RESEARCH COUNCIL OF ALBERTA

Information Series No. 37

PILOT PLANT INVESTIGATIONS OF LOW-TEMPERATURE COAL CARBONIZATION IN A FLUIDIZED BED

by

G. Davies and C. Moreland

Research Council of Alberta 87th Avenue and 114th Street Edmonton, Alberta 1961

CONTENTS

															Page
Abstract .	• • • •		•	•	•	•				•	•	•	•	•	1
Introduction											ė		•		. 2
Lo	w-temperature c	arboni	zati	ion	•										2
Fl	uidized low-temp	peratu	re c	arbo	oni z	zati	on	•	•	•	•	•	•	•	5
Experimental	program										•				6
	rst pilot plant		•	÷		•				•		•			6
Se	cond pilot plant		•			•		•		•	•				6
Th	ird pilot plant		•	•	•	•	•	•	•	•	•	•	•	•	12
Discussion .															15
	perimental result	s .	_				•				_	•	•	•	15
	onomics of low-t		atuı	re c	arbo	oni z	zati	on	•		•	•		•	18
Conclusions .	• • • • •	• •	•	•	•	•	•	•	•	•	•	•	•	•	19
Acknowledgn	nents		•	•	•	•	•	•	•	•	•	•	•	•	19
References .			•	•	•	•	•	•	•	•	•	•		٠	20
Appendix 1.	Pilot plant desig	gn .	•	•	•	•	•	•	•	•	•	•	•	•	22
Appendix 2.	Plant operation	and re	sult	ts	•		•	•	•	•	٠.	•	•	•	38
Appendix 3.	Heating method	s .	•	•	•	•	•	•	•	•	•	•	•	•	53
Appendix 4.	Dust entrainmen	ıt and	coll	ecti	ion					•		• •	•	•	57

FIGURES

			rage
Figure	1.	Generalized block flow diagram for fluidized low-temperature carbonization processes	4
Figure	2.	Flow sheet for first pilot plant	7
Figure	3.	Reactor and heater for first pilot plant	8
Figure	4.	Flow sheet for second pilot plant (original form)	9
Figure	5.	Reactor for second pilot plant	10
Figure	6.	Flow sheet for third pilot plant	13
Figure	7.	Glass-wool bag filter unit	24
Figure	8.	Shaking mechanism for glass-wool bag filter unit	26
Figure	9.	Insulation walls, heater arrangement, and circuit diagram for bag filter unit	28
Figure	10.	Tar condenser	30
Figure	11.	Demister	32
Figure	12.	Gas sampling apparatus	32
Figure	13.	Detailed flow sheet for third pilot plant	36
Figure	14.	Air and coal feed rates, and outlet gas rate – Test 1 (December, 1958)	40
Figure	15.	Portion of temperature chart - Test 1 (December, 1958)	42
Figure	16.	Air and coal feed rates, and outlet gas rate - Test 2 (March,1959)	46
Figure	17.	Portion of temperature chart - Test 2 (March, 1959)	48
		PLATES	
_			
l. Se	cond	pilot plant	11
2. Pai	nel b	poard for third pilot plant	14
3. Thi	ird pi	ilot plant (coal feed side)	16
4 TL 9	ا :	that when the collection side	17

PILOT PLANT INVESTIGATIONS OF LOW-TEMPERATURE COAL CARBONIZATION IN A FLUIDIZED BED

ABSTRACT .

A pilot plant scale investigation of fluidized low-temperature carbonization of Alberta subbituminous coal has been conducted by the Research Council of Alberta. The pilot plant in its final form successfully carbonized predried coal from the Edmonton area at 600°C. in a continuous operation. The reactor operated very smoothly and the main technical difficulties remaining unsolved are associated with dust entrained in the gas stream, thermal cracking of the tar, and tar mist entrainment. Modifications to the plant aimed at overcoming these problems have met with limited success.

Tar yields from Alberta subbituminous coals are disappointingly low, and a preliminary economic assessment of the process indicates that, unless a market for char at a premium price arises, low-temperature carbonization of coal will not be commercially attractive in Alberta in the foreseeable future.

INTRODUCTION

The Research Council of Alberta has for several years been interested in the development of a low-temperature coal carbonization process suitable for use in Western Canada. Earlier thinking in this field was directed toward producing a high grade, smokeless solid fuel by upgrading indigenous subbituminous coals, and little attention was paid to the byproducts which might be obtained during the process. The pilot plant described by Gregory and McCulloch belongs to this period.

Subsequent developments, which cost coal a number of major markets that could advantageously have used such a solid fuel, led to a change in thoughts regarding low-temperature carbonization. It was considered that some of the future large scale coal users, in particular the electric utility companies, might benefit by adopting a carbonization process which would supply a solid char product as fuel for the main operation, and simultaneously yield liquid tar and oil products that could be sold for other uses. The potential value of the tar and oil might cover the cost of carbonization, and leave additional revenue for reduction of the effective cost of fuel to the main operation.

Of the many processes which have been described for low-temperature carbonization, the fluidized bed technique seemed to offer the greatest promise under prevailing local conditions. Therefore the Research Council of Alberta undertook a pilot plant investigation of fluidized low-temperature carbonization of Alberta subbituminous coals.

This report outlines the development of the experimental work and includes a general discussion of low-temperature carbonization and a brief description of the design and operation of the pilot plants used. This is followed by a short discussion of the plant operation and of the economics of any commercial application of such a process. Detailed descriptions of modifications made to the plant and the experimental results appear as appendixes.

Low-Temperature Carbonization

The low-temperature carbonization of coal is technically a relatively simple process: essentially, it involves heating coal to a temperature of between 500°C. and 600°C. under conditions that do not permit combustion. At these temperatures the coal decomposes to give four main products:

- 1. A solid residual char or semicoke;
- 2. A liquid tar which is a complex mixture of organic compounds;
- 3. Water;

4. Gas - the chief constituents of which are methane, hydrogen, carbon dioxide and carbon monoxide.

The yields of the various by-products, per ton of coal carbonized, depend upon the type of coal used, the carbonization temperature, and the carbonization process chosen.

Parry et al.² have drawn up a series of tables giving the yields and analyses of these products obtained from various United States subbituminous coals and lignites, while Davies et al.³ have carried out a low-temperature carbonization assay of many Alberta coals. Medium and high volatile bituminous coals give similar products, but tar yields are generally higher and chars are lumpier and much more strongly agglomerated than with low rank coals. If desirable, this agglomeration can be prevented by pretreatment.

The two products of carbonization which have attracted the most interest are the char and the tar. The char is a solid, reactive fuel which has a high heating value, burns smokelessly, and makes a satisfactory domestic or industrial fuel which has often commanded premium prices. The calorific value and volatile matter content of the char depend upon the carbonization temperature, the type of coal used, and, to a lesser extent, upon the carbonization process. In general, calorific values range from 12,000 to 14,000 B.t.u. per lb., and volatile matter lies between 10 and 25 per cent.

The tar is a dark-colored liquid of specific gravity between 0.95 and 1.10. It is a much more complex mixture than high-temperature tar and one of its more interesting features is its relatively high content of high-boiling tar acids. The value and possible outlets for the tar have been the cause of much speculation in the past, and up to the present considerable difficulty has been met in attempts to refine and fully utilize this tar.

The potential value of the char as a high-grade fuel, and of the tar as a chemical raw material, has attracted many engineers and promoters during the last 50 years, and references to low-temperature coal carbonization are numerous⁴. Most of these units were doomed to commercial failure; the few, relatively successful processes (e.g. the Coalite⁵, Disco⁶, and Lurgi⁷ processes) depend mainly upon receiving a premium price for their solid product, usually in the domestic market.

The history of low-temperature carbonization has been beset with difficulties, due mainly to the low heat transfer rates at the low temperature gradients involved, the adverse effects of the swelling properties of coking coals, the long carbonization times, and in some cases the complicated mechanical parts involved. These factors often led to very high capital investment per tonnage throughput for the unit. In addition to these difficulties, the anticipated high price outlets for the tar were never attained.

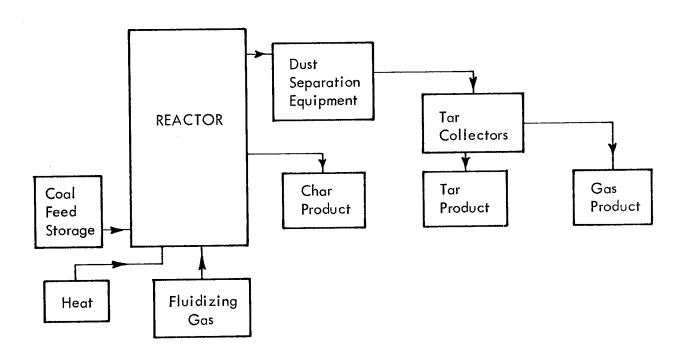


FIGURE 1. Generalized block flow diagram for fluidized low-temperature carbonization processes

Fluidized Low-Temperature Carbonization

The fluidized bed technique for contacting gases and solids, which has developed rapidly during recent years, seemed to offer several advantages over the older conventional processes for low-temperature carbonization of coal. These advantages, which result in savings in capital, labor and energy costs, include high heat and mass transfer rates, ease of solids handling, ability to treat coal fines, and high throughput per unit of reaction space. It is therefore not surprising that several organizations have undertaken experimental work to investigate the possible development of large scale low-temperature carbonization using a fluidized bed reactor. Pilot plant investigations in North America, Europe, and Asia have been described 15. Initial reports of the results of these workers have been promising, but apart from the Parry Unit built at Rockdale, Texas 16, no full scale unit has been operated.

It is not necessary to discuss here the details of the various experimental units, all of which, although differing considerably in detail, consist of the same essential items. These are shown in a simplified block flow sheet in figure 1. Ground coal is fed into the reactor and kept fluidized in a stream of fluidizing gas, and some means of supplying heat is provided such that the contents of the reactor are held at the carbonization temperature. Two streams leave the reactor: (i) the solid char product which usually overflows from the top of the fluidized bed, and (ii) the gaseous products which are carried away in the stream of fluidizing gas. The latter contains the tar, water, and carbonization gases together with entrained solid particles.

The char product is cooled and stored for use as a solid fuel, while the gaseous product is passed through the equipment designed to remove entrained solids, and then cooled to condense the tar and water, which are collected both from the condensers and in subsequent tar collection equipment. The noncondensible gas is then passed to waste or used as a fuel to provide heat for the carbonization process. In some units a portion of this gas provides the fluidizing gas for the reactor.

As might be expected, there are a number of problems associated with the use of the fluidized technique. The major one would appear to be that due to entrainment of fine particles of char in the gas stream leaving the reactor. All pilot plants have installed equipment designed to handle this dust, but none have been completely successful. A second problem lies in the choice of methods used to provide heat for the process; of the many alternative systems none is without some major disadvantage. The possible methods of providing heat and the dust entrainment problem are discussed at some length in appendices 3 and 4 of this report.

A further limitation of the fluidized system is that the coal feed to the reactor must be nonagglomerating. This presents no difficulty when noncoking coals are used, but in the case of coking coals, some pretreatment of the coal, or modification of the process, must be adopted to satisfy this condition.

EXPERIMENTAL PROGRAM

The fluidized low-temperature carbonization program undertaken by the Research Council of Alberta, may be conveniently divided into three sections corresponding to three successive pilot plants. The first of these was built and operated by C. G. Sinclair in a co-operative program with the Department of Chemical Engineering of the University of Alberta. As this section of the work is fully described in Sinclair's M.Sc. thesis ¹⁷, it will only be very briefly mentioned in this report. The second and third pilot plants represent distinct stages in the modification of the original unit towards more satisfactory performance. The third unit gave reasonably satisfactory continuous operation when carbonizing subbituminous coal from the Edmonton area.

First Pilot Plant

A flow sheet of the first pilot plant is shown in figure 2, and details of the reactor in figure 3. The unit was designed to carbonize 60 lbs./hr. of noncoking, 1/16" x 0 subbituminous coal "as received", i.e. containing about 18 per cent moisture. One of its more interesting features was the heating method, which employed an external hot fluidized bed of sand to supply heat to the reactor. The coal charge was fluidized by recirculated carbonization gases.

Coal feed rates of 42 lbs./hr. were achieved, and steady operating conditions were obtained for periods up to two hours, the carbonization temperature being 600° - 650°C. Blocking of the products transfer line, and of the condensers due to coal dust entrained in the gas stream leaving the reactor, limited the length of each test to about 6 hours including 2 1/2 to 3 hours start-up time.

