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VAN SUID-AFRIKA**

**FUEL RESEARCH INSTITUTE
OF SOUTH AFRICA**

ONDERWERP: **LOW-TEMPERATURE CARBONISATION ASSAY OF SOUTH AFRICAN**
SUBJECT: _____

COALS IN A LABORATORY SCALE TEST APPARATUS

PART 1. THE APPARATUS AND EXPERIMENTAL PROCEDURE

CHEMISTRY

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TITLE : LOW-TEMPERATURE CARBONISATION
ASSAY OF SOUTH AFRICAN COALS IN A
LABORATORY SCALE TEST APPARATUS.
PART 1. THE APPARATUS AND EXPERI-
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LOW-TEMPERATURE CARBONISATION ASSAY OF SOUTH AFRICAN COALS IN A LABORATORY SCALE TEST APPARATUS

PART 1 : THE APPARATUS AND EXPERIMENTAL PROCEDURE.

SUMMARY

This report describes the apparatus used to determine the low-temperature carbonisation properties of South African coals. The test procedure is one based upon a precision test method developed by the United States Bureau of Mines, but with the incorporation of changes to the product recovery train.

INTRODUCTION

The rapid increase in the price of imported crude oil has served to generate renewed interest in the technology of coal carbonisation, gasification and the utilisation of carbonisation by-products. This study represents the initial phase of one aspect of an investigation into the low-temperature carbonisation properties of a typical cross section of South African coals, namely : the determination of tar and gas yields in laboratory scale retort tests.

By common acceptance the low-temperature distillation assay of coal has virtually centred around the Fischer assay procedure introduced by Fischer and Schrader¹ over fifty years ago. Notable among other workers who have published assay methods are Strache² who distilled 0,1 gram coal samples in a glass tube ; Dancy and de Jersey³ who proposed a technique of using 2 gram samples and Stansfield⁴ who used iron retorts of up to 3,5 kilogram capacity.

In 1953⁵ Goodman and co-workers published details of a low-temperature retort assay devised to yield increased data accuracy and efficiency for predicting the yields of products from non-caking and low-caking coals.

For the purpose of this study of the carbonisation properties of South African coals, it was decided to use the method of Goodman with, however, alterations to the product collection train to simplify subsequent analyses. Most workers in the low-temperature carbonisation field collect or condense a so-called light-oil fraction in addition to the char, tar, water and gas produced when coal is destructively distilled. The light-oil collected in many cases is of a wide boiling point range and indefinite composition and consists of varying amounts of benzene, toluene, xylenes and even higher aromatics as well as small amounts of carbon disulphide, unsaturated and saturated hydrocarbons and even carbon dioxide where activated charcoal traps are used. Goodman⁶ conducted a series of tests to ascertain the actual recoverability of liquid light-oil from a charcoal trap. He found that only half of the mass gain of a trap in a 500°C carbonisation test was due to liquid oil and that the rest was due to absorbed gases, mainly carbon dioxide.

Also in most cases where tar and light-oil are collected separately, the heavier tar contains a proportion of lighter fractions. Therefore, for the purpose of this study, it was decided to replace the light-oil adsorbent trap with a cold trap from which any condensable products could be returned to the bulk of the tar, and to collect only three carbonisation products namely char, tar (including water), and gas.

Of the various methods available for the collection of non-condensable gases, confinement over saturated or strong brine solutions is probably the most widely used. Corrosion caused by the concentrated solutions used and crystallisation from saturated solutions can prove troublesome, and for this reason it was decided to measure the gas produced during an assay by venting it through a precision test meter.

DESCRIPTION OF THE COMPLETE APPARATUS. THE RETORT ASSEMBLY.

The retort based on the design of Goodman⁵ is made from an 18 cm length of mild steel tube, 5 cm outside diameter, with a welded top and bottom. An 18 cm length of 15 mm inside diameter tubing forms the gas offtake tube. The side gas offtake tube made from the same tubing is welded at an angle of 45° to the vertical tube. A ground seat pipe union welded to the vertical tube forms the gas tight closure to the retort. A thermo-couple well welded to the

top half of the union extends into the middle of the retort chamber. The retort is heated in an electrical furnace made from a 52 mm inside diameter by a 23 cm long length of 10 mm wall tubing around which a 2 kW heater is clamped. The tube furnace is suitably insulated. The temperature of the furnace is sensed by a thermocouple in a well drilled into the wall of the furnace tube. The same thermocouple, connected to a cam-follower type controller, is used to control the temperature of the furnace. Both the temperature of the furnace and the coal being carbonised are registered on a multipoint potentiometric recorder.

