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FUEL RESEARCH INSTITUTE OF SOUTH AFRICA

TECHNICAL MEMORANDUM NO. 37 OF 1966.

TESTING OF COKING PROPERTIES OF COAL
IN THE LABORATORY.

by

F.H. KUNSTMANN

and

H.D.T. KERKOVIVS

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South Africa has extensive coal deposits but only a small percentage of this coal is suitable for coke production. It is, therefore, essential that the known good coking coal be used efficiently and sparingly, to which end only blends with the minimum quantity of good coking coal **should be** carbonised. In addition, a thorough study should be made of the properties of the coal in newly opened coalfields.

Quick and simple methods are needed for routine control of the coking ability of blends as well as for determining the coking properties of the components of blends and of the coal from newly opened mines or boreholes.

The question whether laboratory tests, carried out with coking coals or blends, allow predictions about their behaviour in the coke oven with reasonable reliability has often been raised; it can be answered with a distinct "yes". Laboratory tests in their present form do not make final large scale tests unnecessary, but they help to reduce the costly and slow large scale tests to a minimum. In many cases where only a small sample of the coal is available, laboratory tests are the only means of obtaining information about the coking ability of the coal.

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To yield a coke, a coal must become plastic at a certain temperature (350° - 450°C), exhibit some swelling in the plastic state and resolidify again with further heating (at about 430° - 500°C) after having given off some gas. It is on these properties of coking coal that all the tests to determine its coking ability in the laboratory, are based.

The best known, and because of the simple procedure involved probably the most frequently used test for the caking and swelling properties of a coal, is the determination of the CRUCIBLE SWELLING NUMBER¹⁾.

In this test 1 g of coal, ground to -72 mesh (B.S.S.), is shock-heated to 820°C in a covered crucible until the flame of burning volatile matter dies out. The swelling number is determined by comparing the resulting coke button with numbered standard profiles, the highest number 9 corresponding to the highest degree of swelling.

This quick test is suitable for controlling the coking ability of coal originating from the same seam and mine, but its results may be misleading when coals of different origin are compared. Certain South African coals with a swelling number of only $3\frac{1}{2}$ yield a reasonable coke whilst for others a minimum swelling number of 7 is necessary to give an acceptable coke.

As an alternative parameter to the swelling number the ROGA INDEX²⁾ is proposed in the International Classification System of Coal. In this test the mechanical strength of a crucible coke, obtained by the carbonisation of a mixture of the coal with standard anthracite, is determined by subjecting it to a drum test. The procedure is briefly the following:

1 g of coal (-0.2 mm) is mixed in a crucible with 5 g of standard anthracite (0.3 - 0.4 mm) and the mixture is carbonised by placing the covered crucible for 15 minutes in

a furnace preheated to 850°C. The coke button obtained is subjected to a drum test, consisting of three treatments of 250 revolutions each. From the weight of the coke button before treatment and the weight of the + 1 mm material after treatment, the Roga Index is calculated according to a formula given by Roga.

The Roga method differs from other methods for the determination of the caking power of a coal in that it uses an inert substance related to coking coal, viz. anthracite. Comparative tests established only a fairly rough correlation between the values of the swelling number and of the Roga Index³⁾. This loose correlation may partly be explained by the fact that the swelling number is mainly determined by the plastic properties of the coal at the moment of resolidification, whilst the Roga Index characterizes more the plastic behaviour over the whole plastic range⁴⁾.

The Roga Index is a very useful test for good and medium coking coals but its results are less reliable with weakly coking coals. Hamaker⁵⁾ gives an explanation for this fact and proposes an improved method, consisting mainly of a modified treatment in the drum to obtain more reliable results.

Another test with a high heating rate, which gives results with a reasonable reproducibility, even with weakly coking coals, is the determination of the BONDING INDEX⁶⁾.

In this method the coking coal is carbonised with such an excess of inert material (coke powder), that virtually every particle of coking coal should be surrounded by coke particles only. A coherent coke is, therefore, not formed, but small isolated conglomerates with coking coal in the centre are obtained. After having separated the unbonded coke powder

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with a suitable sieve, the Bonding Index is calculated from the weight of the sieve residue and that of the coking coal used.