Valuable experience was gained from this unit, particularly as regards many of the operating difficulties inherent in fluidized low-temperature carbonization processes. The fluidized heater showed some promise as a general method of external heating although anticipated difficulties of scale-up caused it to be dropped from the later pilot plants.

Second Pilot Plant

A simplified flow diagram of the original version of the second pilot plant is presented in figure 4 and details of the reactor appear in figure 5. A completely new reactor was designed for the second unit. Heating was entirely internal by means of partial combustion of the char in the air used to fluidize the coal charge. This heating method was thought to be simpler to scale-up than an external heating system. It was also anticipated that it would be easier to operate than the system used in the first pilot plant.

The conical shape of the reactor arose from two considerations, both requiring an increasing cross-sectional area at higher levels of the reactor. Firstly, since gas is generated within the reactor, an increasing cross section is necessary to

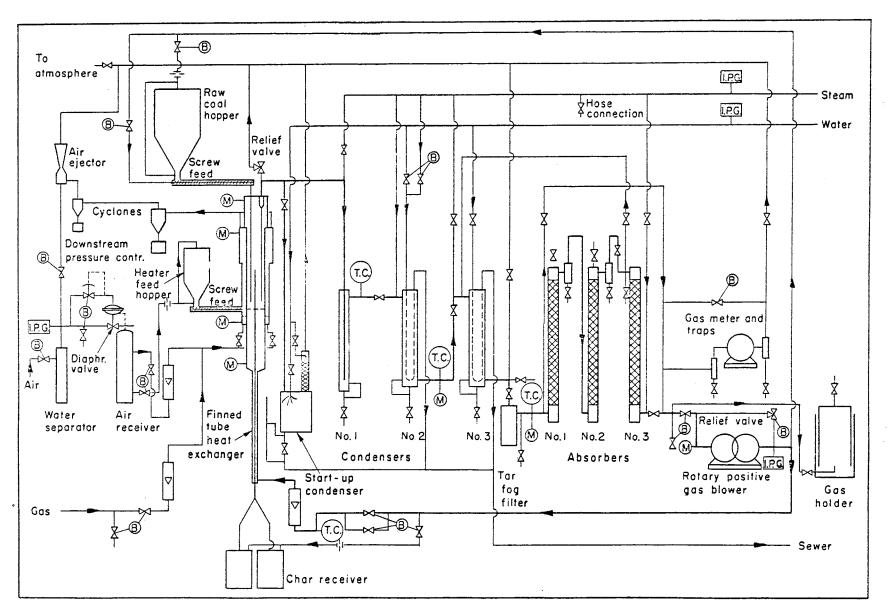


FIGURE 2. Flow sheet for first pilot plant

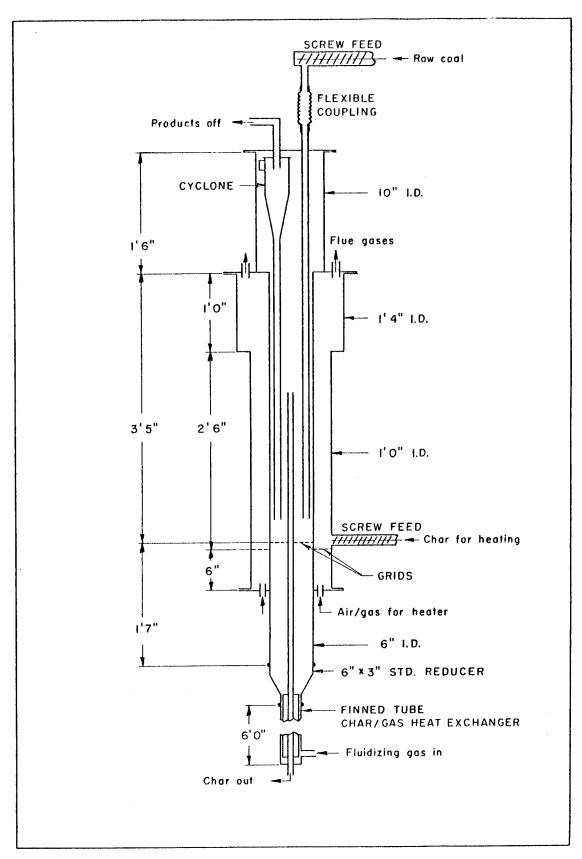
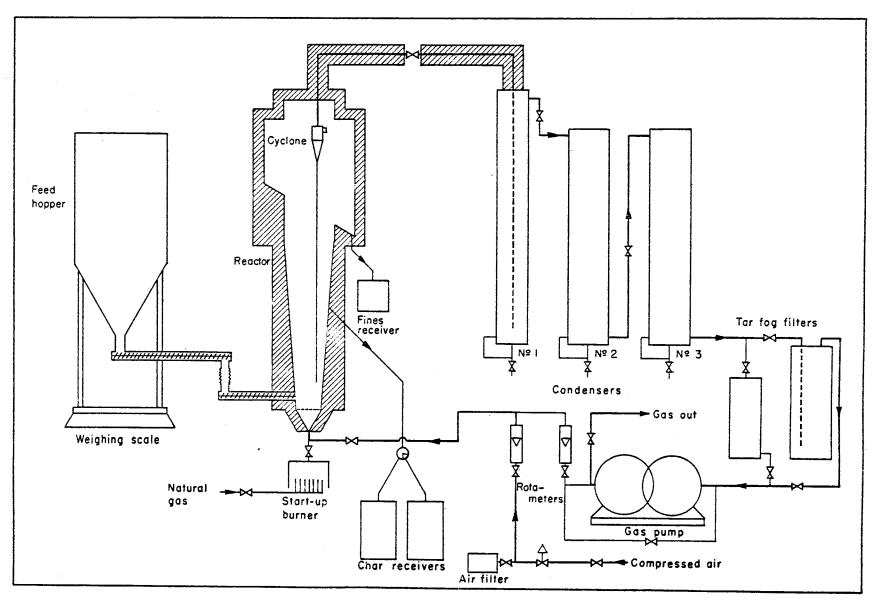


FIGURE 3. Reactor and heater for first pilot plant



9

FIGURE 4. Flow sheet for second pilot plant (original form)

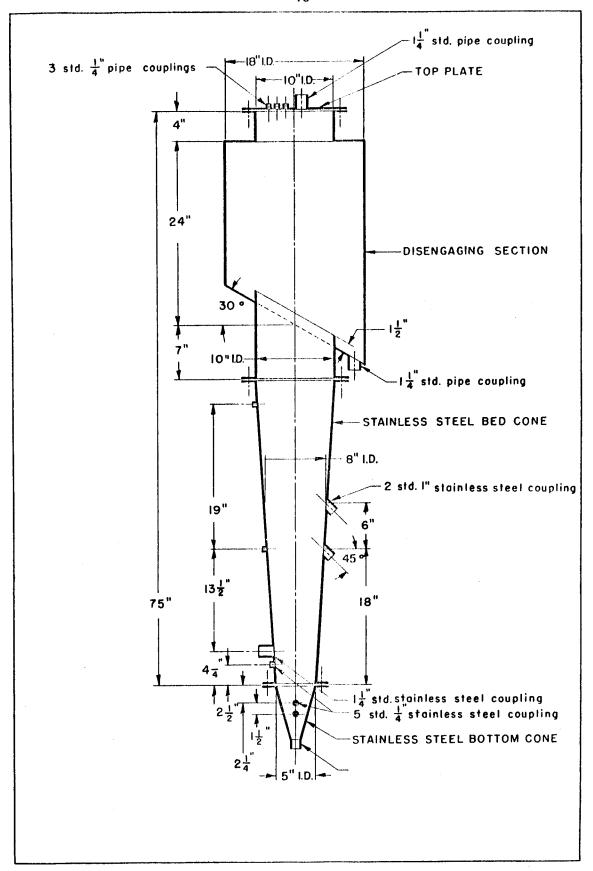


FIGURE 5. Reactor for second pilot plant

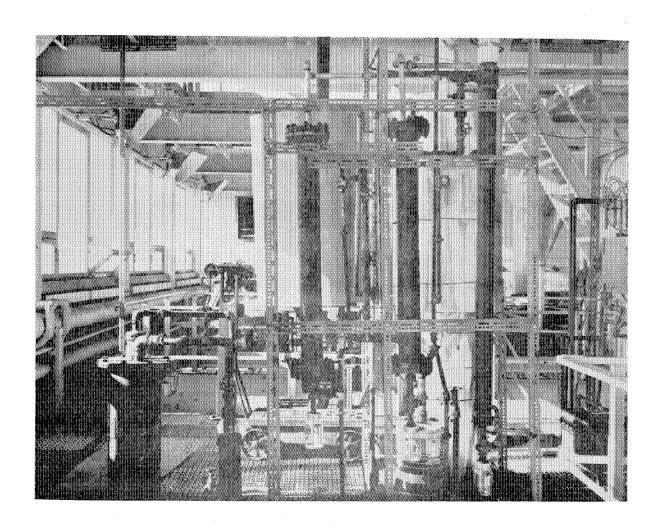


Plate 1. Second Pilot Plant.

In the front of the picture are condensers 1, 2, and 3 running from right to left; the two tar-fog filters are situated to the left of, and below, the third condenser in the lower left corner of the picture. Just below, and slightly to the right of the second condenser is the bin receiving the dust from the disengaging space; the char outlet pipe from the reactor can be seen going down through the floor just to the right of this bin. The product transfer line from the reactor to the first condenser is visible at the top of the picture. The rear of the panel board is at the extreme right.

avoid higher velocities at higher levels of the reactor. Secondly, it was felt that a reactor having higher gas velocities at lower bed levels would be more suitable for fluidizing material having a wide size range. Too large a cone angle, on the other hand, would probably result in poor gas flow distribution. The cone used was a compromise result of these three factors.

The condenser train was the same as was used in the first pilot plant. Except during start-up, the unit was adjusted so that the reactor was at atmospheric pressure. Several short runs, each of about 8 hours duration, were carried out, during which about 420 lbs. of 1/16" x 0 subbituminous coal from the Edmonton area, containing about 15 per cent moisture, were carbonized at 500 - 550°C. The following observations were made during these tests:

- 1. The reactor temperature was found to be easily controlled. It was very sensitive to changes in the air rate, and in coal feed rate. There was no difficulty in maintaining the bed-level within the reactor.
- 2. A throughput of about 10 lbs./hr. was obtained. This was limited by the maximum amount of air which could be fed to the reactor (3.5 standard cu.ft./min.) without causing excessive dust entrainment.
- 3. No tar was collected during these tests. The only product from the condensers was a malodorous aqueous liquor.
- 4. A considerable amount of fine char was carried over from the reactor, and collected in the transfer line to the first and second condensers, and in the condensers themselves. This dust was associated with small amounts of tar, forming a thick immobile paste which caused increases in the pressure drop across this section, and sometimes led to complete blocking.
- 5. A layer of sintered ash was found to have built up to a level of between 3 and 4 inches on the grid at the base of the reactor. A similar observation has been reported by Peytavy and Foch 10.

Third Pilot Plant

During the course of the subsequent testing program a number of modifications were made to the pilot plant at different times. The plant in its final form is shown in the simplified flow diagram presented in figure 6. The reactor and coal feed system, which had given satisfactory performance in the previous tests, were left essentially unchanged. The major differences between this plant and the previous one were in the dust removal, and tar condensation and collection equipment.

Two tests at 600°C. using predried Edmonton coal were carried out with this unit. In each case the plant ran very smoothly and did not have to be shut down until the supply of coal was exhausted – after about 30 hours of operation.