THE PRODUCT COLLECTION TRAIN

The tar and water receiver is a 500 ml flask fitted with an additional side-neck at 45°. The side gas offtake tube from the retort connects to this tube of the receiver flask with a bored-through rubber bung. In practice this seal is slightly flexible, yet completely gastight.

For the duration of an assay the receiver flask is immersed in an ice bath at 0°C. The small amount of tar mist and water vapour which remains in the gas is effectively removed by condensation in the vertical double-walled dry-ice trap at the flask exit. Strong convection currents in the flask due to the large temperature gradient between the flask and the dry-ice trap aid the collection of the tar mist.

The non-condensable gas which passes through the trap is fed to a wet test gas meter for measurement. A tee-piece in the line to the gas meter allows gas samples to be taken in double-ended gas sample bottles for gas chromatographic analysis. The complete thermal reaction and product collection apparatus is shown in Figure 1. The furnace temperature, retort temperature and cumulative gas yield plotted as a function of time for a typical assay run are shown in Figure 2.

Subsequent to the reproducibility tests reported in Table 1, flexible Teflon* gas sample bags were obtained and used to collect the total gas yield of an assay.

OPERATING PROCEDURE

Except for variations in the assay procedure necessitated by alterations to the product recovery train, the test method is the same as that of the USBM.

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* Alltech Associations Inc USA.

Coal samples for the test are obtained from the normal quarterly product samples of the Survey Division of the Fuel Research Institute. Coal is kept in sealed containers prior to the assay. Replicate distillation runs on minus 4 mm material and also minus 1 mm material showed no substantial differences in the tar or gas yield. The finer material did, however, give more reproducible results and is used for all routine determinations.

For the purpose of a test 150,0 g of air-dried coal is weighed into a tared beaker and transferred into the retort through a metal funnel. Care must be taken to prevent the loss of sample as dust, or down the retort outlet tube. The retort closure and thermocouple well is tightened in place and the complete charged and assembled retort weighed to the nearest 0,1 g. The receiver flask weighed to the nearest 0,01 g is connected to the retort outlet. The trap filled with dry ice is connected in line between the flask and the gas meter. For the alternate gas collection system the trap outlet is connected to an empty gas sample bag. If the totalising gas meter is used to measure the gas produced during a test, two or more "grab" gas samples are taken at suitable intervals during the test for analysis.

The temperature of the retort furnace is increased at a linear rate from ambient temperature to 500°C within an hour and held at 500°C for an additional hour. At the termination of the assay the dry ice is removed from the trap which is then allowed to warm up to ambient temperature. The trap is so constructed that the condensed tar and water drip back into the receiver flask. The receiver flask is weighed to determine the yield of tar plus water. Water is removed by azeotropic distillation and the dry tar determined by difference.

If the gas has been collected in a Teflon bag, its composition is determined by gas chromatography and its volume measured by venting the contents of the bag through a wet test meter.

ASSAY RESULTS AND CONCLUSION

Data in Table 1 summarize results of replicate test assays done to determine the reproducibility of the method. The distillations reported were conducted at 500°C. This was done to correlate future results with numerous data reported for foreign coals.

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Carbonisation product yields for char, tar, gas and water are reported as well as the composition of the carbonisation gas. The chemical composition of the gas is reported on an oxygen- and nitrogen-free basis. Air is entrained in the assay apparatus and is not a part of the gas yield.

Reproducibility of the assay data is satisfactory. As shown in Table 1 standard deviations for the major carbonisation products char, tar and water are 0,35 or less. Coefficients of variation which reflect standard deviations as a percentage of the mean values are 3 per cent or less. The standard deviation for the gas yield is a factor of four higher, possibly due to the use of a faulty gas meter.