Presumably some relation exists between the caking power as determined by the Roga Index and this parameter, but obviously the Roga method is not intended to determine the quantity of particles cemented together, but rather the strength of the bond which holds them together. With the Bonding test, however, the maximum quantity of coke powder is determined which can be cemented to small conglomerates with a certain amount of coking coal. The results of the Bonding test show a very good correlation with the maximum speed of rotation of the Gieseler plastometer (see p.6).

In the tests described so far a high heating rate is applied. To be able to draw reasonably reliable conclusions about the behaviour of coals in the coke oven, additional tests with a low heating rate should be carried out. The GRAY-KING test⁷⁾ and the DILATOMETER test⁸⁾, which have been proposed as alternative tests for the coking power of coal in the International Classification System of Coals, are such tests.

In the Gray-King test 20 g of coal ground to -72 mesh (B.S.S.) is heated in a silica tube from 325°C to 600°C at a heating rate of 5°C per minute. The coke obtained is classified by reference to a series of standard cokes marked with the letters A to G. When testing a highly swelling coal, part of it has to be replaced by an inert substance such as electrode carbon. The number of grams of coal which has to be replaced by electrode carbon to give a coke of type G (normal coke) is given as a suffix to the letter G. With the Gray-King low temperature carbonisation assay at 600°C in its original form, apart from the coke type, the yields of

coke...../

coke, gas, tar and ammonia are determined whilst the determination, as described in the British Standard Methods for the Analysis and Testing of Coal deals only with the assessment of the coke type. The Gray-King test is mainly used in the U.K. while on the continent the dilatometer test is preferred.

For the dilatometer test the Audibert-Arnu dilatometer in its technically improved form (Ruhr-Dilatometer) with automatic heating regulation and automatic registration of the curve is mostly used.

The procedure in this determination is briefly the following: The coal sample, ground to -100 mesh (B.S.S.), is pressed into the shape of a pencil of 60 mm length and heated in a precisely calibrated steel tube with a heating rate of 3°C per minute. The coal pencil is topped by a steel piston, connected to a writing device, which registers the changes in the volume of the coal pencil on a revolving drum. Figure 1 shows a dilatometer curve obtained with a good coking coal and also gives the figures which are read from it and reported. The recording paper is calibrated so that the dilatation and contraction are read as percentages of the original length of the pencil.

The contraction is caused mainly by the closing of the voids and filling of the small space between coal pencil and wall of the tube under the weight of the piston when the coal softens, but the shrinkage due to devolatilization also plays a role. The main factors determining the dilatation are the degree of plasticity reached and the course of volatilization. The shape of the dilatometer curve is strongly dependent on the heating rate applied and, therefore, the heating rate has to be observed as strictly as possible.

The PLASTOMETER is another apparatus which is widely

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used to study the plastic properties of coal. It is very popular especially in the U.S.A. Several types of plastometers are available for determining the plasticity of coal, but today mainly the Gieseler plastometer^{9,10} is used (see Figure 2). The principle involved in the determination is the following:

A steel retort furnished with a stirrer is filled under a certain pressure with coal ground to -36 mesh (B.S.S.), the stirrer being fixed in a certain position. On top of the stirrer shaft a pulley is fixed with a cord wound around it. The cord passes over a second pulley of the same diameter fitted with a pointer, to indicate movement around the dial which is divided into 100 divisions. The cord carries a weight of 40 grams.

The retort is heated in a metal bath and as soon as the softening temperature of the coal is reached the stirrer, and with it the pointer, start to move. The speed of the pointer movement corresponds to the fluidity of the coal and increases to a maximum, then decreases again to zero at the resolidification temperature. The following characteristic points are noted:

1. The initial softening temperature.
2. The resolidification temperature.
3. The width of the plastic range.
4. The temperature of maximum fluidity.
5. The maximum fluidity in divisions per minute.

Considering the relatively poor reproducibility of the values obtained as a measure of the plasticity, it seems advisable to report not the number of divisions completed per minute, but the logarithm of this figure.

The plastometer method has the inherent weakness of all methods based on the measuring of movement of a solid body

in softened coal. It does not measure the true viscosity because the solid body is not moving in softened coal only, but in a two phase system of softened coal and gas which is formed during the softening process.

