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2. CARBONIZATION

Carbonization, destructive distillation, or pyrolysis is the heat treatment of organic substances in the absence of air to decompose the substances into a solid residue, liquid products, and gases. The temperature level applied during the process of carbonization influences greatly the yields and characteristics of the recovered products.

According to A. C. Fieldner (8): $\frac{1}{2}$

Low-temperature carbonization of coal may be defined as the heat treatment of coal in the absence of air at temperatures of 450° to 700° C. (842° to $1,292^\circ$ F.) as distinguished from the usual high-temperature carbonization at temperatures of 900° to $1,200^\circ$ C. ($1,652^\circ$ to $2,192^\circ$ F). At 450° to 500° C. (842° to $1,292^\circ$ F.) the tar yield is 2 to 3 times that of the ordinary high-temperature process for making coke or gas.

Table 12 shows approximate comparative yields obtained from low- and high-temperature carbonization processes.

TABLE 12. - Approximate comparative yields of high- and low-temperature carbonization processes 1/2/

	Low-temperature	carbonization	High-temperature
	Externally heated	Internally heated	carbonization
	retort	retort	byproduct coke oven
Cokepercent	. 70 - 80	60 - 75	60 - 70
Volatile matter in			
cokepercent	7 - 15	7 - 15	1 - 2
Gascu. ft./ton	3,000 - 5,000	20,000 - 50,000	11,000 - 12,000 🍋
Heating value of			*
gasB.t.u./cu.ft.	800 - 1,000	150 - 250	520 - 580
Tar gal./ton	20 - 30	18 - 20	10 - 12
Specific gravity of tar	1.07 - 1.09	1.02 - 1.07	1.19
Light cilgal./ton	2.5 - 3.0	None	2.5 - 3.0
Ammonium sulfatelb./ton	10 - 12	12 - 18	25 - 30

Fieldner, A. C., Low-Temperature Carbonization of Coal: Bureau of Mines Tech. Paper 396, 1926, p. 43.

2/ Data apply to the carbonization of coal and not to lignite.

The yield of coke is higher for low-temperature carbonization processes because it retains 7 to 15 percent of volatile matter, as compared with 1 to 2 percent in high-temperature coke. Volume of gas is only one-fourth to one-third that obtained in high-temperature coking, but the heating value is almost twice that of gas from high-temperature carbonization when externally heated retorts are used. Low-temperature-carbonization processes internally heated by products of combustion produce large quantities of gas of low heating value. The total yield of tar at low temperatures is from 2 to 3 times that obtained at high temperatures. The quantity of light oil is about the same for both processes. However, the concentration of light-oil vapors in gases obtained by internal heating is too low for efficient recovery.

Although these data apply to the carbonization of higher rank coal, yields obtainable from carbonization of lignite show the same general trend as a function of the processing temperature. Unlike the bituminous coals that form a coherent coke upon heat treatment, lignite and subbituminous coal do not become plastic and therefore produce a residue or char, consisting of nonagglomerated particles smaller than the material charged.

Results of Assay Tests on North Dakota and Other Selected Lignites

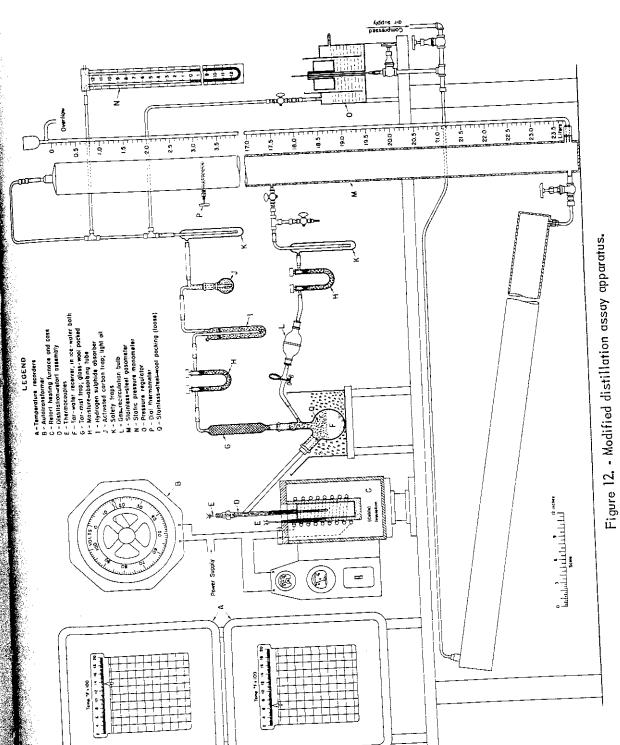
Preliminary evaluation of the economic promise of low-temperature carbonization of a given coal or lignite is primarily based on the results of laboratory investigations, which supply information on yields of tar, coke, and gas. Laboratory-scale carbonization in properly designed assay apparatus has been shown to indicate the approximate maximum tar yields to be obtained at a given carbonization temperature. Variation of yield as a function of processing temperature is also readily determined. Comparison of yields obtained in commercial plants with those from assay tests is helpful in determining optimum operating conditions and evaluating various processing methods. The Franz Fischer assay apparatus developed in Germany has been widely used in carbonization assay investigations. The method, which uses 250 grams of coal, is described by Davis and Galloway (7), who assayed numerous coals by this technique.