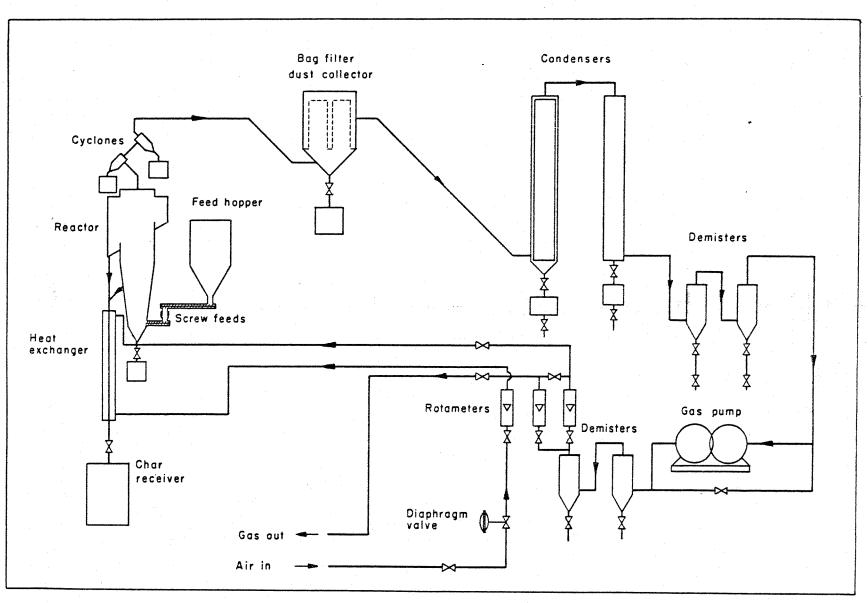


FIGURE 6. Flow sheet for third pilot plant

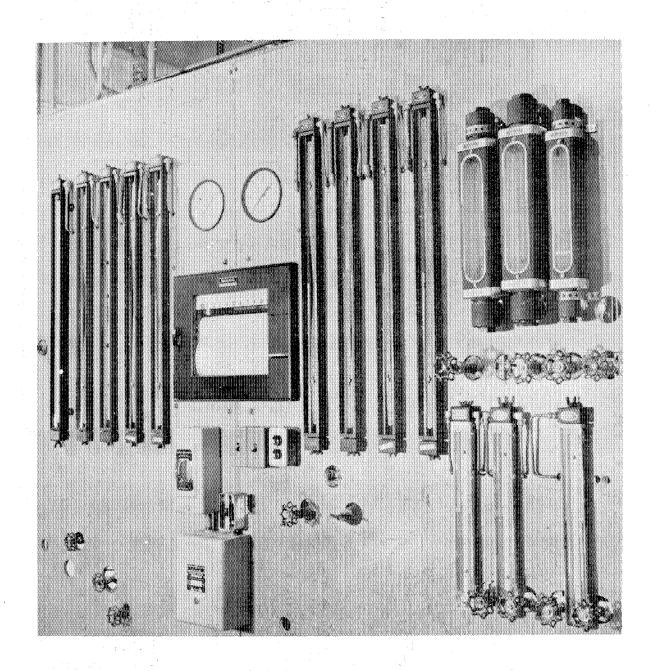


Plate 2. Panel Board for Third Pilot Plant.

This includes the inlet air and outlet gas rotameters, the 12 manometers, and the temperature recorder.

The yield of tar was still found to be abnormally low -- only about 1.5 per cent of the dry coal feed. Laboratory assays³ indicated that a tar yield of 3.7 per cent might be expected when carbonizing this coal at 600°C. No conclusive explanation for the lower yield can be offered, but it was possibly due to combustion of some tar within the reactor or to cracking of the tar before it was condensed. As a result of the laboratory assay tests³ which indicated that, of the readily available noncoking Alberta coals, those from the Lethbridge area gave the highest tar yields (ca. 7.5%); tests were continued using Lethbridge coal prepared by grinding and drying.

In all, four tests were carried out using Lethbridge coal, but in each case the plant had to be shut down shortly after starting the coal feed due to continued severe blocking of the reactor gas outlet, the cyclones, and the connecting pipe work.

Further details of the tests carried out using the third pilot plant are described in a later section.

DISCUSSION

Experimental Results

The test program showed that the conical reactor, heated internally by partial combustion of the charge in the fluidizing air, performed very well as regards solids flow and ease of temperature control. Apart from some possible trouble in establishing and maintaining suitable flow patterns of coal and air, no difficulty is envisaged in scaling-up such a reactor. This is the big advantage of using an internal heating method. This type of reactor should be suitable for the production of low-temperature char (sized $1/8" \times 0$, or smaller) from noncoking coals if the off gases could be wasted or used directly as fuel. This process would be quite feasible for a market in which a premium price could be obtained for such a char.

The dust entrainment problem, which is discussed in more detail in a later section, remained unsolved at the end of the test program. The hot bag filter showed some promise but it is doubtful if such a unit would be suitable in larger scale work.

The difficulty in separating the tar mist from the gas stream which persisted throughout the experimental work, should be relatively easily overcome in a larger scale unit. However, the problem of the thermal cracking of the tar might be more troublesome. There was some slight evidence of tar cracking during the tests with Edmonton coal, while with Lethbridge coal there appeared to be a considerable amount of cracking. Thermal cracking will reduce the tar yield and the carbon formed may cause blockages. To minimize cracking, the tar vapors should be cooled as soon as possible; on the other hand the tars must be kept above their dew point until the entrained dust has been separated.

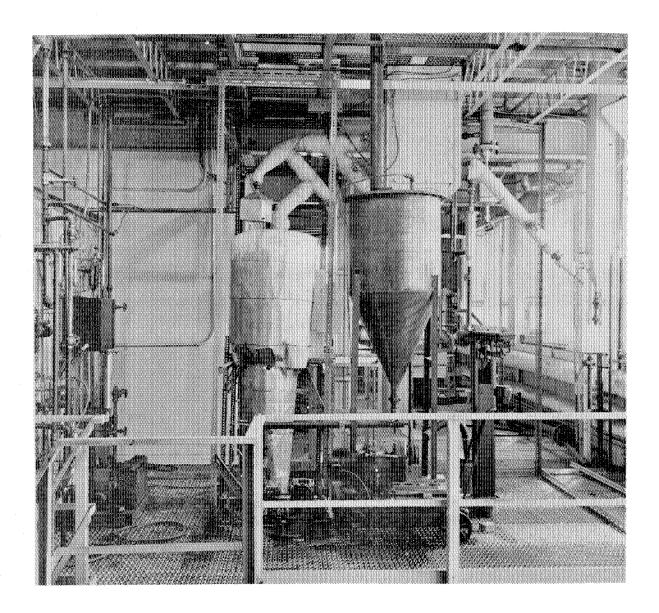


Plate 3. Third Pilot Plant (coal feed side).

In the centre are the coal feed hopper (carried on the platform scale), and the insulated reactor. Immediately above the reactor are the two insulated heated cyclones with their respective dust receivers. The rear of the bag filter unit can be seen above and behind the coal feed hopper, while the tar condenser is in the upper extreme right of the picture. The rear of the panel board lies on the left, the temperature recording box, the third demister, and the gas pump being clearly visible. The outlet gas pipe runs along the top of the picture, and the gas sampling points can be seen leaving this line just at the top of the panel board.

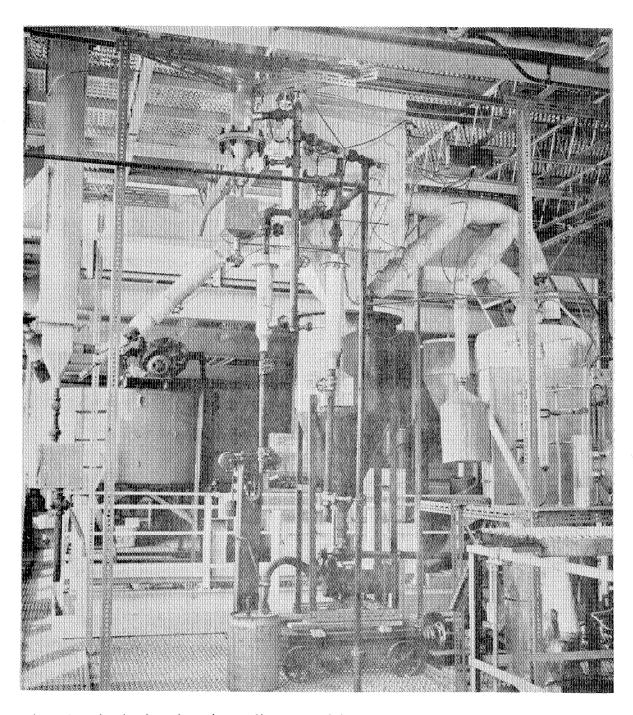


Plate 4. Third Pilot Plant (tar collection side).

The reactor is at the right, and in the lower right corner the top of the char inlet-air finned tube heat exchanger can be seen. The gas sampling equipment is on the shelf in front of the reactor. The front of the bag filter unit, and the first two demisters are in the upper centre of the view, while below and behind these is the coal feed hopper. The first tar condenser and receiver are on the extreme left of the picture, and the bottom of the second condenser and its receiver can be seen to the left of the bag filter. The two screw feeders, their drive units, and the connecting bellows are also visible.

Economics of Low-Temperature Carbonization

The economics of low-temperature carbonization can vary considerably from one case to another, and each situation should be considered separately.

The main product of the carbonization is the solid char, and the price obtainable for the char is a major factor in the economic assessment of the process. The only known commercially successful units have depended upon obtaining a premium price for the char.

Most of the recent investigations have visualized a situation in which the char would be used as an alternative fuel to coal in a thermal power plant. In this case the char would have little, if any, premium price over coal (on a heat value basis), and the cost of processing together with the cost of the loss of heat in the solid fuel would have to be recovered by sale of the tar. The loss of heat involved would be 20 - 25 per cent of the heating value of the coal. Parry ¹⁶ has presented general formulas showing the relationship between cost and value of coal, char, and tar for cases where the char is used as a power plant fuel.

It is not possible at present to assess accurately processing costs for low-temperature carbonization. The cost should decrease as the throughput of the operation is increased, and lower priced coals will have lower operating costs. Minet ¹² has suggested that processing costs of between \$0.50 and \$1.50 per ton of coal should be attainable.

As the operation depends for its success upon the sale of the tar, the yield and value of the tar are two important factors. The yield of tar will depend primarily upon the type of coal carbonized, and to a lesser extent upon the carbonization process. Tar yields can range up to 20 per cent by weight of coal for high volatile bituminous coals. Alberta subbituminous coals give tar yields of about 4 per cent as estimated by laboratory assaying³.

The value of the tar is not so easily assessed, and its poor acceptance has been very disappointing to a number of potential producers. There appears to be no satisfactory process for refining low-temperature tar into valuable fractions. The recent literature includes a number of reports of experimental investigations of the chemical composition, quality, possible refining methods, and uses for the tar⁵, 18-22, but as yet no large volume outlets for the tar have been found.

A recent joint study by the Saskatchewan Research Council and the U.S. Bureau of Mines²³ estimated that for a low-temperature carbonization unit operating on lignite from the Estevan area, the tar would have to be sold for at least 20 cents a gallon to pay for the carbonization process and the loss of potential heat. Other specific cases have been analyzed by Minet ¹², ²⁴ and Pursglove²⁵. The latter project is now reported to have been shelved because of unattractive economics²⁶.

The comparatively low yields of tar obtainable from Alberta subbituminous coal, and the probable lack of local markets for the tar, together with the disappointing reports from other parts of the world have led to the conclusion that there is no prospect of setting up a commercially successful low-temperature carbonization unit, working in conjunction with a coal-fired thermal power plant, within Alberta in the foreseeable future.

Fluidized low-temperature carbonization could however prove to be a very attractive process for the production of low-temperature char if a demand for the char at a premium price should arise.

CONCLUSIONS

- 1. The pilot plant, in its final form, saccessfully carbonized predried Edmonton coal at 600°C., in the fluidized state.
- 2. The conical reactor operated very smoothly. Both temperature and flow of solids were easily controlled.
- 3. The tar yield obtained was very low, being less than 2 per cent of the dry coal carbonized.
- 4. Char dust entrained in the gases leaving the reactor is difficult to separate completely from the gas stream. This dust gives rise to blockages if not removed before the vapors cool below their dew point.
- 5. Thermal cracking of the tar occurs at temperatures above 350°C., leading to reduced tar yields and to blockages.
- 6. Condensed tar mist which was difficult to separate from the gas stream, tended to foul equipment installed downstream from the condensers.
- Low-temperature carbonization is not economically attractive in Alberta at present. The position might be reversed if an outlet for char, at a premium price, is found.

ACKNOWLEDGMENTS

The authors are indebted to Mr. E. Kyto who assisted in constructing and operating the unit, and who prepared the drawings for this report.

Acknowledgment is also due to Mr. J. F. Fryer for coal, char, and gas analyses; to Dr. R. M. Elofson for tar analyses; and to these and other members of the Research Council of Alberta staff for their helpful suggestions and for their cooperation in operating the plant during the longer tests when shift work was necessary.

