the construction of the Boksburg surfacing experiment and on occasions during the paving of a tar mix overlay near Ladysmith in Natal.

5.3.2 Analyses of hydrocarbons

The primary objective was to identify and quantify the hydrocarbon constituents. Ideal equipment for this purpose would have been a combined gas chromatograph-mass spectrometer system but as this was not available a combination of gas-chromatography and infra-red spectroscopy was considered to be the next most suitable method. Preparative gas chromatography was used to separate and trap constituents responsible for the major peaks. The first indication of their identity was given by infra-red spectroscopy and this was confirmed, or otherwise, by spiking the sample with pure constituents and examining the chromatograms given by high-resolution packed columns. This procedure was very similar to that used for the identification of the constituents of Lurgi tar oils (Part 2, Chapter 2.2.4).

Preparative chromatograms of a test mixture and of hydrocarbons trapped during paving operations with coke-oven tar and with PVC/tar

are shown in Figure 5 - 1. This figure gives an indication of the resolution obtained with a 1,1 m x 9 mm I.D. column packed with 10 per cent E30 on 100-120 mesh diatomite C. A standard Pye manual preparative kit providing for splitting of the gas stream between the column and the detector was used. The split ratio was 100/1. The kit has a "collector oven" to prevent condensation of the volatile constituents prior to entry into the glass trap. After some experimentation it was found preferable to use simple glass u-tubes for collecting the constituents and these were packed with glass wool and about 2 ml of carbon disulphide was added. The trap was placed in position when the specified constituent peak emerged as indicated by the GC recorder.

The infra-red spectra were obtained as described in Appendix B.

5.3.3 The composition of tar volatiles emitted during the paving of tar mixes

The concentration of tar volatile constituents emitted during the paving of tar mixes (coke-oven tar - unmodified and PVC modified) in the Boksburg and Bergville-Winterton-Ladysmith surfacing operations are given in Table 5 - 1. These were estimated according to procedures described in Appendix A 1.5, Tables VII to XIX. (Threshold Limit Values (ACGIH) have been included in Table 5 - 1.)

In this work the traps were set up on the paving machine in front of the driver. They were positioned at about chest height and approximately 0,5 metres in front of the driver. The air was drawn through each trap at 2 litres/minute which is slightly below the volume respired per minute by the average adult human.

The problems in obtaining samples representative of the emissions of volatile hydrocarbons are indicated by the results which show that a higher emission occurred during the paving of the unmodified tar mix than in the case of the PVC/tar mix. This cannot be explained on the basis of the volatile content of the respective binders (see Appendix K, Table XXIV) and the mix temperature (Part 4, Table 4 - 13) and must be due to variations in emission concentrations at the sampling points together with unrecorded factors such as wind direction and speed.

The total concentration of volatiles is many times greater than the benz(a)pyrene concentrations previously recorded by Sawicki et al. (1962) (100 times greater) and Louw (1965) (1 000 times greater).