V. F. Parry and coworkers refined the Fischer method (13) for use with low-rank coals; the apparatus is shown in figure 12. From 150 to 200 grams of coal is used for a test. A carbonization temperature of 500°C. was adopted as a standard, whereas in the Fischer method 550°C. was originally employed. Because of the modifications used, results of the assay tests with the original and modified methods are not strictly comparable. However, yields of tar and light oil differ by not more than approximately 0.2 percent. Gas yield is slightly higher and coke yield lower at the higher temperature. The influence of changes on assaying temperature on yields from Sandow lignite is shown in figure 13. Results of assay tests on lignites made by the Fischer method have been reported by J. D. Davis and A. E. Galloway (7); H. F. Yancey, K. A. Johnson, and W. A. Selvig (31); W. A. Selvig (29); and W. H. Ode and W. A. Selvig (21).

Results of tests made on North Dekota lignites are reported in table 13. To eliminate difference in moisture and ash content of the various lignites, yields were calculated to moisture- and ash-free basis, unless MAF data were already available in the original publications. Average yields and range of yields for North Dakota lignites calculated from data of table 13 are given in table 14.

The rather wide variation of tar yields, ranging from 5.0 to 9.4 percent for all tests listed in table 13, should be noted. Lignites from Divide, Williams, Burleigh, Morton, Stark, and Grant Counties had higher tar yields than those obtained from Burke, Ward, and Mercer Counties. Although results of a number of assay tests run on North Dakota lignites are available, lignites from some areas of the State have not as yet been assayed.

Additional results of assay tests on other United States lignites are given in table 15. Only one lignite from South Dakota was tested. The yield of tar and light oil from this lignite was 5.8 percent. Lignites from Arkansas had yields of tar and light oil as high as 20 percent calculated to the moisture- and ash-free basis. Primarily this results from a different petrographic composition, as pointed out by Selvig (29).



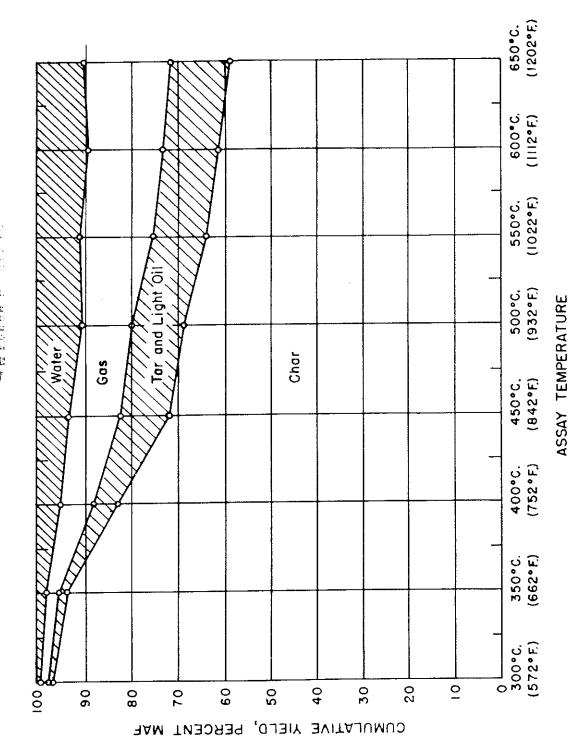


Figure 13, - Low-temperature carbonization yields in laboratory precision assay from Sandow lignite (sample 482).

TABLE 13-Results, of asser carbonization tests on North Dakota, Monites by the Flesher, method