REFERENCES

- Gregory, J., and McCulloch, A.: Ind. Eng. Chem. 41, 1003-1011 (1949).
- 2. Parry, V. F., Landers, W. S., Wagner, E. O., Goodman, J. B., and Lammers, T. C.: U.S. Bur. Mines Rept. Investig. 4954, 43 pages (1953).
- 3. Davies, G., Moreland, C., and Berkowitz, N.: A low-temperature carbonization assay of Alberta coals, Res. Coun. Alberta Inform. Ser. No. 34, 29 pages (1961).
- 4. Soule, R. P.: Proc. Third Int. Conf. on Bit. Coal, Nov. 1931, p. 272-298.
 Pittsburgh: Carnegie Inst. Tech.
- 5. Bristow, W. A.: J. Inst. Fuel 20, p. 109-130 (1947).
- 6. Lesher, C. E.: Min. Eng. 4, 287-299 (1952).
- 7. Heptinstall, W. G.: Bull. Can. Inst. Min. Met., No. 210, p. 1187-1196 (1929).
- 8. Parry, V. F., Landers, W. F., and Wagner, E. O.: Min. Eng. <u>8</u>, p. 54–64 (1956).
- Coppens, L., Venter, J., and Ledent, P.: Inst. Natl. d'Ind. Charbonniere (Belgium) Bull. Tech. Houille et Derives, No. 11, p. 336-370 (1957).
- 10. Peytavy, A., and Foch, P.: A study of semi-carbonization by means of fluidization; Conf. on Chem. Eng. in the Coal Ind., Stoke Orchard, June 1956, p. 75-99. London: Pergamon Press.
- 11. Jenkins, G. I.: Chem. and Ind., No. 35, 1068-1076 (1955).
- 12. Minet, R. G.: Coal Utilization, Feb., p. 33-36 (1956).
- 13. Asai, K., and Tanno, H.: Ann. des Mines de Belgique, Nov., p. 971-980 (1955).
- 14. Shrikhande, K. Y., Das Gupta, A. K., and Lahiri, L.: J. Sci. and Ind. Res. (India) 13B, p. 634–641 (1954).
- 15. Lang, E. W., Smith, M. G., and Bordenca, C.: Ind. Eng. Chem. <u>49</u>, p. 355-359 (1957).
- 16. Parry, V. F.: U.S. Bur. Mines Rept. Investig. 5123, 27 pages (1955).
- 17. Sinclair, C. G.: unpublished M.Sc. thesis, Univ. of Alberta, 1956.

- 18. Gomez, M., Goodman, J. B., and Parry, V. F.: U.S. Bur. Mines Rept. Investig. 5302, 44 pages (1957).
- 19. Watson, G. H., and Williams, A. F.: The study of tars obtained in fluidized carbonization; Conf. on Chem. Eng. in the Coal Ind., Stoke Orchard, June 1956, p. 100–121. London: Pergamon Press.
- Sabatier, J. L.: Industrial treatment of low-temperature carbonization tars;
 Conf. on Chem. Eng. in the Coal Ind., Stoke Orchard, June 1956,
 p. 122–131. London, Pergamon Press.
- 21. Anon.: Chem. Eng. 67, No. 7, p. 70-72 (1960).
- 22. Sask. Res. Coun. Twelfth Ann. Rept., p. 26 (1958).
- 23. Sask. Res. Coun. Twelfth Ann. Rept., p. 60 (1958).
- 24. Minet, R. C., Smith, H. B., and Trilling, C. A.: Chem. Eng. Progr. 50, p. 342-347 (1954).
- 25. Pursglove, J.: Coal Age 62, p. 70-73 (1957).
- 26. Anon.: Chem. Eng. 67, No. 8, p. 156 (1960).
- 27. Minet, R. G.: Operation of a low-temperature carbonization pilot plant; presented at Am. Gas Assoc. Chem. Eng. and Mfd. Gas Conf., New York, May 1955.

APPENDIX 1. Pilot Plant Design

This section outlines the main considerations of the various parts of the pilot plant, and lists many of the modifications made periodically. For clarity, the equipment is discussed in the order in which the various sections appear along the flow diagram of the plant.

(a) Choice of Coal

Initially, the choice of the coal to be used in the pilot plant tests was arbitrary. A typical Alberta subbituminous coal was expected to give the most useful results, and for convenience, local Edmonton coal was chosen. Lethbridge coal was used in later tests because of its greater potential tar yield.

The coal size chosen was 1/16" x 0; a typical size analysis is given in table 3 (page 28). It was felt that the size range used should be a top size to zero because of the difficulty of preparing, on a large scale, a coal fraction containing no fines. A top size of 1/16" was found by Sinclair 17 to be the maximum for reasonably smooth fluidization without excessive entrainment of fines. The use of a smaller top size would involve higher grinding costs and smaller throughputs in an internally heated carbonizer where the capacity is limited by the air-velocity.

Initial tests were carried out on undried coal containing about 18 per cent moisture. This moisture imposed high heat loads on the reactor with a consequent low throughput. Evaporation of moisture followed by condensation in the coal feed system caused sporadic plugging of the coal feed lines. To overcome these difficulties, the coal was predried to about 3 per cent moisture in all subsequent tests. Lack of more suitable drying equipment led to the coal being dried by heating it in a warm (70 - 80°C.) oven for 2 1/2 days. This treatment allowed some preoxidation of the coal which reduced its potential tar yield.

(b) Coal Feed System

A large scale unit would undoubtedly use pneumatic transport, or fluidized transport, of the coal and char. However, these systems cannot be conveniently scaled down, and therefore a screw feed system was used throughout the pilot plant program.

The coal feed system used in the first pilot plant was very difficult to control, and caused large fluctuations in the temperature and in the bed level in the reactor. The difficulty was due to two factors; firstly the purge gas fed into the top of the coal feed hopper would sometimes blow the coal past the screw feed into the reactor, and secondly there was no positive means of feeding the coal down the vertical section of the coal feed pipe beyond the end of the screw feed. This section frequently blocked due to condensation of water or tar on the coal in the cooler end of the pipe.

In the later pilot plants these difficulties were overcome by having only one purge gas line which entered the feed system at four points. This balanced the gas pressure throughout the feed system. The design of the new reactor made it a simple matter to arrange the screw feed to reach the end of the coal inlet line. This system gave satisfactory performance and no further modifications were made.

(c) Reactor

Two different reactors were used during the course of this work; these are shown in figures 3 and 5. The first design was rejected primarily because of scale-up difficulties which would be involved in this type of reactor. No serious attempts were made to modify the design to improve the solid flow rates which were very irregular and caused large temperature fluctuations.

The second reactor was found to be very easy to control both as regards to temperature and solid flow rates, and no further major alterations were made to the original design. The grid, which was initially installed just below the coal feed inlet, was removed when it was found that it gave rise to sintered ash building up on the grid. After removing the grid the bed was not as easily fluidized at the commencement of a test, but no further build up of ash was encountered. An inlet-air preheater was installed on the reactor to recover some of the sensible heat from the outgoing char.

No investigation of the reactions taking place within the reactor was carried out, but the low tar yields obtained suggest that possibly some of the tar was being oxidized. In any future work consideration should be given to modifying the reactor with a view to reducing the possibility of tar oxidation, possibly by altering the positions of the air and coal inlets. It might also be advantageous to reduce the size of the disengaging space; this would reduce the residence time of the gases within the reactor, and consequently reduce the probability of cracking the tar vapors, and reduce heat losses. The resulting increase in the amount of entrained solids in the gases leaving the reactor would probably not be very serious, and by the time the gases had been through the cyclones the amount of dust still entrained should not be much different than was the case with the large disengaging space.

(d) Dust Separation

Char dust entrained in the gas stream leaving the reactor caused the major operating difficulty associated with this pilot plant investigation. The general problem of dust entrainment and collection is discussed in appendix 4, while our experience is outlined here.

The first pilot plant relied upon a relatively small disengaging space and one 3-inch diameter internal cyclone for dust removal. This proved quite inadequate, and a change from a 3-inch to a 2-inch diameter cyclone did not show any improvement. The lines between the reactor and the condensers, and the condensers themselves, continually blocked with entrained dust. This blocking severely limited the length of time of each test.

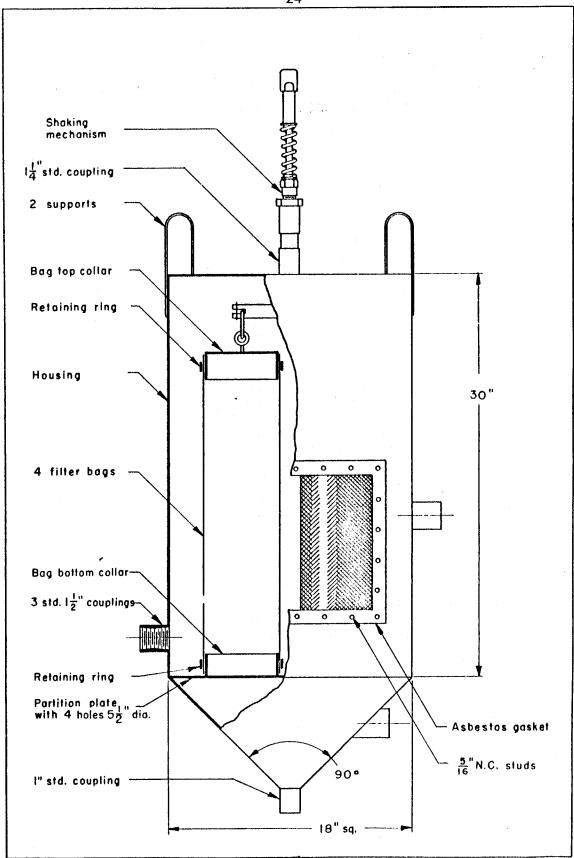


FIGURE 7. Glass-wool bag filter unit

The second pilot plant had a much larger disengaging space together with one 3-inch diameter internal cyclone. Dust carry-over was found to be less than in the first pilot plant, but still was severe enough to cause blocking of the reactor outlet line, and of the line between the first and second condensers. The first condenser was also found to contain a large amount of dust on cleaning out between tests. In later tests using this plant the internal cyclone was removed, and two 3-inch diameter, externally heated cyclones were installed in series between the reactor and the first condenser. These resulted in a considerable reduction in the amount of dust entrained in the gas stream entering the condensers.

It has been estimated that these cyclones removed about 94 per cent of the coal dust which was entrained in the gases leaving the reactor. The amount of dust getting past the cyclones amounted to 0.3 per cent of the dry coal fed to the reactor (a similar figure has been published by Parry⁸). This small amount of dust, associated with the low tar yields obtained, was sufficient to form a thick, highly viscous paste which eventually blocked the condenser lines.

In order to remove these last traces of dust, an externally heated bag filter was included immediately downstream from the cyclones, and the pipe work was reassembled to be as short and straight as possible, in order to minimize the number of locations where dust might collect. Using the bag filter, continuous operation of the pilot plant was achieved with Edmonton coal. The filter unit is described in detail in the following section.

Bag Filter Design - As we had no experience of operating a glass bag filter unit at elevated temperatures, nor was there any information available on the subject, the design both as regards filter area and heat input to the unit was quite conservative.

Results of previous tests on the pilot plant indicated that the gas flow rate at the filter unit, at 350°C. and atmospheric pressure, would be of the order of 15 cu. ft./min. Having decided to use a filtration rate of about 1.5 cu.ft./sq.ft./min., a filter unit having about 10 sq.ft. of filter cloth area was designed.

The filter cloth chosen was type G-202-C, manufactured by the National Filter Media Corporation of Salt Lake City; this cloth had been heat treated followed by silicone lubrication. Unfortunately at the time of building the unit this cloth was not available, so type G-202 was used. This was a similar weight and weave of material, but had not had the heat and silicone treatment. Having decided the filter area to be used, and bearing in mind the sizes of electrical heaters readily available, the desirability of using fairly small individual filter bags and of keeping the whole unit within reasonable dimensions, and the necessity of providing a bag shaking mechanism, the unit shown in figure 7 was finally designed.

The bag housing is square in cross section, and divided into two compartments by a partition plate. The lower compartment is in the form of an inverted square

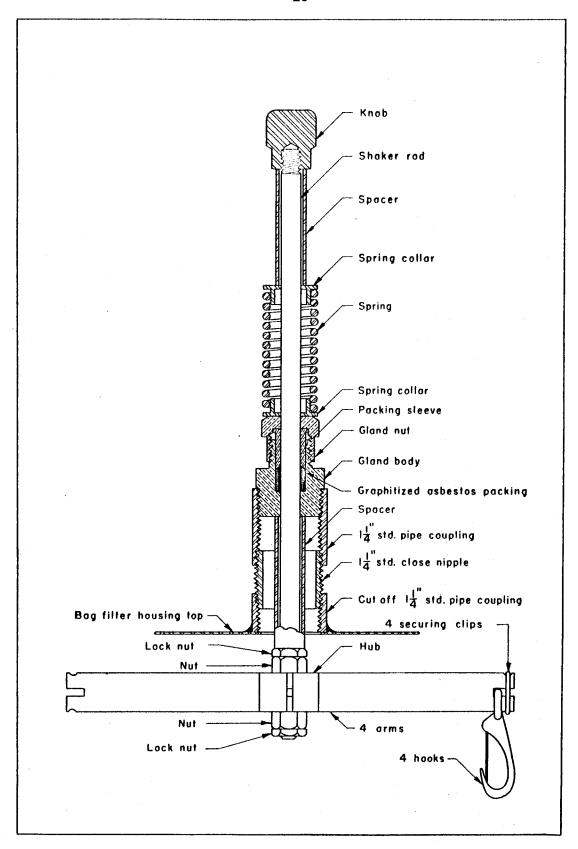


FIGURE 8. Shaking mechanism for glass-wool bag filter unit

pyramid and carries fittings for the gas inlet, and dust outlet to the dust hopper. The upper compartment which contains the bags, has fittings for the clean gas outlet, and a gas inlet fitting which is only used if the gases are to bypass the bags in case of a breakdown of the unit or while the bags are shaken. There is a fitting for the shaking mechanism at the centre of the roof. The front wall has a doorway (10 in. x 12 in.) to allow access to the bags.