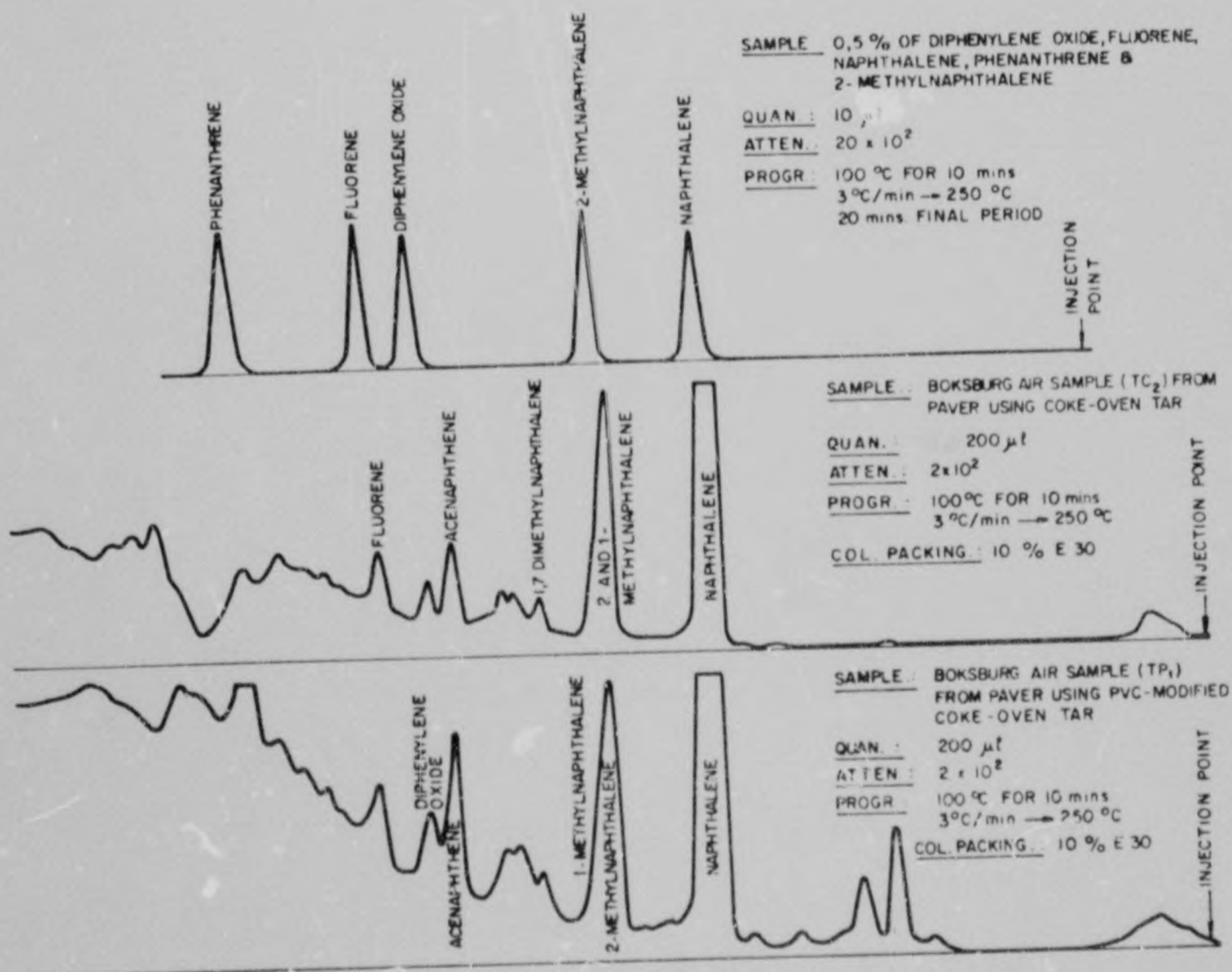


FIGURE 5-1
PREPARATIVE CHROMATOGRAMS OF HYDROCARBONS USING 10% E30
PACKING IN A 1.1m x 9mm ID COLUMN

TABLE 5-1
VOLATILE HYDROCARBONS IN THE AIR SURROUNDING THE
DRIVER-OPERATOR OF A PAVING MACHINE DURING
THE LAYING OF MIXES CONTAINING COKE-OVEN
TAR BINDERS

Hydrocarbon identified	Boksburg paving operation				Bergville-Winterton- Ladysmith paving operation		Threshold* Limit Value (ACGIH) Time weighted average mg/m ³
	Unmod. adsorbed mg	coke-oven tar conc. mg/m ³	PVC mod. adsorbed mg	coke-oven tar conc. mg/m ³	Unmodified adsorbed mg	coke-oven tar conc. mg/m ³	
Phenol + Cresols and other constituents boiling at about 200°C.	0,1	0,2	0,7	1,9	0,3	0,8	19
Naphthalene	3,8	8,4	2,6	6,9	7,7	19,9	50
2 Methylnaphthalene	1,0	2,1	1,1	2,9	1,8	4,7	
1 Methylnaphthalene	0,4	0,8	0,4	1,1			"Coal tar pitch volatiles (benzene soluble fraction Anthracene benz(a)pyrene phenanthrene chrysene pyrene)" = 0,2
Dimethylnaphthalenes	0,5	1,0	0,6	1,6	0,5	1,4	
Acenaphthene	0,2	0,4	0,3	0,8	1,0	2,5	
Diphenylene oxide	0,2	0,4	0,2	0,5	0,6	1,4	
Fluorene	0,2	0,5	0,2	0,5	0,4	1,1	
Phenanthrene and Anthracene	0,5	1,1	0,3	0,8	0,4	1,0	
Carbazole	0,3	0,8	0,7	1,9	-	-	
Fluoranthene	0,3	0,7	0,2	0,5	0,1	0,3	
Pyrene	-	-	-	-	0,1	0,3	
	-	-	-	-	0,2	0,4	
Total	7,5	16,4	7,3	19,4	13,1	33,8	0,2 as above

The main pollutant is naphthalene but the concentration of phenol and cresols is also high (though both are well below the TLV's for these substances). Phenol-cresols mixtures are described as carbolic acid and these are also toxic substances (Aerospace Medical Research Laboratory 1972,) and it is thought a toxic dose ranges from 300 - 500 mg/kg (body mass).

5.3.4 Threshold Limit Values for tar fumes

The data in Table 5 - 1 show that the total volatile emission, which would be benzene soluble material, exceeds the TLV adopted by ACGIH for "coal-tar-pitch" volatiles. However, in defining "coal-tar-pitch volatiles" the ACGIH report makes particular mention of benz(a)pyrene (as well as some non-carcinogenic hydrocarbons), whereas it has been shown by Louw, (1965) and previously by Sawicki et al. (1962) that the concentration level of these particulates is well below the TLV of the coal-tar-pitch volatiles, $200 \mu\text{g}/\text{m}^3$. It would appear that criteria should be established for truly volatile hydrocarbons as distinct from the particulate matter, (the TLV for the latter might remain at $200 \mu\text{g}/\text{m}^3$). Further investigation is required to establish an appropriate TLV for the volatiles and in the interim, in view of the relatively low toxic effect of the volatile constituents which are present in the highest concentration, a value of $25 \text{ mg}/\text{m}^3$ might be used. This is suggested on the basis of providing minimal irritation at a level which may be accommodated by the tar industry. It is apparent that this object would also best be achieved by minimising the volatile content of road tars.

5.4

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