Y N. Dak. Grent Knupp	New Letpaig	34.1 10.1 550	9.0	48.1 2.1 15.6 20.1 20.1 7.4 4.12 2.613 1078 12.30
K. Lak. Horton New Salem	New Salem	37-1 7-0 550	60,0 12.3 8.2 18.6	23.3 23.5 23.6 23.6 25.6 25.00 11.3
N. Dak. Stark	Dickinson	200	6•9	
N. Dak. Stark Fittsburgh	blekinson	42.6 5.7 550	8.3 22.4	53.0 1.6 11.6 11.6 18.9 5.860 5.860 5.860 1093
W. Dak. Burleigh Wilton	Wilton	40.9 5.5 550	62.3 11.6 8.6 18.3	23.8 23.8 116.5 116.5 22.5 22.5 22.5 22.5 23.0 23.0 23.0 23.0 23.0 23.0 23.0 23.0
3/ N. Dak. Burleigh Wilton	Wilton	39.7 6.0 350	21.2	50.3 1.4 11.7 11.7 11.8 16.9 5.9 2.800 5.000 1019 8.1
N. Dak. Mercer Beulah	Beulah	200	6*5	
N. Dak. Mercer Beulah	Beulah	30	7-6	
N. Dak. Mercer Beulah	Beulah	35.6 5.7 550	62.7 12.6 6.1 18.1	25.6 20.0 20.0 20.0 20.0 20.0 20.0 20.0 20
N. Pak. Mercer Pratt	a Z	33.6 6.2 550	64-4 10-3 6-0 19-3	2,77.2 1.6 10.1 15.2 21.7 2,660 53.00 53.00 1066 8.6
N. Dak.	Velva	30	5.4	
N. Dak. Ward Volva	Velva	900	0.2	
M. Dak. Ward Volva	Velva	35.4 5.0 550	60.8 14.3 7.2 17.1	2.2 2.4 2.4 2.4 2.4 2.3 2.3 2.3 2.3 2.3 2.3 2.3 2.3 2.3 2.3
N. Dak. Williams Reclams -	Williston	36.1 9.9 550	59.1 13.9 18.7	46.3 117.5 117.5 119.2 2,320 2,320 4,54 4,64 4,64 4,64 4,64 4,64 4,64 4,64
3/ R. Dak. Rurks Truex	Columbius	32.2 8.5 550	5.71 17.9	46.7 1.48 1.48 19.3 19.3 5.5 408 2.405 4010 4010 4010 4010
2/ E. Dak. Divide Baukol- Moman	Hooman	200	7.6	
M. Dak. Divide Bankol- Noman	Поолел	200	85 83	۵. د څ
State County Mine name		Moleture, as recd., % Ash, as recd., % Temp. of distill., °C. sear yields. MAR. %.	Char Water Tar + 11ght odl Gas Rydropen Sulfide	Control doctor Thermonia Thermonia Performe Reforme Reforme

2) ofth, W. G. and Salvig, W. A., Lon-Semerature Distillation Yinlis of Friency for and ident Oil from Goals of Warlous Ranks and Types: fures to investigations 3748, 1944, 19 pp. L Selviy, W. A., Properties of Lignites of the United States: Fast, vol. 32, No. 1, January 1953, pp. 28-35.

Novie, J. D., and Gallony, A. F., Low-Temporature Carbonization of Lightless and Sub-lithmatnone Conjec. Ind. Eng. Cham., vol. 20, No. 6, 1936, pp. 612-17.

L'Ancry, H. F., 3 demon, X. A., and Selvis, W. A., Printlilly, Shekkin; Characteristics, Low Tesperature Carbonization Asses, and aggluithating Walne of Wanington and Other Genia: Bureau of Mines Tech. Paper 323, 1932, 94, pp.

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TABLE 14. - Assay-carbonization yields of North Dakota lignites by Fischer-assay method

lightes by riocas-		
Assay yields, MAF, percent1/	Average 61.6	Range 59.1 - 64.4
har	12.5 7.4	10.3 - 14.3 6.0 - 8.6 17.1 - 19.3
Water Far + light oil	18.4	17.07 - 73.03
Cos data:	431	389 - 466 4,640 - 5,320
Net gas B t 11 /lb MAF	1,040	902 - 1,135 8.6 - 11.2
Heat in gasgal /ton lignite as mined Tar + light oilgal for this calculation.	<u> </u>	1. Itemites

1/ Only complete tests used for this calculation. TABLE 15. - Results of assay tests on other United States lignites by Fischer-assay method

	By F TB OLL			
	17	2/	2/	3/
) - 1, - Daleata	Arkansas	Arkansas	Washington
	South Dakota	Ouachita	Ouachita	Lewis
State	Harding	003022	1	Prospect
County	Ellis	}	_	
<u> </u>	Gobert	 		
Moisture, as recd percent	41.0	1		
Moisture, as recu do	7.4	500	500	500
Moisture, as recudo. Ash, as recddo.	550	700		
Temperature of discre-				
MAW percent		1		
Char	' I ' I	ļ		
	' \ - 0	19.8	20.5	10.7
- 13 mb# 07 1				
	-			
Gas Hydrogen sulfide	• -			
Hydrogen surrent:	1			
Gas composition, percent:	. 55.3			
Carbon dioxide	1.6			Ì
Illiminants	7.0			
Carbon monoxide	16.1			
W -3	~ l 1.	ţ		ĺ
	1 1. 6		1	1
Ethane	' -			
/ a) and gross-	337			ì
B.t.u./cu.ft., calc., gross				1
(== 36ATi	2.570		1	
Net gas, cu.ft./lb. MAF	5,140		1	
Net gas, cu.ft./ton MAF			1	1
/- *****	866	- 1	1	1
Heat in gas, B.t.u./lb. MAF	•••		ļ	
	7.2		<u> </u>	on of
Tar, gal./ton as recd	A E Low-Tem	perature Ca	arbonizati	M2 6
1/ Davis, J. D., and Galloway,	Cools: Ind.	merature of Eng. Chem.	, vol. 20,	110.0,

Lignites and Subbituminous Coals: Ind. Eng. Chem., vol. 20, No. 6, 1/ Davis, J. D., and Galloway, A. E.,

2/ Selvig, W. A., Properties of Lignites of the United States: Fuel, vol.