With the intention of developing an apparatus which would give more reproducible values than the Gieseler plastometer, Echterhoff¹¹⁾ designed for the BRABENDER PLASTOGRAPH a kneader with a vertical kneading chamber and shaft, with a very well regulated heating system, which made it possible to use the Brabender Plastograph for studying the plastic properties of coal. The kneader shaft is driven by a motor which is arranged as a dynamometer. The torque due to kneading resistance is shown on a balance and at the same time recorded on a chart as a function of time.

Unlike the Gieseler plastometer, the Brabender plastograph is a rotary viscosimeter with a constant speed of rotation. The reproducibility of the values, the initial softening temperature and the resolidification temperature, is excellent. Apart from the temperature points, the recorded curve shows the resistance the kneader experienced during the whole softening process, allowing certain conclusions on the degree of plasticity of the softened mass to be made. Finally the value of the end-maximum in the curve, just before the semi-coke breaks, gives some idea of its hardness. Figure 3 shows a simplified plastograph curve representing only the important details of the automatically registered curve.

Modifying the apparatus for the determination of the expansion pressure of Kortzen-Damm, Sapozhnikov and Bazilevich¹²⁾ developed an apparatus which permits simultaneous measurements

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of the thickness of the plastic layer and of the linear expansion and after-shrinkage of the coal charge during carbonisation. On the figures obtained with this apparatus, known as the SAPOZHNIKOV PLASTOMETER, the classification system of Russian and Polish coal is based. In Western Europe and in English speaking countries this apparatus is little used. The reason for the unpopularity of the Sapozhnikov test may be that its performance is tedious and requires an experienced operator.

In the Sapozhnikov test 100 g of coal are placed in a steel cylinder, the bottom surface of which is heated at a rate of 3°C per minute. A piston exerting a pressure of 1 kg/cm^2 on the charge is connected to a lever which carries a writing device at its end, registering every movement of the piston. In the coal charge vertical channels are provided, one for housing the thermocouple and the other for measuring the thickness of the plastic layer with a calibrated needle. The Sapozhnikov test gives two parameters of the coal, firstly the contraction and expansion curve of the coal charge, which is automatically registered and secondly the more important curves showing the thickness of the plastic layer which is recorded periodically during the test. Bazilevich later replaced the separate plastometer needle and thermocouple with a combined thin plastometer needle and thermocouple.

According to Klepel¹³⁾ the thickness of the plastic layer as determined with the Sapozhnikov plastometer shows a good correlation with the width of the plastic range as determined with the Brabender plastograph.

The penetrometer was also used for the determination of the softening and resolidification temperature of coal. The penetrometer is an apparatus which indicates the softening of a material by the movement of a loaded needle penetrating into it. The formerly frequently used penetrometer, developed

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by Agde and Lyncker¹⁴⁾ for coal testing, is scarcely used today.

Finally, another test, which was formerly frequently used for the determination of the plastic properties of coal, may be mentioned: the "Gas-Flow" method developed by Foxwell¹⁵⁾. In this method the change in pressure is registered which a stream of nitrogen experiences when passing through a column of ~~coarse~~ ^{- 1/4"} coal which is heated at a certain heating rate. The gas pressure rises when the coal starts to soften, reaches a maximum and drops again when, after resolidification, the semi-coke fissures. The test was modified by various authors, and the interpretation of the curve was changed, but the information gained could be provided more reliably by other test methods like the dilatometer and plastometer methods which have more and more replaced it.

It may be mentioned here, that the coking properties of some coals deteriorate quickly with oxidation. To preserve the coking properties the samples have to be kept under nitrogen or under water.

As already mentioned, predictions about the behaviour of a coal in the coke oven should not be made after having carried out one type of test only. At least two or three different tests have to be carried out before reasonably reliable conclusions on the coking behaviour of the coal can be drawn. One of these tests should involve a low rate of heating. The reliability of the predictions is increased considerably when, apart from these test results, the results of a petrographic analysis are available.