The partition plate has four 5 1/2-inch diameter holes arranged in a symmetrical square pattern. The holes are fitted with collars on the upper side of the plate, over which the lower ends of the bags are clamped. Four filter bags were used, each 5 1/2 in. in diameter and 2 ft. long, giving a total filter area of just over 10 sq. ft. The upper ends of the bags were closed off by clamping them around the collars of four blind cups which hung from the ends of the arms of the shaking mechanism.

The shaking mechanism is shown in detail in figure 8; its operation was manual and is self-explanatory. Some difficulty was experienced in finding a suitable packing material as the gland operated at too high a temperature for the majority of packings. Even the graphitized asbestos used became very hard after several hours' use, and had to be replaced between each test.

The whole of the filter housing is insulated with fitted sheets of 1 1/2 in. thick 'Marinite-23', a bonded asbestos fibre material, manufactured by Johns-Manville Co., Ltd. Electrical strip heaters are inset into the insulation sheets so that the surface of the strip heaters is flush with the inner surface of the insulation. Specifications of the Marinite indicated that the expected heat loss with the filter unit operating at 350°C. was about 220 B.t.u. per hr. per sq.ft. This would require a heating load of 65 watts per sq.ft. to balance the heat loss. Using a conservative design, the heater circuits were arranged to give about 85 watts per sq.ft. of housing area. The arrangement of the heaters, and a circuit diagram are shown in figure 9.

Table 1 gives the wattage supplied to the different sections of the filter housing, together with their areas, showing the wattage/sq.ft. for each section. The designation symbols in the second column show the manufacturer's (Canadian Chromalox Co. Ltd.) number for the various elements.

Comparison of the traces of thermocouples 7 and 8 in figures 15 and 17 show that the change in temperature of the gases in passing through the bag filter was very small indicating that the electrical heating was just balancing the heat loss, as designed.

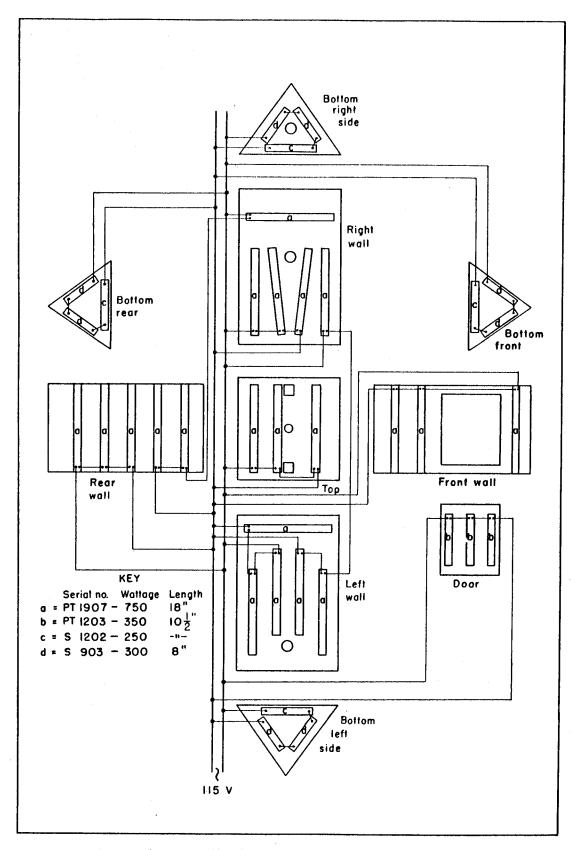


FIGURE 9. Insulation walls, heater arrangement, and circuit diagram for bag filter unit

Table 1. Heat input per unit area used for bag filter housing

Section	Elements	Wattage	Internal area of section, sq.ft.	Watts/sq.ft.
Тор	3 × PT 1203	192	2.25	85.3
Rear wall	5 × PT 1907	320	3.75	85.3
Right wall	п	11		n ·
Left wall	u ·	: 11	u u	u,
Front	3 × PT 1907	192	2.92	65.9
Door	3 × PT 1203	116.5	.83	140.0
Front and door combined	3 × PT 1907 &3 × PT 1203	308.5	3.75	82.3
Lower triangular walls (each)	2 x S 903 1 x S 1202	4 × 93.8	4 × 0.8	117
Total		1835.7	20.45	89.7

(e) Cooling and Condensation

The same condenser train was used for both the first and second pilot plants. It consisted of three condensers connected in series. The first of these was air-cooled, with an external area of about 6 sq.ft.; this was followed by two water-cooled trombone condensers, each having 20 ft. of 1/2-inch copper tubing.

This condensing system, although giving sufficient cooling, was found to be unsuitable due to a tendency to block with accumulations of a sticky paste of water,

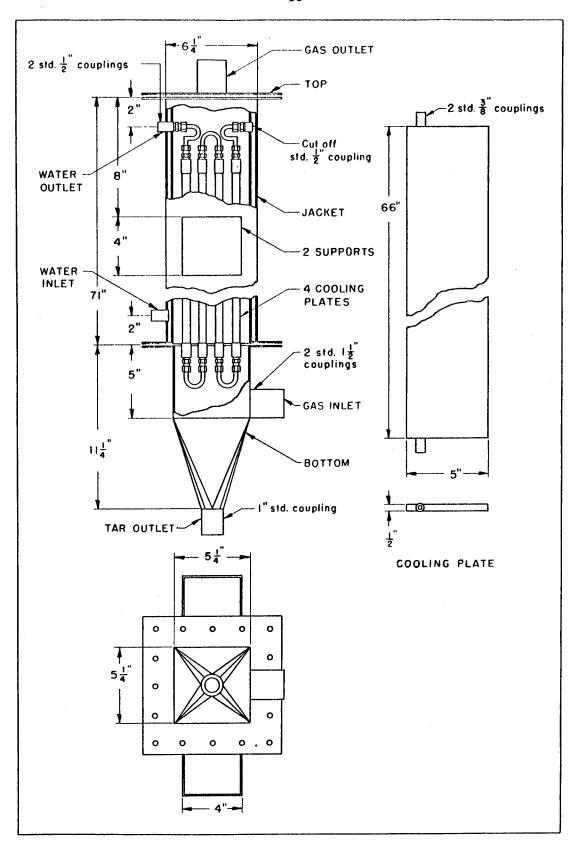


FIGURE 10. Tar condenser

tar, and char dust which would not drain down into the condensate receivers. These accumulations were particularly bad on the lower end plates of the condensers, in the lower U-bends of the water tubes, and in the interconnecting pipe work.

As a result, a new condensing system was installed in the third pilot plant, consisting of a vertical plate, water-cooled main condenser (described below) followed by one of the previous trombone condensers modified to prevent accumulation of condensate on the lower end plate. This system performed reasonably well, and no further modifications were made.

Condenser Design - The requirements for the heat exchanger were that it should cool the gas stream entering at about 325°C., to 35° - 40°C., that it should be easy to clean, and if possible that it should be self-cleaning to some extent. Also its general dimensions should be such as to fit in with the layout of the rest of the pilot plant.

Previous tests had indicated that the rate of flow of hot gas was 44 lbs. per hr. uncondensibles and 3.7 lbs. per hr. of condensibles, mostly steam. This gives a heat load for the heat exchanger of 12,000 B.t.u. per hr. Designing for 15,000 B.t.u. per hr. and using an over-all heat ransfer coefficient of 5 B.t.u. per hr. sq.ft. °F., a cooling area of 24 sq.ft. is necessary.

A plate condenser, containing four long vertical plates was constructed; this is shown in figure 10. The area of the plates together with the area of the cooling jacket gives a total cooling area of about 28 sq.ft. The plates are loosely supported within the condenser so that they can be taken out for cleaning and their walls being plane areas they should be easily cleaned.

The gases enter the heat exchanger at the lower end and flow upward inside the condenser, thus the lighter condensible fraction condensing higher up in the unit should run down the plates and walls, thus cleaning them of any heavier tars deposited in the lower sections. Except for the connecting pipes between the plates all walls of the unit are vertical with no projections where material might accumulate. A number of strengthening studs were welded to both walls of each plate to prevent them bulging under the cooling water pressure.

This condenser performed reasonably well, cooling the gases to below room temperature, and it gave no signs of blocking. However, it has a number of weak features. The plates are not as easy to remove as was planned, and in fact have never been removed since installation, but the unit was steamed out between tests. There is no way of completely removing the air from inside the two plates which are fed with water at the top, there being scarcely sufficient space above the plates to install air bleeds. However, as the unit was giving satisfactory cooling no further attempts to remove this entrapped air have been made. Since the outer cooling jacket is not made of heavy enough metal and has no strengthening supports, it will only withstand pressures up to about 18 p.s.i. The cooling water pressure at the

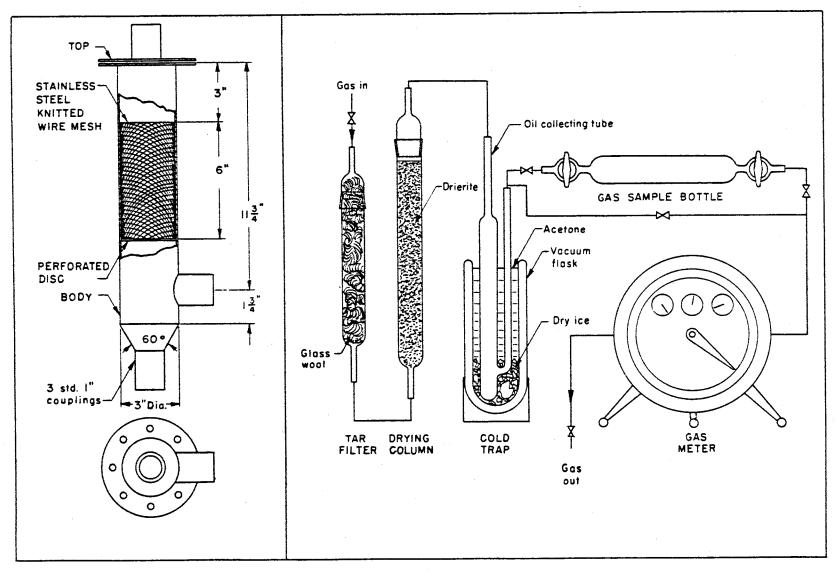


FIGURE 11. Demister

FIGURE 12. Gas sampling apparatus

inlet was therefore not allowed to exceed 11 p.s.i. This meant a lower flow rate of cooling water than had been intended. Nevertheless, sufficient cooling of the gases was achieved.

(f) Tar Mist Collection

One annoying problem which persisted throughout the pilot plant carbonization program was that due to tar mist entrained in the cooled gases down stream from the condensers. The tar mist, if it is not removed, tends to foul any equipment through which it passes - e.g. pump, meters, light oil absorbers.

Tar fog filters were installed in the first two pilot plants; these consisted of chambers, packed with either glass wool or wood shavings, through which the cooled gases were passed. These units were not satisfactory as they did not trap all the tar; their resistance to gas flow increased considerably after a few hours operation, and their tar hold-up made accurate assessments of tar yields and good mass balances impossible.

The wood-shavings type of tar fog filter was dispensed with in the third pilot plant in favor of small knitted wire mesh demisters. The construction of these is shown in figure 11. Initially, two of these units were used, one installed near the condenser outlet, and the other immediately following the gas pump. Tar collection was much better using this system; in fact about half of the total tar collected was obtained from the demister following the gas pump. It was obvious that the rotary action of the pump caused the tar mist to coalesce so that it was fairly easily removed from the gas stream discharged by the pump. The accumulation of tar in the outlet gas rotameter showed that some tar was still getting past the demisters. In later tests. four knitted wire mesh demisters of the type shown in figure 11 were used. Two of these were installed in series downstream from the condenser, one at the pump discharge, and the fourth just prior to the outlet gas rotameter where the gas was cooler, the heat generated by the pump having dissipated. As a result, much less tar was present in the outlet gases, but there was still some tar to be seen in the outlet gas rotameter. Again, more tar was collected from the demisters downstream from the pump, than from the condensers.