3/ Ode, W. H., and Selvig, W. A., Low-Temperature Distillation Yields of Primary Tar and Light Oil From Coals of Various Ranks and Types: Bureau of Mines Rept. of Investigations 3748, 1944, 10 pp.

Attrital lignites, like the Arkansas lignites, which contain more or less debris of leaves, bark, spores, pollen, and resins, yield more tar than woody lignites, which predominate in North Dakota.

V. F. Parry and coworkers have tested numerous coals and lignites in the modified distillation-assay apparatus and reported the results in various publications (12, 23, 24), from which complete data on North Dakota lignites and selected data on Texas lignites are presented in tables 16 and 17. Among the Texas lignites, only the Sandow lignite is significant in extent of deposits.

Average yields and range for both North Dakota and Texas lignites are shown in table 18.

Yields of products from low-temperature carbonization of North Dakota lignites, recalculated to as-mined basis and expressed in English units of measure per ton of lignite carbonized, are given in table 19.

On the average, approximately 82 percent of the potential heat of the lignite was recovered in the char, about 10 percent in the tar and light oil, and 6 to 8 percent in the gas.

An extensive series of determinations of carbonizing properties of coking coals has been carried out by the Bureau of Mines in cooperation with the American Gas Association. In the BM-AGA test (26), 13- or 18-inch-wide retorts containing charges of 75 to 180 pounds of coal are used, in comparison to the 150 to 250 grams employed in the assay tests previously described. This procedure and apparatus have been used to investigate the carbonizing properties of a single North Dakota lignite, Velva lignite from Ward County, at temperatures ranging from 500° to 900° C. (932° to 1,652° F.) (27).

At 500° C. approximately half of the yield of tar and light oil indicated by Fischer assay was obtained in the BM-AGA test. The reduced yield is the consequence of longer residence time of the charge in the BM-AGA test, coupled with a considerable temperature gradient across the retort which promotes cracking of tar vapors at the wall and in the hotter section of the charge.

Lignites shrink considerably during the process of carbonization as a result of the large amounts of water vapor and gas released during decomposition of the coal substance.

The contraction of volume of Velva lignite as a function of test temperature is shown in table 20, as determined in the EM-AGA test.

TABLE 16. - Results of assay tests on North Dakota lignites by modified distillation-assay apparatus

	Bankol - Bankol	Bankol-									
Mine name	Noonan	Noonan	Kincaid	Custer	Garrison	Garrison	Garrison	Garrison Beulah	Beulah	Zap	Dickinson
County	Divide	Divide	Burke		McLean	McLean	McL ean	McLean	Mercer Mercer		Stark
Moisture, as recd.		\ \ \ \		9		C	o c	000	,	l c	- 00
percent		ð,	34.0	0.66	ر د. د.	5.0°	30°	30.0	٥. ج	7,	30.1
Ash, as recd do.	5,2	6.9	6.2	⊅.	 6.3	6.3	1. 6	1. 6	0.6	6.3	7.7
Temp. of distillation,					-			-			
0	200	200	200	200	200	주	200	元0	200	500	500
Assay Vields MAH nement:											
Char.		9. 29	68.2	67.3	69.3	72.2	68,2	73.5	68.7	67.9	66.3
	10.0	10.5	11.11	6	11.2	9.0	9.3	8,5	9.6	10.6	13.0
Tar		5.4	7, 4	5.6	9.9	7.3	0	9		6.4	٠ ٣
Light oil		† ⊤	դ.⊤	1.5	1.6	9.1	1.1	1.4	1. 9.	ω	1.2
Tar + light oil		9.9	5,6	7.1	8.2	6.8	4.6	8.0	5.9	6.2	5.1
Gas	13.4	15.1	15.0	15.8	11.0	10.6	13.1	10.3	15.2	15.0	14.9
Hydrogen sulfide		0.3	0.3	0.1	ł	į	ı	1	φ.	ij	6.
Gas composition nercenti											
Carbon dioxide	19.5	54,3	52.2	51.1	36.2	55.8	† † †	61.9	75.4	8,64	54.3
Illuminants	0.7	0.7	0.5	0.5	1.3	, T	1.0	6.0		ω.	, , ,
	13.9	11.3	13.0	14.5	11.4	12.3	11.4	13.5	11.0	12.9	11.4
		11.4	8.6	9.6	16.2	0.0	13.9	8.4	10.1	10.3	6.6
Methane	24.3	21.2	23.2	23.2	28.7	22.1	29.3	18.6	20.4	25.5	22,8
Ethane	1.9	1,1	1.3	1:1	Q 9	س ش	0.0	0.3	†•. L	1	1.1
B.t.u./cu.ft							-		•		
calc. gross	366	318	337	338	511	366	394	568	310	358	325
NOT GRE	1 657	1 Rof	80	000 [1,000	00,	7/4 [061 1	ממי	7 860	30%
Heat in cas	-	10.1	700.1	1.750	ر مر•۲	764.4	2,	OCT • T	9	000° T	1.(%)
B.t.u./Ib. MAF	607	580	617	650	809	1,36	688	303	555	999	584
										-	