Since it is known that certain coal macerals play an active part in the formation of the coke and others behave only as inert substances during carbonisation, the results of a maceral analysis allow certain conclusions to be drawn on the

coking...../

coking ability of a coal. Naturally the rank of the coal must also be known to draw these conclusions, because a coal with even a very high vitrinite content will not give a reasonable coke if its rank does not lie within certain limits. A rough idea of the rank of a coal is given by the proximate analysis. To get a more definite indication of the rank the results of an ultimate analysis should be available.

The Fuel Research Institute of South Africa has well equipped laboratories for the determination of coking properties as well as the petrographic composition of coal samples. These laboratories co-operate closely with the division of the Institute concerned with the testing of the coke produced on a pilot plant scale or in commercial coke ovens.

Table 1 shows the results of the determinations and tests carried out at the Institute on four samples. The purpose of the compilation of as many results as possible is to establish the correlations existing between the results obtained in laboratory experiments and on large scale carbonisation tests.

In Table 1 the four coals are arranged in the order of decreasing quality of the coke obtainable from them. In the proximate analyses of the four coals, a decrease in the fixed carbon can be observed, from left to right, indicating a decline of the rank of the coals. The coking properties of these samples also deteriorate from left to right, apart from an insignificant improvement of the plastometer result from the second to the third sample.

In the microlithotype analysis no clear trend from left to right can be observed but the maceral analysis shows it again. On the one hand the vitrinite plus exinite content decreases from left to right and on the other hand the percentage of inertinite increases in this direction, resulting

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in a decreasing ratio of the active to the inactive constituents.

Such tests, admittedly empirical, have been designed to provide information on important characteristics of coking coal. Considering the complexity of the mechanism involved with coke formation one cannot expect that the results obtained will necessarily be entirely conclusive. As already mentioned, their special value lies in the screening of samples submitted, so as to reduce the number of expensive and tedious pilot- or large scale oven tests.

Efforts are also being made at the Fuel Research Institute to improve existing tests and to develop new ones.

PRETORIA.

4th October, 1966.

F.H. KUNSTMANN
Principal Research Officer.

and

H.D.T. KERKOVIOUS
Technician.

TABLE 1.

PROXIMATE ANALYSIS, COKING PROPERTIES AND
PETROGRAPHIC ANALYSIS OF COKING COALS AND
TEST RESULTS OF THE COKE MADE FROM THEM

Sample No. of Coking Coal or Blend →		63/206A	61/762A	61/863A	61/679A	
Coking Properties	Proximate Analysis	{ % Moisture	1.0	1.9	2.1	2.4
		{ % Ash	13.0	11.1	12.7	9.9
		{ % Volat. Matter	22.8	30.9	29.9	34.0
		{ % Fixed Carbon	63.2	56.1	55.3	53.7
	Dilatometer	{ Swelling No.	7½-8	4½	3	3
		{ Roga Index	67	52	39	37
		{ % Contraction	23	30	26	30
		{ % Dilatation	74	-7	-21	-30
		{ % Dilat. Amplit.	97	23	5	0
		Plastograph	{ Init. Soft. Temp.	409°	-	-
	{ Resolidif.		468°	-	-	-
	{ Plastic Range		59°	-	-	-
	Plastometer	{ Log. Max. Div./Min.	2.6	1.9	2.0	1.1
	Petrographic Analysis	Microlitotype Analysis	{ % Vitrite	49.5	26.5	23.0
{ % Clarite			0.0	1.8	4.3	4.7
{ % Vitrinertite			45.6	11.4	21.4	19.9
{ % Interm. Mat.			2.9	53.1	46.6	45.0
{ % Fusite			1.5	1.2	1.7	2.1
{ % Carb. Shale			0.5	6.0	3.0	1.6
Maceral Analysis		{ % Vitrinite } Active	73.5	56.3	57.0	55.9
		{ % Exinite }	0.5	13.7	11.0	11.8
		{ % Inertinite	21.6	24.8	28.3	29.2
		{ % Vis. Minerals	4.4	5.2	3.7	3.1
	{ Ratio Act./Inact.	2.8:1	2.3:1	2.1:1	2.1:1	
Sample No. of Coke →		63/206B	61/762B	61/863B	61/679B	
Characteristics of Coke	{ B.S. Shatter Index on 1½"	91	88	89	81	
	{ B.S. Abrasion Index	78	74	70	62	
	<u>Micum Indices</u>					
	{ On 40 mm (M40)	75	70	67	49	
	{ Through 10 mm (M10)	10.0	11.7	15.5	24.1	