The wire mesh demisters were a decided improvement over the wood-shavings unit. They had low resistance to flow which did not increase with time; also their operation was continuous and there was no need to renew the packing periodically. It is not thought that the tar mist problem would be much of an obstacle in a larger unit, as several items of equipment which collect such mists quite effectively are commercially available for large scale operation.

(g) Light Oil

The first pilot plant included a silica-gel adsorption tower for collecting the light oils from the gas stream. However, because of operational difficulties with other sections of the pilot plant the adsorber was never used.

At one stage during the tests with the second pilot plant a carbon adsorber was installed immediately downstream from the tar-fog filter. This did not prove satisfactory as it soon developed a very high resistance to flow, probably due to accumulation of sticky tar mist on the carbon. On steaming out after the test a small amount of oily tar was recovered from the adsorber.

In the third pilot plant it was decided not to attempt to collect all the light oil from the gas stream, but rather to estimate the amount of oil formed by condensing and weighing the light oil contained in a sample of the outlet gases. Arrangements were made whereby a sample stream could be taken off the gas line, metered, dried, and passed through a cold trap to remove the light oil, before returning the uncondensed gas to the main gas stream (see figure 12). Initially, the gas sample was taken from a point between the condensers and the gas pump. This system worked reasonably well and samples of gas and light oil were obtained.

The mist carried in the gas tended to foul the sample inlet line and the drying column. This problem was overcome in a later test by installing extra demisters in the main gas line, and by moving the gas sampling point to a location downstream from the outlet gas rotameter. Some of the light oil solidified in the cold trap, and the solid slowly accumulated until the sample line eventually blocked.

The sample collected indicated that the amount of light oil in the exit gases was very small, amounting to about 0.2 per cent of the weight of dry coal carbonized.

(h) Gas Pump

A Rootes-Connersville blower specified to deliver 9.5 cu.ft. per min. of 0.5 sp.gr. gas at 5.5 lb. per sq.in. above suction pressure was installed in the first pilot plant. This pump proved quite satisfactory in operation and was used in all subsequent work. It was found to be quite successful in coalescing the tar carried along with the gas as a mist. The pump would occasionally jam after cooling down due to the thickening of tar accumulations within the pump; however, it was easily freed by steaming it out for a short while prior to running it.

(i) Gas Metering

The outlet gases were metered using an ordinary household type leather bellows gas meter in the first two pilot plants. This meter was installed in the pump discharge line in the second pilot plant and initially appeared to be quite satisfactory. However, when the pilot plant had been improved to the stage where measurable amounts of tar were being collected, the gas meter slowly fouled up with tar and water and eventually ceased to function altogether. This type of meter was replaced by a rotameter in the third pilot plant, and because of the tar in the outlet gas stream, the gas was only passed through the rotameter for short periods when a

flow rate reading was required. This system was perfectly satisfactory, and the presence of tar in the gases did not impair its performance.

(i) Instrumentation

The third pilot plant was equipped with 12 thermocouples located at appropriate points throughout the system. The 12 temperatures were continuously recorded on a multipoint temperature recorder. The locations of the thermocouples are shown in figure 13, and typical examples of the record charts obtained are shown in figures 15 and 17.

In addition, thermometers were installed in the outlets of the air and gas rotameters, and at the pump discharge.

A total of 12 manometers were used to measure absolute pressures, and pressure drops over different sections of the pilot plant. The locations of the manometer tappings are numbered in figure 13, and the manometers are listed in table 2. A pressure gauge was installed at the discharge of the gas pump.

The air inlet and gas outlet flow rates were measured by rotameters. Coal feed rate was measured by regularly weighing the coal feed hopper which stood on a platform scale. Char production rate was best estimated from the coal feed rate; however, an independent estimate was made by weighing the char receivers each time they were changed when full.

(k) General Comments

Connecting unions were located at frequent intervals along the gas line, in order that any piece of equipment could be easily removed. A steam line was connected at several points in the system; and the condensers, demisters, gas pump, and the line itself were thoroughly steamed out following each test. So far as was practical, all valves, switches and meters were mounted on the control panel.

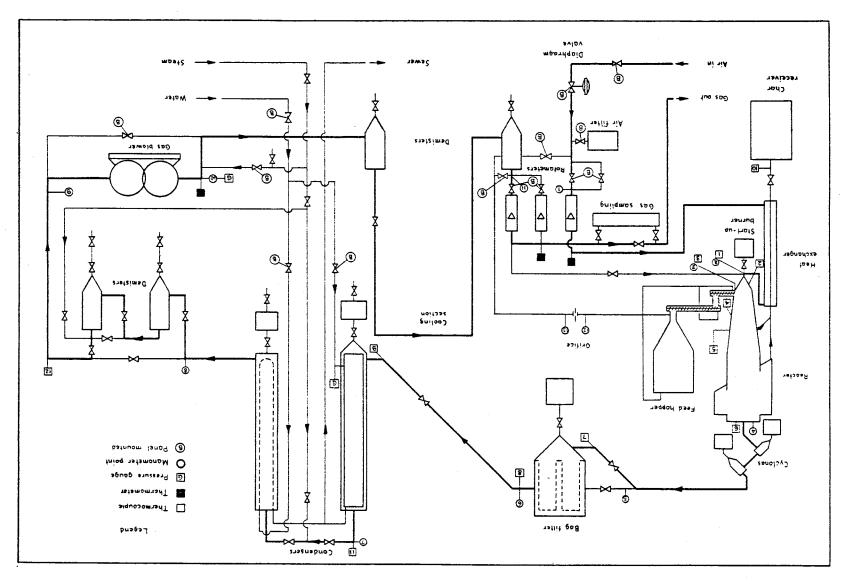


FIGURE 13. Detailed flow sheet for third pilot plant

Table 2. Manometer connections for pilot plant (The figures in the third and fourth columns refer to the manometer points as numbered in figure 13)

Manometer number	Fluid	Tapping high pressure side	Tapping low pressure side	Remarks
1	Hg	1	Atmosphere	Inlet air pressure
2	Hg	2	3	Across lower part of bed
3	H ₂ O	3	4	Across upper part of bed
4	Hg	4	5	Across cyclones
5	Hg	5	6	Across bag filter
6	H ₂ O	6	7	Across condenser 1
7	H ₂ O	7	8	Across condenser 2
8	Hg	8	9	Across 1st two demisters
9	Hg	10	9	Across gas pump
10	H ₂ O		neter – not diagram)	Purge gas to char bins. Not used in later work.
11	H ₂ O	13	12	Air meter to feed system
12	Hg	11	Atmosphere	Outlet gas pressure

APPENDIX 2. Plant Operation and Results

The operation of the third pilot plant was relatively simple, and while the plant was running smoothly only one operator was necessary.

Prior to start-up, about 1,000 lbs. of coal were prepared by crushing to $1/16" \times 0$, and drying to less than 3 per cent moisture. The coal feed hopper was filled with this prepared coal. The cyclone and bag filter heaters were switched on a few hours before start-up.

The gas pump was started with the valve above the start-up burner open, and the air velocity through the reactor, which contained char from the previous test, was increased by throttling the pump bypass valve, until manometers 2 and 3 indicated that the char was smoothly fluidized. At this point the start-up burner was turned on and hot combustion gases were drawn through the bed until it reached a temperature of about 250°C., - at this temperature the char would burn in air. The burner was turned off and air drawn through the bed until it reached the carbonization temperature of 600°C. By enriching with oxygen the air (drawn in through the start-up burner), this temperature could be attained within an hour.

The valve above the start-up burner was now closed while air from the compressed air supply was fed to the reactor via the inlet air rotameter. By adjusting the compressed air valve and the pump bypass valve the pressure at the top of the reactor was set to approximately atmospheric pressure. A small amount of air was bled into the coal feed system through the orifice meter.

The coal feed was started and set at such a rate that the temperature of the bed in the reactor remained steady at 600°C. There was no difficulty in holding the reactor temperature which responded immediately to any changes in coal feed or air feed rates. The whole of the plant reached equilibrium temperature about 6 hours after start-up.

All manometers, rotameters, and thermometers, together with the weight of the coal feed hopper were read and recorded every 30 minutes. The char bins were changed, and weighed as they became full. Condensate was removed from the condenser receivers and tar from the demisters periodically. The cyclone dust receivers were emptied when convenient. The pressure drop across the bag filter was held to between 0.3 and 0.4 in. of mercury by shaking the bags when the pressure drop rose to the latter figure.

Failing a breakdown, the plant was operated smoothly in this manner until the supply of coal feed was exhausted – about 30 hours after start-up.

Results

The more pertinent results of the two prolonged tests carried out with the third pilot plant are outlined below.

Table 3 gives typical size analyses of the coal feed and the char obtained from the main char receiver. It is noticed that the amount of fines in the char is significantly less than in the coal. This reflects the entrainment of the fine material in the gas stream, from which it was recovered in the cyclones and the bag filter.

Table 3. Size analyses of coal feed and char product

Tyler sieve mesh no.	% by weigh Coal feed	it retained Char produc
10	1.6	0.6
10 - 14	4.5	7.3
14 - 20	15.5	18.3
20 - 28	20.9	22.4
28 - 35	14.6	16.0
35 - 48	12.7	10.8
48 - 65	8.0	8.1
65 - 100	6.7	6.2
100 - 150	4.7	3.8
150 - 200	3.5	3.1
200 – 0	7.3	3.4
	100.0	100.0

The two tests were very similar, the main difference being the considerably greater throughput rate used in the second test. In each case the results quoted are for the period during which steady conditions were maintained, and do not include the short periods during start-up and shutdown of the plant.

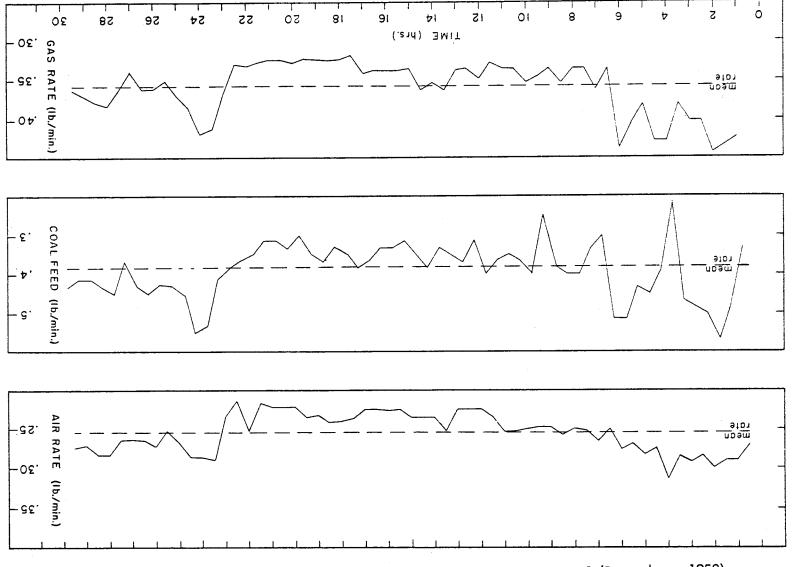


FIGURE 14. Air and coal feed rates, and outlet gas rate - Test 1 (December, 1958)

Test 1. (December, 1958)

Coal - Edmonton coal, crushed and dried

Reactor temperature - 600°C.

Average coal feed rate - 23.5 lbs./hr.

Average air feed rate - 15.3 lbs./hr.

Time of steady operation - 29 hours

The records taken of the air and coal feed rates, and of the outlet gas rates are shown in figure 14. A copy of a portion of the temperature recorder chart taken during the test is given in figure 15. It is seen that the bag filter unit operated at 300°C., and that the difference in temperature between the bag-filter inlet and outlet was only 10°C., showing that at this temperature the heaters installed just balanced the heat loss.