TABLE 17. - Results of assay tests on Texas lignites by modified distillation-assay apparatus

(11 50111				
	Malakoff	Sandow	Sandow1	Freestone
Mine name		Milam	Milam	Freestone
	Henderson	35.2	35.6	36.2
County Moisture, as recdpercent	36.3	8.4	7.9	8.1
Moisture, as recd. do.	8.0	500	500	500
Ash, as recd	500)00		
Assay yields, MAF, percent:	() (66.1	68.0	64.2
	64.9	8.6	7.3	8.0
	7.6	11.5	10.9	14.6
Water	14.7	2.0	1.8	1.8
Light oil	1.8	1	12.7	16.4
Tar + light oil	16.5	13.5	12.1	11.1
Tar + 11ght off	10.9	12.3	1	
G85		1	Ì	
Gas composition, percent:	10.5	42.4	41.7	36.2
a a a dioride	42.5	1.8	1.9	1.8
ral	' 1	9.8		9.8
a tem monoyide	0	6.2		15.6
T Jung mon	* l = 2 1.	38.1	^	34.6
16 blooms	1.0	1.7	1	2.0
Ethane	• 4.0	,		1
		499	465	500
B.t.u./cu.ftcalc. gross	9	1.400		1.627
B.t.u./cu.ft	F 678	697	-1.0	814
Net gasB.t.u./lb.MA	<u> </u>			_
Heat in gas	ъте.			

1 Average of 4 tests on same sample.

TABLE 18. - Comparative assay yields of North Dakota and Texas lignites, assay temperature 500° C. (932°F.)

	No rt	n Dakota	T	exas				
		Range	Average	Range				
Assay yields, MAF, percent: Char	68.1 10.5 5.4 1.4 14.3	66.3 - 69.3 9.3 - 13.0 3.9 - 8.0 1.2 - 1.6 11.0 - 15.8	65.8 7.0 12.9 1.9 11.6	64.2 - 68.0 7.3 - 8.6 10.9 - 14.7 1.8 - 2.0 10.9 - 12.3				
Gas data: B.t.u./cu.ftcalc.gross Net gascu.ft./lb.MAF Heat in gasB.t.u./lb.MAF		310 - 511 1.583-1.860 555 - 809	481 1.527 735	460 - 500 1.400-1.627 678 - 814				

TABLE 19. - Yields of products of low-temperature carbonization of North Dakota lignite

	Baukol-			T	Ī	T	1	
Mine name	Noonan	Kincaid	Custer	Garrison	Beulah	Zap	Dickinson	
County	Divide	Burke	McLean	McL ean	Mercer			
Moisture in coal as		!				1 201	Doulk	Average
minedpercent	36.6	34.0	39.0	39.9	36.0	33.7	38.1	36 . 8
Ash in coal as	İ				50.0	22.1)0.1	20.0
minedpercent	6.9	6.2	4.4	6.3	9.0	6.3	7.7	<i>-</i>
Yields, per ton of		1		3	7.0	,	1 - 1	6.7
coal, as mined:		ļ						
Charlb.	902	940	850	871	936	941	873	000
Targal.	7.4	6.0	7.6	8.5	5.7	7.1		902
Light oildo.		2.6	2.6	2.6	2.7	2.4	2.2	6.8
Gascu. ft.	2,062	2,187	2,173	1,703	2,010	2,230		2.5
Waterlb.		813	890	919	826	794	1,954	2,046
		3	• • • • • • • • • • • • • • • • • • • •	7-7	020	174	902	856
Gas B.t.u./lb. of								
lignite	328	369	368	435	305	400	276	
CharB.t.u./lb.	12,330	12,610	12,970					360
Potential heat of	,555	,010	 ,,,,,	12,410	11,770	12,000	12,060	12,400
lignite recovered								
in charpercent	80.1	82.0	81.4	84.2	02.2	07.0	0	
Volatile matter in	00.1	02.0	01,4	04.2	83.3	81.8	80.7	81.9
charpercent	17.8	18.0	18.6	18.6	10.0	70.0	7.5	- 0 - 4
Ash in char do	13.9	11.5	9.1	7.7	19.0	18.8	19.2	18.6
wo.	-3.5		9.1	12.4	17.6	13.0	15.5	13.3

TABLE 20. - Contraction in volume of charges of Velva
lignite as mined in RM-AGA tests as a
function of processing temperature