M₄₀

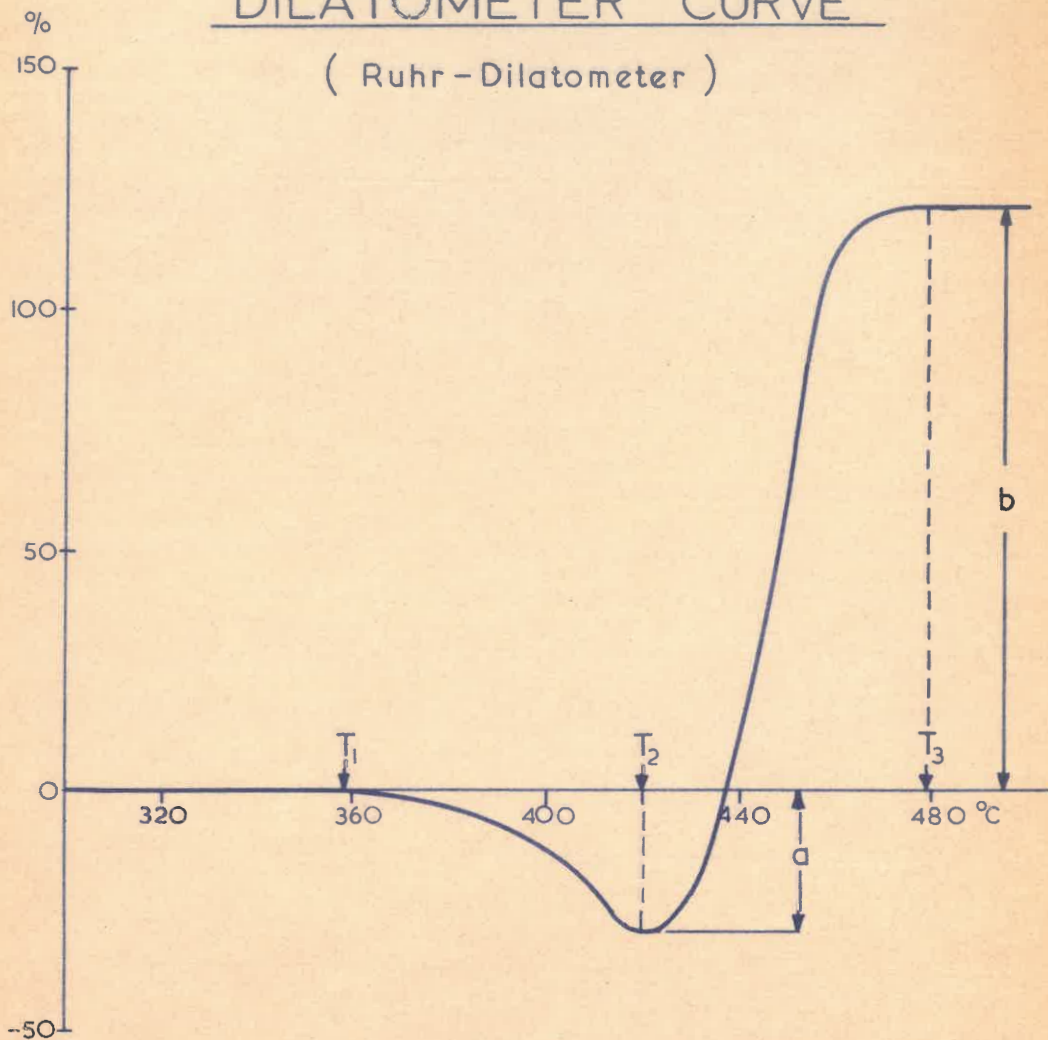
M₁₀

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DILATOMETER CURVE

(Ruhr-Dilatometer)