Material Balance and Product Yields

Input:	Lbs.
Air	444.0
Coal	680.0
Air purge	17.0 (estimated)
Air leaks	13.0 (estimated)
	1,154.0
Output:	
Char	408.0
Char fines: Cyclone	20.5
Bag filter	2.0
Condenser liquor	85.5
Demister tar	3.0
Outlet gas	615.0
	1,134.0

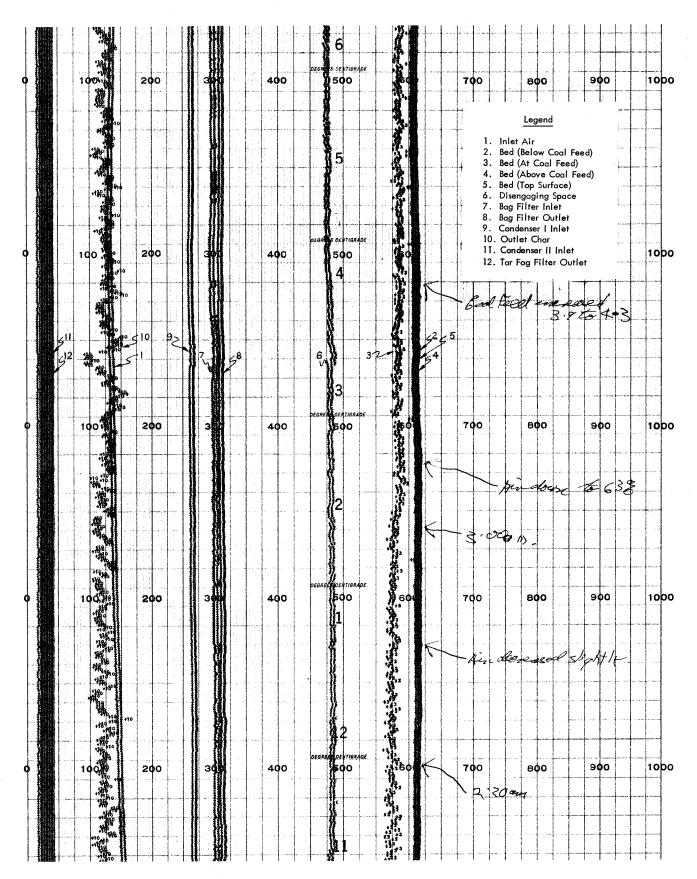


FIGURE 15. Portion of temperature chart - Test 1 (December, 1958)

On analysis the condenser liquor was found to contain approximately 4.5 lbs. of tar.

The yields of the various carbonization products obtained, expressed as a percentage of the coal feed as carbonized, are given below in table 4. The tar and light oil yield is seen to be only 1.4 per cent.

Table 4. Yields of carbonization products - Test 1 (December, 1958)

Product	Yield (% of coal feed as carbonized)		
Char	60.0		
Fines	60.0 3.3 } 63.3		
Tar	1.1)		
Light oil	1.1		
Gas	23.3		
Water	12.0		
	100.0		

Analysis of two samples of the outgoing gas are presented in table 5 and the proximate analyses of the coal feed and the various solid products are given in table 6. The high ash content of the fines taken from the bag filter is, to a large extent, due to some burning of this very reactive char dust having taken place during the shutdown period, when air was allowed into the bag housing.

Table 5. Outlet gas composition - Test 1 (December, 1958)

Composition by volume %	Sample A	Sample B	
Hydrogen	11.2	13.7	
Oxygen	0.9	1.7	
Nitrogen	59.0	53.0	
Carbon monoxide	4.2	5.4	
Carbon dioxide	19.8	20.4	
Methane	4.0	5.3	
Other hydrocarbons	trace	0.05 (approx.)	
	99.1	99.5	
Calculated average density = 0.0769 lbs./cu.ft.			
Calculated average h	neating value = 100 B.t	.u. per cu.ft. gross	

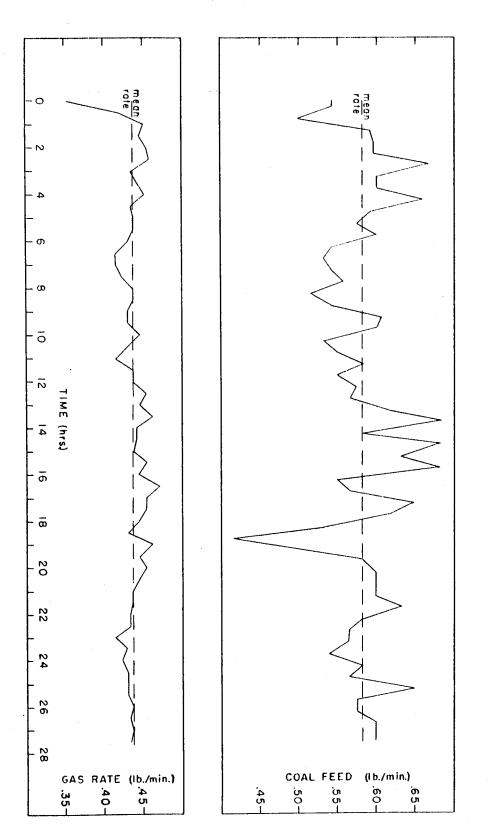
Table 6. Proximate analyses of coal feed and solid products for test 1 (December, 1958)

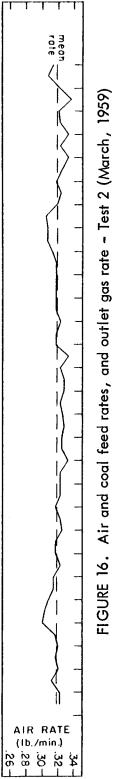
Material	Moisture %	Ash %	Volatile matter %	Dry Ash %	Dry volatile matter %
Coal feed	2.0	10.1	35.1	10.4	35.8
Char	0.4	14.0	9.6	14.1	9.6
Cyclone dust	1.0	19.8	16.2	20.0	16.4
Bag filter fines	1.2	32.3	14.8	32.7	15.0

A preliminary analysis was made of two samples of the tar obtained, one from the condensers and the other from the demister. The results of these are shown in table 7.

Table 7. Tar analysis - Test 1 (December, 1958)

	Condenser Tar	Demister Tar
Specific Gravity	1.013	1.042
Water	10%	0.7%
Distillation, wet sample		
110°	11.4%	1.8%
110 - 170°	0.7	0.3
170 - 235°	18.6	9.7
235 - 270°	15.3	14.0
270 - 300°	10.3	9.3
300 - decomp.	24.3	39.8
pitch	13.4	20.6
Decomposition temperature	358°C.	352°C.
Distillate analysis		
% Phenols	24	21
% Bases	3	3
Neutral Oil		
% Olefins	24	22.5
% Aromatics	64	69.5
% Paraffins	13	12
Refractive index	1.5404	1.5490





Test 2. (March, 1959)

Coal - Edmonton, crushed and dried

Reactor temperature - 600°C.

Average coal feed rate - 35 lbs./hr.

Average air feed rate - 19.2 lbs./hr.

Time of steady operation - 27.5 hours

Records of air and coal feed rates and of outlet gas rate are shown in figure 16 and a portion of the temperature recorder chart in figure 17. The bag filter operated at about 330°C., somewhat higher than in the previous test due to the increased gas rate.

Material Balance and Product Yields

Input:		Lbs.	
Air		527.5	
Coal		967.1	
Air purge		9.4	
			1,504.0
Output:			
Char		613.5	
Char fines	: Cyclone	33.5	
	Bag filter	2.0	
Condenser	liquor	120.0	
Demister to	ar	7.5	
Outlet gas		723.0	
			1,499.5
Difference			4.5 lbs. or 0.3%

The condenser liquor was found to contain about 6 lbs. of dry tar.

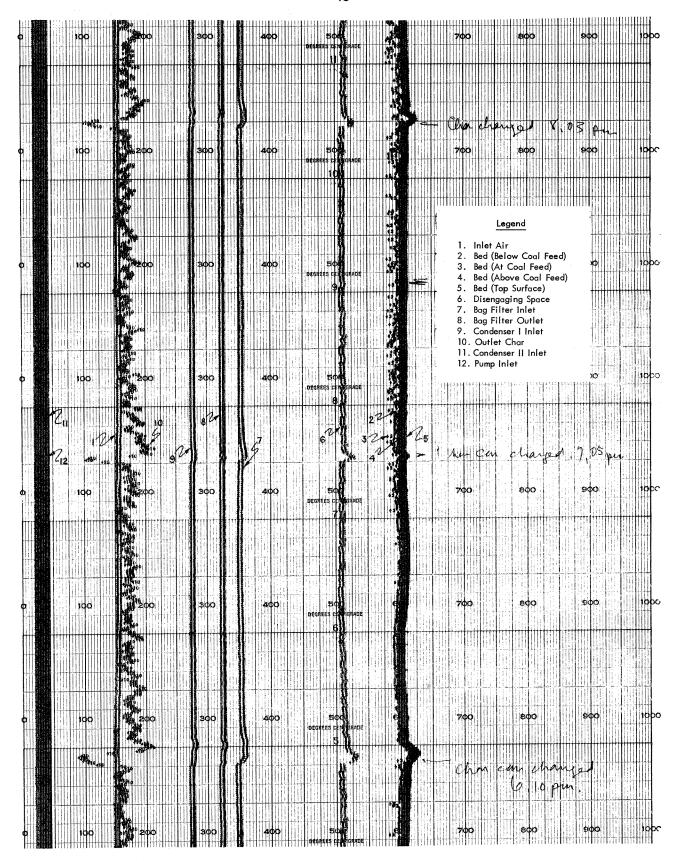


FIGURE 17. Portion of temperature chart - Test 2 (March, 1959)

The yields of the various carbonization products, expressed as a percentage of the coal feed as carbonized, are given in table 8. The main difference in the yields obtained in the second test, compared to those in the first (see table 4), is in the higher char yield and lower gas yield. This indicates that in the second test a lesser proportion of the char was burnt to provide process heat. This would be expected for higher throughput rates as the rate of heat loss to the surroundings is practically independent of throughput rate. The tar + light oil yield of 1.6% was still very low.

Table 8. Yields of carbonization products - Test 2 (March, 1959)

Product	Yield (% of coal feed as carbonized)
Char	63.3
Fines	63.3 3.7 } 67.0
Tar	1.3
Light oil	0.3
Gas	19.6
Water	11.8
	100.0

No gas analysis is available for the second test, but it is not expected to be much different to that found in the first test. Proximate analyses of the coal feed and of the various solid products are given in table 9, and a preliminary tar analysis is presented in table 10.

Table 9. Proximate analyses of coal feed, and solid products for test 2 (March, 1959)

Material	Moisture %	Ash %	Volatile matter %
Coal feed	3.5	10.1	33.8
Char	0.0	14.7	10.0
Cyclone dust	0.4	19.8	14.9
Bag filter fines	1.2	28.8	15.9
Pitch deposit at condenser inlet	0.4	0.7	70.4

Table 10. Analysis of tar for test 2 (March 1959)

	Condenser Tar	Demister Tar
Specific gravity	. <u>-</u>	1.04
Water content	34 (est.) %	0.66%
Acetylatable OH	4.4	4.2
Distillation, dry basis		
110°C.	1.9%	0.9%
110 - 170°	1.3	1.2
170 - 200°	0.9	1.5
200 - 235°	19.8	12.4
235 - 270°	19.1	12.7
270 - 300°	10.5	12.4
300 - Decomp.	21.0	35.2
Residue	22.8	18.8
Loss	3.4	4.7
Decomp. temperature	330°C.	363°C.
Distillate		
% Phenols	24	28
% Amines	3.5	3.5
% Neutral	72.5	68.5
Neutral Oil		
% Olefins	22	22
% Aromatics	71	64
% Saturates	7	14
Refractive index	1.5474	1.5492
Light oil		
% Olefins	64	
% Aromatics	28	
% Saturates	8	

Test with Lethbridge Coal

As reported earlier, the tests using Lethbridge coal were not successful. However, sufficient tar and filter bag fines were obtained for analysis. The proximate analysis of one of the troublesome pitch deposits is also given in table 11.

Coal - Lethbridge, crushed and dried

Reactor temperature - 600°C.

Average coal feed rate - 35 lbs./hr.

Average air feed rate - 19.3 lbs./hr.

Time of steady operation - 1.5 hours

Table 11. Proximate analysis of coal feed and some solid products of test using Lethbridge coal

Material	Moisture %	Ash %	Volatile matter %	Fixed carbon %
Coal feed	2.2	11.9	36.4	49.5
Deposit from filter bags	4.0	18.0	20.6	<i>57</i> .4
Deposit from pipe at condenser inlet	0.8	3.8	37.9	<i>57.</i> 5

Table 12. Analysis of tar from test using Lethbridge coal

Specific gravity	1.07
Water content	16.0%
Distillation (dry tar)	
110°C.	0.6%
110 - 170°	1.8
170 - 235°	7.3
235 - 270°	9.7
270 - 300°	8.3
300 - decomp.	15.7
Residue and losses	56.6
Decomposition temperature	330°C.
Benzene insoluble	11.0%
Ash content of benzene insoluble	13.3%
Dry distillate	
% Neutral oil	63
% Phenols	34
% Bases	3
Neutral oil	
% Aromatics	65.1
% Olefins	22.3
	12.6

APPENDIX 3. Heating Methods

There are several possible ways of providing the heat required by the coal for low-temperature carbonization. A number of these have been considered by Peytavy and Foch 10 but are further discussed here. These may be classified as to whether the heat is provided externally or internally.