Test temperature, °C.	Contraction, percent by volume
500	33.1
600	47.3
700	47.5
800	51.5

Process Methods

Experimental carbonization of lignite has been carried out in this country since the middle of the last century. Objectives of lignite carbonization have changed from time to time as a result of contemplated end uses for the products. Before development of convenient oil-burning equipment and construction of numerous power plants, almost all of the lignite mined in the North Central region was used for domestic heating. The main objective of lignite carbonization under these circumstances was to provide a smokeless, agglomerated fuel from the resultant char. Recovery of tar and oils was considered of secondary importance. When coal gas was rather extensively used for municipal distribution in North Dakota, gas coals for manufacturing the gas had to be imported. It was therefore proposed to use lignite in the conventional retorts of gas plants for manufacture of city gas. Because gas was the desired end product, carbonization temperatures had to be very high, which resulted in low tar yields. Meanwhile, oil and natural gas have increasingly replaced solid fuels for domestic heating, and electricity and petroleum gases are predominantly used for cooking and hot-water service. The more recent concept of the end uses for products of carbonization of lignite are utilization of the char as fuel for power generation and utilization of the tar as a source for chemicals. In this case, the char must compete with nonprocessed lignite, natural gas, and heavy fuel oils, and revenue from tar must absorb the cost of the carbonization process. Therefore, the potential tar content of the lignite should be high, recovery of the tar should be as complete as possible, and the carbonization process should be cheap.

Pilot and Commercial Plants for Carbonizing American Lignites

Glenk (11) has reported that Arkansas lignite was carbonized as early as 1859 by Robert Peter to produce gas for generating heat and power. A very crude process to carbonize Arkansas lignite was operated in 1885. The resultant char was pulverized and mixed with the oils and other ingredients and the mixture sold as rough black and mixed with was used principally on iron and steel.

In the summer of 1900, the Laclede Gas Light Co. of St. Louis, Mo., installed an experimental bench of 4 horizontal D-shaped fire-clay retorts in which 20 carloads of Arkansas lignite were carbonized. As a result of high carbon dioxide content of the gas, which was too high to be removed economically, it was concluded that lignite gas could not be used as illuminating gas (6).

Later, Darling (6) designed an inclined slowly rotating cylindrical iron retort, which was placed in a combustion chamber. The process proved satisfactory with respect to yields, but the iron retort deteriorated rapidly under the high temperature.

E. J. Babcock, of the University of North Dakota, and his coworkers took a leading part in early work on development of North Dakota lignite. Extensive investigations were made on carbonization of lignite and briquetting of residual char. An tions were made on carbonization of lignite and briquetting of residual char. An experiment station was established at Hebron, N. Dak., where experiments were carried out on a large pilot-plant scale.

Babcock (1) and Babcock and Odell (2) reported in detail on types of carbonizing equipment and presented operating data. Hood and Odell (15) reviewed development of the technique of lignite carbonization up to that time.

Although the Bureau of Mines since its establishment had taken an active interest in utilization of lignite and development of the lignite deposits of the United States, in utilization of lignite and development of the Secretary of the Interior to Congress, by an act of February 25, 1919, authorized the Secretary of the Interior to make investigations, through the Bureau of Mines, of lignites and peat to determine the

practicability of utilization as fuel and in producing commercial products. As a result of this authorization, a cooperative agreement between the Bureau of Mines and the University of North Dakota was signed in 1922 covering the joint use of a lignite carbonizing oven and accessories. Results of runs made in this oven were reported by Odell $(\underline{22})$.

Externally heated cast-iron or clay retorts first used were operated at fairly high temperatures, ranging from 1,200° to 1,800° F., with the maximum quantity and quality of gas obtained at 1,200° to 1,400° F. The clay retort was capable of carbonizing about 400 pounds of lignite in 4 hours. Lignites from North Dakota, Montana, Colorado, and Texas were tested. Average figures resulting from 25 experiments run on air-dried lignite containing 8 to 27 percent moisture were reported as follows:

Operating temperature, 1,110° - 1,700° F.

Gas yield

11,038 cu. ft. per ton of air-dried lignite

Heating value of gas

396 B.t.u. per cu. ft.

Char yield

54.6 percent of the original charge of air-dried lignite

Heating value of char

11,838 B.t.u. per pound

After further preliminary experiments, a large, externally heated, inclined retort having a capacity of about 20 tons of lignite per day and consisting of 6 double retorts with 18 gas offtakes arranged to remove gas at 3 separate levels, was constructed and operated. Tops of the retorts were not closed, a feed hopper of lignite acting as a seal. A single retort was 16 feet long, 30 inches wide, and 10 inches high. Gas and byproducts could be collected, and stripped gas could be returned to the fire box. The influence of temperature, size of lignite, and time of carbonization on quality and quantity of products was investigated. Yields of tar were small because some tar was carbonized in the fuel mass. The char obtained was satisfactory for briquetting. A typical material balance for a ton of lignite, calculated from data for a test rum of 1 week, is given in table 21.