Initial Softening Temperature	T_1
Temperature of Maximum Contraction	T_2
Temperature of Maximum Dilatation	T_3
Plastic Range	$T_3 - T_1$
Maximum Contraction	a
Maximum Dilatation	b
Dilatation Amplitude	$a + b$
Temperature Rate	$3^\circ\text{C}/\text{min.}$

GIESELER PLASTOMETER

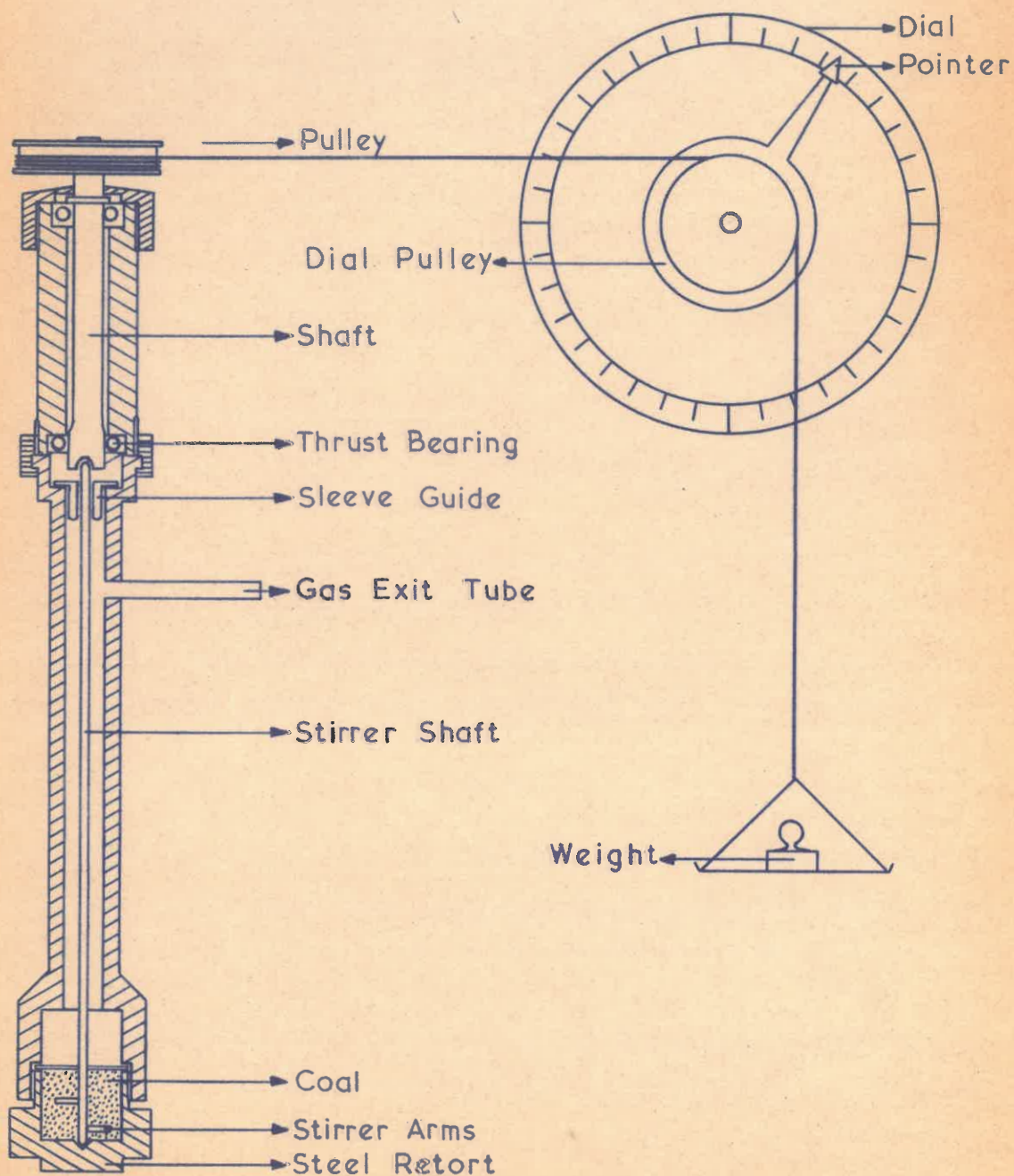
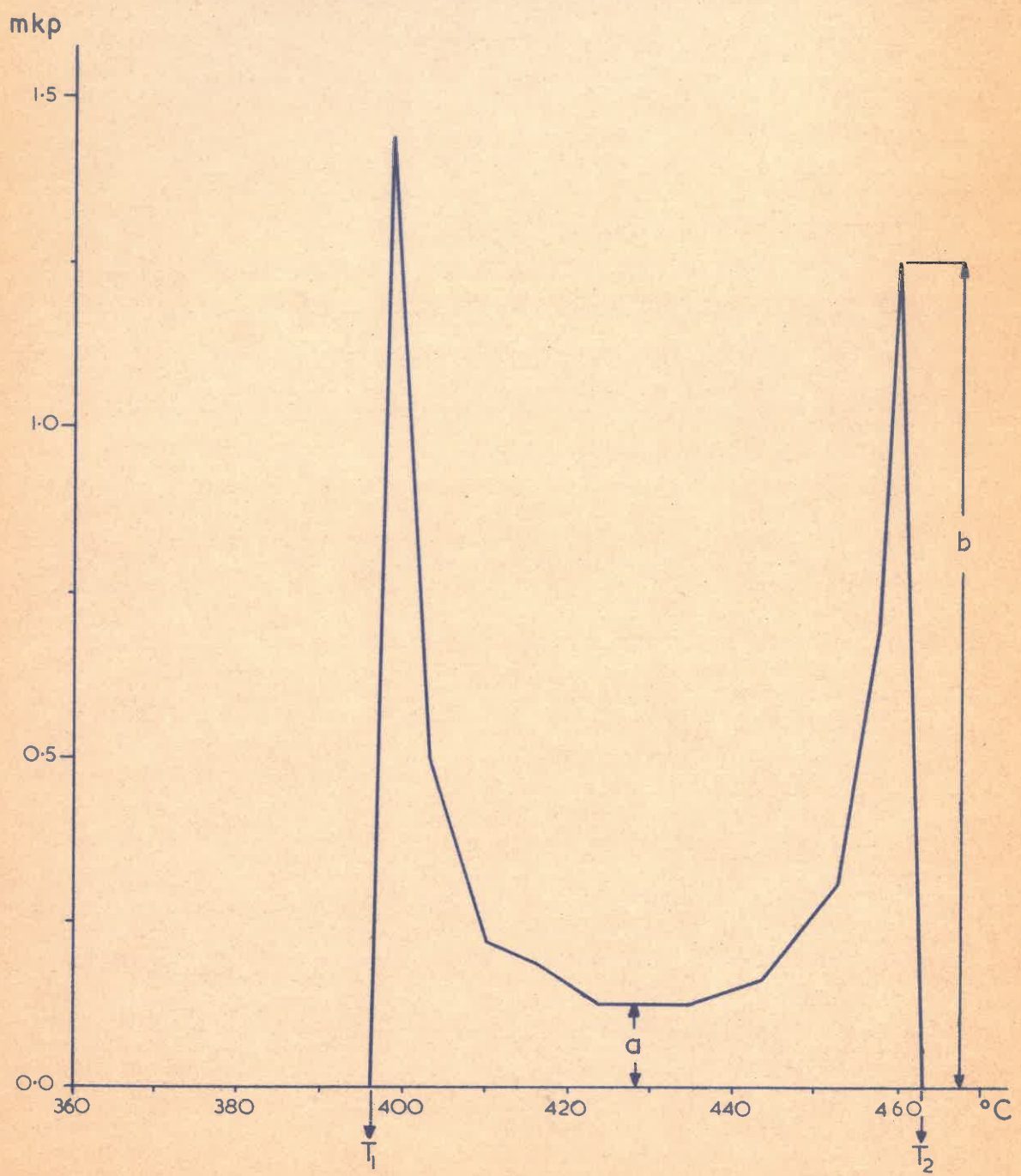


Fig. 3

PLASTOGRAPH CURVE

(Brabender Plastograph)



Initial Softening Temperature	T_1
Resolidification Temperature	T_2
Minimum Resistance	a
End Maximum	b