- (1) External Heating This includes all methods where the heat is transferred to the coal through a wall separating the coal from the source of heat.
- (a) Ovens One of the simplest methods of achieving this heating is in rectangular ovens similar to those used for producing high-temperature coke. The chief difficulty with this method in the case of low-temperature carbonization is the low temperature differential which must be used in order to prevent overheating of the coal adjacent to the oven walls. This method is used in the Coalite process⁵, and the oven residence time can be reduced by preheating the coal charge to about 300°C., and by using metal walls for the ovens which are as narrow as reasonable in design.

In an effort to increase heat transfer rates, numerous processes were designed where the coal charge was stirred while being heated in the oven. Most of these processes failed due to the mechanical difficulties involved.

(b) Rotary Furnace - Probably the most successful of these designs was the rotary furnace, which consisted essentially of a metal tube through which the coal passed. This tube was externally heated and continually rotated so as to stir the coal charge which was usually preheated before entering the tube. This method is the basis of the Disco process⁶ and the rotary furnace process described by Peytavy and Foch ¹⁰.

One advantage of the two methods mentioned above is that the gas leaving the ovens is undiluted carbonization gas which has a fairly high calorific value of between 450 and 750 B.t.u./cu.ft., and as no other gas is added to the system, the load on the condensing units is kept as low as possible. The dust carry-over from these ovens should also be low, and not cause much difficulty.

(c) Fluidized Bed - An efficient method of stirring the coal charge which does not involve any moving parts in the high temperature part of the process is the use of a fluidized bed. This was the method used by Sinclair 17, and some external heating is involved in the Parry process 8, 16.

This process, too, will produce a gas of high calorific value if the fluidization medium is the gas itself, or steam. However, because of the presence of the fluidizing gases, the load on the condensers will be increased, the dust entrainment problem will be involved, and there will be an additional heat load on the process due to the need to heat the fluidizing gases. This heating might be achieved

by preheating so as not to increase the heat load of the reactor itself.

The major disadvantage of all of the processes using external heating lies in the difficulty of scaling-up the units. In general, the throughput of the unit increases as the square of the reactor diameter, while the area available for heat transfer only increases linearly. As a result, a point is soon reached when scaling up where the only way in which plant capacity can be increased, is to build a large number of small units. This is usually a much more expensive undertaking than increasing the size of the main unit.

- (2) Internal Heating In order to make the heating rates independent of the wall area of the reactor, a number of different systems using internal heating have been considered, mostly involving a fluidized bed technique.
- (a) Partial Combustion in Air This is the heating method which was used in the work described in this report, and had been adopted in nearly all the recent experimental work reported8,9,10,12,13,14,15,16.

Here the heat is provided by the partial combustion of the char by the air used to fluidize the charge. The design and operation of such a unit is relatively simple, and the carbonization temperature is easily controlled by adjusting the ratio of the air and coal feed rates.

The system however, has some disadvantages. The gas produced is diluted with atmospheric nitrogen and consequently has a very low calorific value (90 to 150 B.t.u./cu.ft.). This large amount of uncondensible gas also increases the load on the condensing system, and involves the dust entrainment problem inherent in all fluidized systems.

A controversial point about this process concerns the possible oxidation of the tar within the reactor, with a consequent loss of tar yield and tar value. The results given in this report showed that the tar yields obtained from the pilot plant were much lower than those expected from the results of laboratory assays, and it was feared that some of the tar may have been lost by combustion. No further tests were made towards settling this matter. Parry² reports that the lignite char used in his tests was so reactive that the oxygen in the air reacted with the char rather than with the tar vapours, and therefore the tar yields were not affected. Lang et al. ¹⁵ report tests with an excess of acetylene mixed with the air, to ensure that no far vapors were burnt. Since similar tar yields were obtained to those when no acetylene was used it was concluded that the oxygen reacts preferentially with the hot char, rather than tar vapors.

(b) Partial Combustion in Oxygen - This is essentially similar to the above process, except that oxygen is used instead of air. It is likely that the oxygen would have to be diluted with air, steam, or carbonization off gases. The main advantage of this process is that a high quality gas would be obtained as it would not be diluted with nitrogen. However, the cost of the oxygen would probably be prohibitive.

- (c) Heating by Introduction of Hot Gases In this case heat is provided by the sensible heat of the fluidizing gas; this eliminates any possibility of oxidizing the tar. However the heat capacity of gases is small, and the incoming gas would have to be heated to very high temperatures (ca. 1400°C.) in order to get a satisfactory throughput.
 - (i) Flue Gases These would be formed by the burning of a fuel immediately prior to the gas entry to the carbonization chamber. The hot products of combustion would then be used as the fluidizing medium and in cooling within the reactor would provide the heat necessary for carbonization. In this case the off gases would be diluted by the flue gases to such an extent as to make them incombustible. The high gas throughput would also give high condenser loadings.
 - (ii) Superheated Steam This has the advantage over the above method in that the off gas would have a high heat value. However, the cost of the steam and the equipment and fuel necessary to superheat it would probably be prohibitive.
 - (iii) Recycled Gas Here again the off gases would retain a high calorific value. Unfortunately, if the gases were recycled before cooling, the regenerators or recuperators used would probably soon be fouled by dust and the solid products of tar cracking. Also thermal cracking would reduce the tar yield. If the gas were first cooled before recycling, to remove dust and tars, the heat losses would probably be excessive.
- (d) Heating by Solid Bodies In this method the char would be heated by mixing it with a hot solid material. This process would not necessarily involve fluidization, but depending on the solid material used, may involve subsequent separation. Two possible materials have been considered.
 - (i) Char The raw coal charge would be mixed with a hot stream of char which had been heated above the carbonization temperature, probably by partial combustion. The two streams would be proportioned so that the resultant char mixture would attain a uniform temperature equal to the carbonization temperature chosen.

Again, the off gases would not be diluted and would have a high calorific value. Unfortunately, the portion of the resulting solid product which had been used as the hot stream would no longer be low-temperature char, and a considerable amount of volatile matter would be lost from the char in the production of the hot stream.

(ii) Refractory material – Here the raw coal would be mixed with a stream of hot particles of refractory material, sand or pebbles.

There would have to be provision for separating the refractory from the

resulting char, and means to recycle it through the heating chamber to be mixed with more raw coal. This system seems to offer distinct possibilities, although the equipment necessary may be expensive.

Whatever heating method is used, it would seem worthwhile to reduce the heat load within the reactor by predrying and preheating the coal, and preheating the air in the case where partial combustion in air is involved.

APPENDIX 4. Dust Entrainment and Collection

Probably the biggest problem encountered during the course of this pilot plant investigation was that due to dust entrainment. This problem is inherent in all fluidization work and has been discussed in most reports of work in this field.

The char dust entrained in and carried over by the gas stream tends to cause blockages in the equipment following the reactor, and will, if not separated before condensing the tar, reduce the value of the tar and make it more difficult to refine. Several of the possible uses of the tar, particularly for electrode carbons, demand a nearly ash free tar, and as it is not feasible to separate the dust from the tar by filtration or centrifuging it is necessary to remove the dust from the tar vapors before condensation.

The reasons for dust collecting equipment in the case of low-temperature fluidized carbonization are seen to be different to those usual in other processes where the major reason is either to recover the dust because of its value (e.g. in catalytic cracking units), or to prevent an air pollution problem, and where in general dust recovery need not be complete. Another difference is that in this case it is necessary to remove the dust while the gases are still hot (above 350°C., i.e., before the tars begin to condense). This condition excludes the use of several types of dust collecting equipment such as water scrubbers and cloth bags, and necessitates modifications of the units which can be used.

It is also desirable to keep the time of passage of the tar vapors through the hot dust collection system as low as possible in order to minimize thermal cracking of the tar, with consequent loss of tar yield and blockage of the equipment with the hard carbon deposits produced by cracking. The pressure drop through the dust collection system should also be kept low to minimize process power requirements.

Various dust collection systems have been reported by the various investigators in this field, and they all include one or more of the following devices.

- (1) Disengaging Space This is merely the space between the top of the fluidized bed and the gas outlet of the reactor, in which the solid particles that are thrown up from the bed have a chance to fall back through the uprising gases. In some cases 10,17, including the plant described in this report, the disengaging space has a diameter much larger than the reactor itself so that the gas velocity in this region is reduced to allow the particles to fall back more easily.
- (2) Internal Cyclones These cyclones which are situated inside the upper part of the reactor are very common in large scale fluidized processes. Their advantages are that the separated dust is returned directly to the fluidized bed by means of a dip leg extending below the surface of the fluidized bed, thus making a seal to prevent the off gases bypassing the cyclone, and they are easily kept hot because of their position within the reactor.

Minet²⁷ reports satisfactory dust removal by using two such cyclones in series, although he gives no figures for the amount of dust passing the cyclones. An internal cyclone was used in the first and second plants described in this report, but the results were far from satisfactory. It is felt that internal cyclones would prove more successful in a larger unit.

(3) External Cyclones - One or more of these have been used in nearly all the reported units. Cyclones are low in initial cost, have a fairly low pressure drop, and are simple to install and operate, and have no moving parts. For the process in question they have to be heated to prevent condensation of tar within the cyclone.

These cyclones if properly designed will remove a large amount of the dust from the gases, but some of the finer dust particles will pass the cyclones and continue on with the gas stream. In two prolonged tests in the plant described in this report the cyclones removed 91 per cent and 94.5 per cent of the entrained char entering the cyclones. In both cases about 0.07 lbs. per hour of dust passed through the cyclones to be caught by the bag filter. Lang et al. 15 report that when using cyclones only for dust collection, the tar contained 8 to 10 per cent of dust.

- (4) Electrostatic Precipitators Two of the pilot plants reported in the literature have used electrostatic precipitation for dust removal. Lang et al. 15 report the dust content of the tar was reduced from 8-10 per cent to 1.5-3 per cent when an electrostatic precipitator was installed. Parry et al. 8 give some figures showing a reduction of about 60 per cent in the dust content of the gas stream after passing through the electrostatic precipitator; however, the tar still contained 2-3 per cent dust. Electrostatic precipitators are high in initial cost, but have low pressure drops, and low power requirements if operated properly. The unit is usually fairly large in volume which will mean some difficulty in heating it, as well as high residence times with possible cracking of the tar vapors.
- (5) Bag Filters The normal bag filter unit could not be used in this process where the filter has to operate at temperatures well above those allowable for most bag materials. The use of glass cloth for filter bags has developed in recent years and this material might withstand the temperatures required (ca. 350°C.). A heated glass bag filter unit was used with some success in the third pilot plant described in this report when using Edmonton coal. However, on changing to Lethbridge coal, the unit failed owing to cracking of the tar vapors throughout the dust collection system. Glass bags would be expected to give almost complete separation of the dust from the gas stream. However, owing to the capital expense and space requirements with consequent difficulty in designing suitable heating equipment, and the possibility of cracking owing to the high residence time of the tar vapors in the filter unit, it is not thought that a bag filter unit would be satisfactory for a large scale plant. It is not yet possible to estimate the probable life of the bags. Some deterioration of the glass cloth was evident after about 80 hours of operation in the tests conducted.

(6) Partial Condensation - Owing to the difficulty of completely removing the dust from the gas stream, some investigators have suggested the following system of partial condensation. The bulk of the dust is first removed using simple cyclones and the gas stream is then cooled to 200 - 280°C., depending on conditions, when the heavier pitch fraction would condense. This pitch would contain nearly all the dust which had passed the cyclones, so that on further cooling of the gas in subsequent condensers a dust-free tar would be obtained.

Although this scheme seems to have some merit in that it obviates the need for expensive dust removal equipment, it has some disadvantages. The solid matter in the pitch fraction would make it unsuitable for electrode production, and, depending on the physical nature of this fraction, there might be considerable operating difficulties due to blockage of the equipment by the pitch-char dust paste.

Other novel and more complex methods of dust removal have been suggested, but no reports of their performance have been published. To date the problem of entrained dust in the gas stream remains as the major difficulty in the operation of a low-temperature fluidized carbonization unit.