TABLE 21. - Material balance for carbonization of lignite in inclined retort

		Pounds	Percent
Input:	Lignite, as mined (moisture 28.2%)	2,000	100
Recovereā:	Char	807	40.4
	Water, calc. from lignite analysis	564	28.2
	Gas	302	15.1
	Tar	19	0.9
Total accou	nted for		84.6
	ed for and losses		15.4

Losses include water-soluble products, leakage, combustion of gases and tar leaking into the flues, and water formed on distillation of the lignite.

In an attempt to obtain increased capacity and lower processing cost, an internally heated oven of relatively simple design was built. This oven consisted essentially of a vertical shaft 6 feet high inside and 3 by 3 feet wide. A device

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to cool and quench the char was attached to the carbonizing shaft. To supply heat for carbonization, a combustion zone was maintained at the middle of the oven by supplying air through airports opening into the combustion zone, burning a portion of the lignite charge. No provision was made to collect gases and tar evolved during the carbonization process. The oven carbonized 16 tons of as-mined lignite a day, producing 6.4 tons of char suitable for briquetting.

Lurgi-Spulgas Process

The concept of using lignite char as a source for storable, high-B.t.u. briquets led in 1928 to construction of two commercial plants on the North American continent, one at Dickinson, N. Dak., and the other at Bienfait, Saskatchewan, Canada. These two plants use the Lurgi-Spilgas internally heated carbonization process developed in Germany in the early 1920's (3, 17).

The basic principles of the Lurgi process are the following:

- 1. Any noncoking fuel, crushed and screened to suitable size can be used without pretreatment.
- Tar can be recovered with high yield, without excessive cracking, and without dust.
- 3. The charge is slowly dried at controlled temperatures before the actual carbonization process.
- 4. The zones of drying and carbonizing of the coal and cooling of the char are clearly separated.
- 5. The whole three-step process is entirely continuous.

Figure 14 shows a diagram of the Lurgi carbonizer.

The North Dakota plant at Dickinson, formerly known as the Lehigh Briquetting Co. and now operated by the Dakota & Tar Products, Inc., has been described by R. Dawson Hall (14).

In this plant lignite as mined is crushed to 4-inch or less, and the crushed material is passed over a vibrating screen. Undersize, consisting of approximately minus-1/4-inch particles, goes to the power plant, and oversize is charged to the carbonizer.

In the upper section of the retort, moisture is removed from the lignite, and in the lower section carbonization takes place. Part of the gas originating from carbonization and having a heating value of about 190 B.t.u. per cubic foot is mixed with air and burned outside the drier, and the products of combustion are recirculated through the drying zone. Moisture is removed from the lignite during its passage through the drier and is vented to the atmosphere. Considerable disintegration takes place during the drying process, leaving a dried product of approximately 3/4 inch maximum size, which descends through 8 pipes into the carbonization section, which is operated on the same principle as the drier and is equipped with ducts for distributing the hot products of (external) combustion from the recycle gas. The gas mixture enters at $1,300^{\circ}$ F. and heats the lignite to approximately $1,100^{\circ}$ F., decomposing it to char, gas, and tar vapors. Char residue goes to the bottom of the carbonizer, where it is cooled by carbonization gas, as recycled. A gas mixture consisting of carbonization gas, tar vapors, and inert products of combustion is drawn off at the

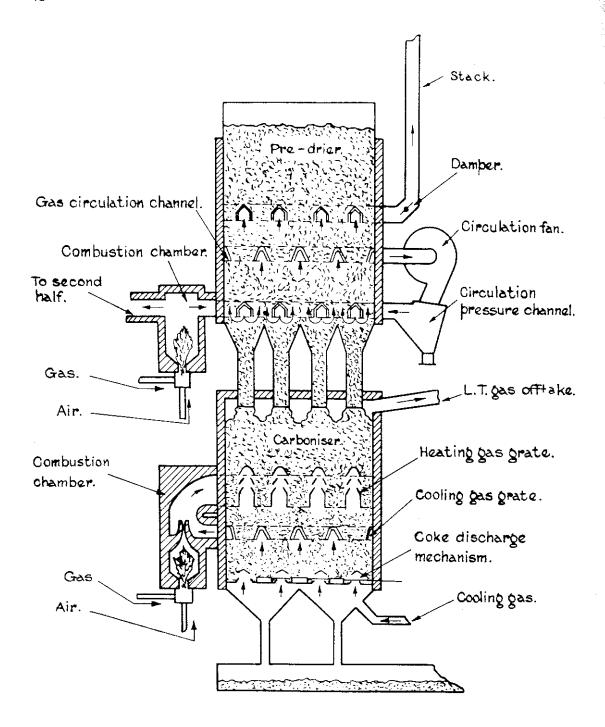


Figure 14. - Diagram of Lurgi carbonizer.