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TABLE 1
REPRODUCIBILITY OF THE PRODUCT YIELDS AND GAS COMPOSITION
FROM TEN REPLICATE ANALYSES OF UNION COAL CARBONISED AT 500°C

Product	Yield										Average	Standard deviation
	Assay No.											
	1	2	3	4	5	6	7	8	9	10		
Char %	80,3	80,9	80,7	80,5	80,9	80,9	80,5	80,1	79,9	80,7	80,54	0,35
Tar %	7,9	7,5	7,8	7,4	8,0	7,8	7,6	7,3	7,9	7,9	7,71	0,24
Water %	6,7	6,4	6,6	6,4	6,4	6,7	6,5	6,2	6,4	6,3	6,46	0,16
Gas litre/kg	32,8	38,4	41,9	32,0	36,1	35,7	-	-	38,8	28,9	35,58	4,20
Component	Gas composition volume per cent										Average	Standard deviation
	Assay No.											
	1	2	3	4	5	6	7	8	9	10		
Hydrogen	8,6	7,6	7,9	7,2	8,7	7,6	7,8	8,1	8,5	8,2	8,02	0,49
Methane	47,3	45,3	45,6	43,9	48,1	45,6	45,8	46,0	47,1	48,0	46,27	1,33
Ethane	13,1	12,8	12,9	12,7	12,9	12,9	12,9	12,8	12,9	13,2	12,91	0,15
Propane	3,3	3,0	3,2	3,2	3,6	3,2	3,2	2,9	3,1	2,9	3,16	0,21
Butanes	0,3	0,5	0,4	0,7	0,4	0,3	0,4	0,3	0,2	0,3	-	-
Ethylene	2,7	2,9	2,8	2,9	2,7	2,9	2,8	2,9	2,7	2,9	2,82	0,09
Propylene	2,7	2,5	2,6	2,6	2,8	2,6	2,7	2,4	2,5	2,4	2,58	0,13
Butenes	0,3	0,3	0,3	0,5	0,3	0,4	0,3	0,3	0,3	0,2	-	-
Carbon Monoxide	8,3	9,3	8,9	9,3	8,2	9,0	9,1	9,3	8,8	7,0	8,72	0,72
Carbon Dioxide	13,4	15,8	15,4	17,0	12,3	15,5	15,0	15,0	13,9	14,9	14,82	1,33

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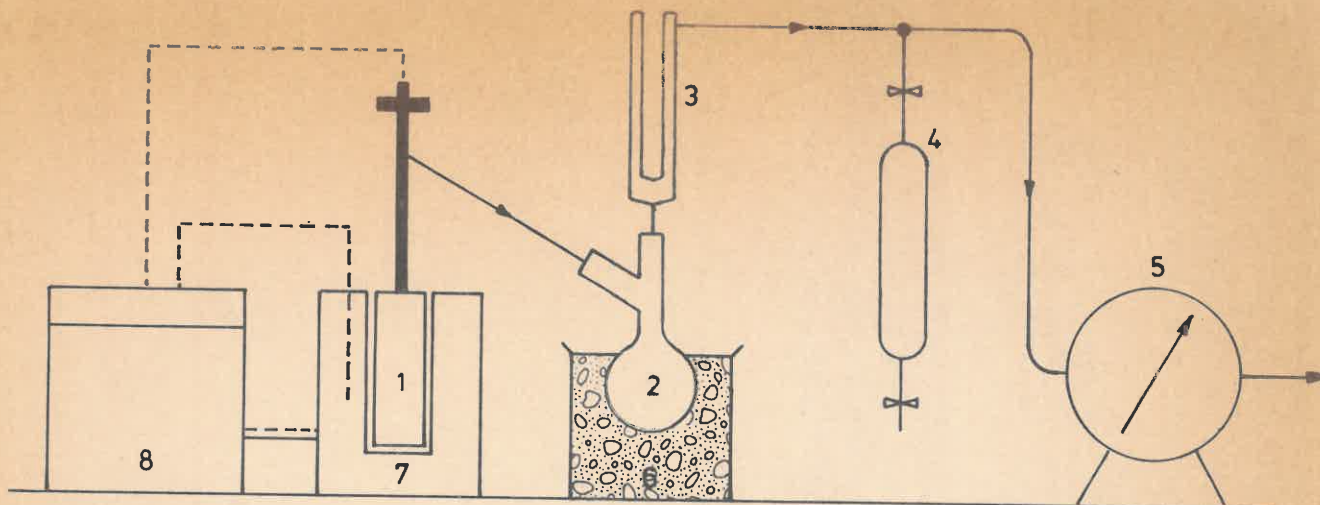


FIGURE 1 THE LOW-TEMPERATURE CARBONISATION ASSAY APPARATUS

- 1) RETORT
- 2) PRODUCT COLLECTION FLASK
- 3) DRY-ICE TRAP
- 4) EVACUATED GAS SAMPLE FLASK
- 5) ELSTER WET TEST GAS METER
- 6) ICE BATH
- 7) RETORT FURNACE
- 8) FURNACE CONTROLLER AND TEMPERATURE INDICATOR

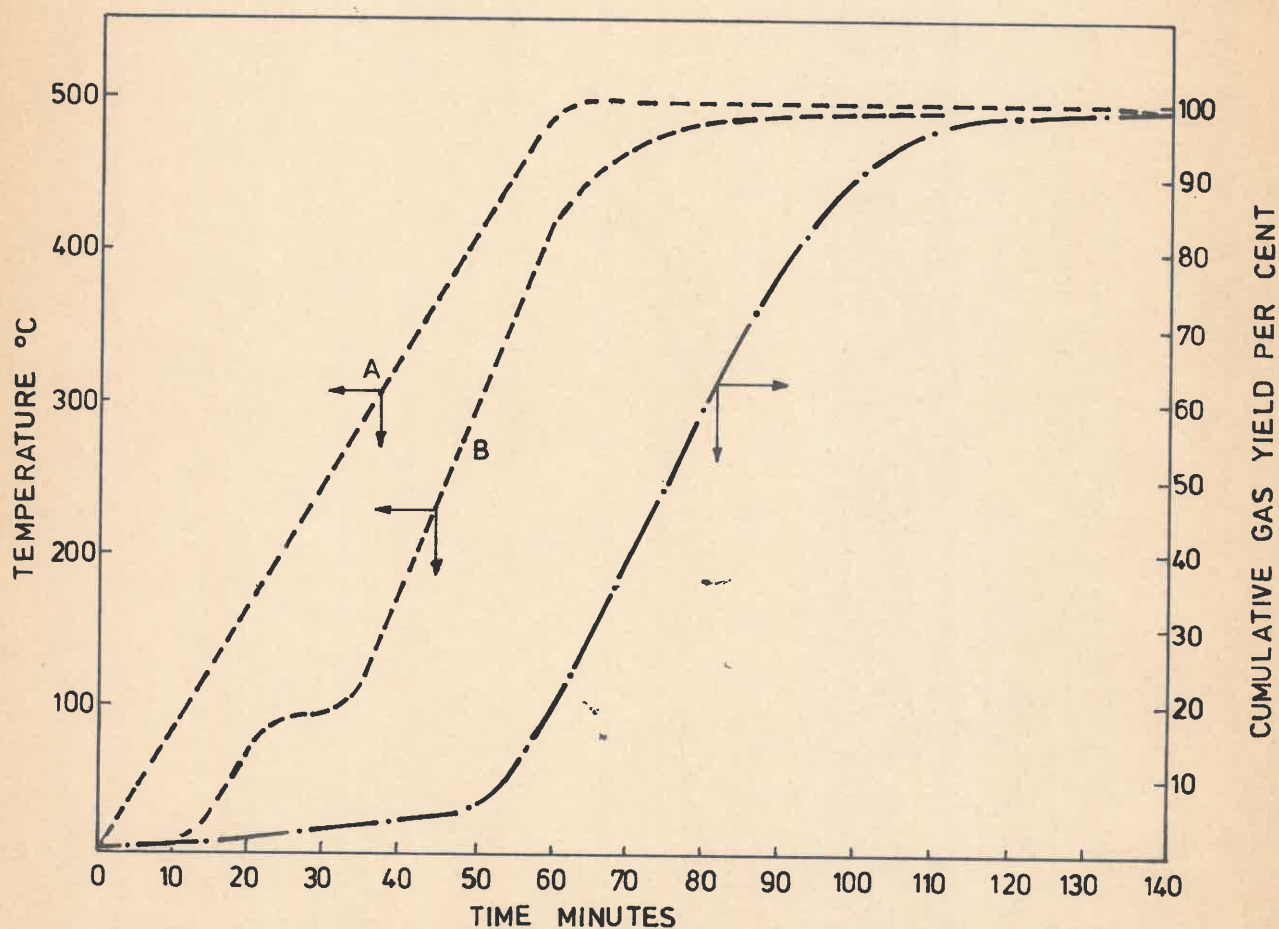


FIGURE 2 OVEN TEMPERATURE, RETORT TEMPERATURE AND CUMULATIVE GAS YIELD PLOTTED AS A FUNCTION OF TIME FOR A TYPICAL ASSAY

- A) OVEN TEMPERATURE
- B) RETORT CONTENTS TEMPERATURE