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top of the carbonization chamber at approximately 350° F. Char is discharged at the bottom at about 200° F. Total height of the carbonizer installed at Dickinson is approximately 85 feet, and the drying and carbonization sections are each 20 feet long and 6 feet wide.

Gas leaving the carbonization section is washed in a water-spray scrubber to take out dust, tar is removed by an electrostatic precipitator, and final cooling is accomplished in two scrubbers filled with grids. Clean gas is returned to the carbonizer to cool the char and supply the heat requirement for drying and carbonization.

There are two Lurgi carbonizers installed at the Dickinson plant, each having a daily capacity of approximately 100 to 150 tons of lignite as mined.

At Dickinson the tar and oils, after separation from water condensate, are distilled into a liquid fraction and a pitch residue. The distillate oil is sold for use as a wood preservative, and the pitch serves as part of the binder for manufacturing briquets from the char. Pitch obtained is not sufficient to meet requirements for briquetting and must be supplemented by petroleum asphalt.

Approximately 5 gallons of tar and oils per ton of as-mined lignite were recovered during actual operation of the plant, or about 70 percent of the yield, as determined in the laboratory. Tar and oil recovery is influenced in part by the fact that facilities are not available for stripping the light oil from the gas after the tar-recovery system. It has been reported (3) that 83 percent of the assay yield was recovered in a newer Lurgi-Spülgas plant, which scrubbed light oil from the gas, whereas the yield was 71 percent of assay disregarding the recovered light oil.

Economic success of the Dickinson plant did not come up to expectation because its main value product is briquets for the domestic market, which has been declining because of competition from other fuels. Quality of the char briquets, having a heating value of about 12,000 B.t.u. per pound, is very satisfactory as far as burning properties and storability are concerned. Tar production is not sufficient in quantity to process it for maximum realization.

Fluidized Carbonization, Bureau of Mines, Denver, Colo.

Work on low-temperature carbonization of lignite, using new techniques, has recently been successfully reactivated by V. F. Parry of the Denver laboratories of the Bureau of Mines. The Texas Power & Light Co. cooperated with the Bureau of Mines in this development work. In this process, carbonization of fine-particle coal in the fluidized state is used to obtain high heat-transfer rates, high capacities, and low operating costs ($\frac{1}{2}$, $\frac{1}{2}$, $\frac{1}{2}$). Feed size of the coal is minus-1/4-inch. Required residence time of the particles in the carbonizer is 10 to 15 minutes.

The process developed by Parry consists of two steps. Lignite, as mined, is crushed to minus-1/4-inch and dried in the entrained state in a stream of hot gases. This process has been described in section 5 part 1. The dried coal is then transferred to the fluidized carbonization retort. In the carbonization retort, process heat is furnished in part by combustion within the retort through limited addition of air and in part by external heating. The mixture of char, gases, and tar vapors leaving the retort at the top is separated in conventional dust-removal equipment and a specially designed hot electrostatic precipitator. Nonagglomerating coals can be carbonized readily in the entrained and fluidized state by this technique.

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Because operating data are not available from the commercial plant now under construction, the equipment used during development will be described. The 300pound-per-hour pilot plant, which was used to develop the technique of fluidized carbonization and to determine yields, is shown in figures 15 and 16. It consists of an 8-inch steel retort having a sprayed coating of 312 alloy to prevent oxidation. The retort is suspended within a refractory-lined outer chamber heated with gases produced by combustion of natural gas and air in an external furnace. Coal is fed by a mechanical vane feeder from the dried-coal hopper into a stream of air or recirculated process gas that transports the dried coal to the fluidizedcarbonization retort. With a gas velocity of 3 feet per second at the base of the retort, the average particle of dried coal remains in the retort approximately 12 minutes. If gas is used to transport the coal, all heat of carbonization is supplied by transfer from the hot-gas zone through the retort walls. If air is used, internal combustion supplies part of the heat required. Approximately 60 percent of the heat of carbonization is supplied by combustion resulting from the use of 3 cubic feet of air to transport 1 pound of dried coal into the carbonizing system. Early tests were made with the annulus shown inside of the retort in figure 15. Clean process gas was recycled down this annulus. The gas absorbed heat from the hot wall and carried it down to the base of the retort, where the gas joined the stream entering the central retort tube. The annulus was intended to distribute the heat evenly throughout the retort and assure uniform temperatures. It was found, however, that this was unnecessary, and later tests were made without the annulus. In these later tests, all of the gas entering the retort entered with the coal stream. Leaving the top of the retort is a mixture of char, water vapor, tar and light-oil vapors, and gas. This mixture is passed through a primary separator, which removes approximately 93 percent of the solids, and is then passed through the heated secondary separator, where 70 percent of the remaining solids is removed. Vapors leaving the secondary separator are at the carbonizing temperature, usually 900° F. They are first passed through an indirect air-cooled condenser, where the heavy ends of tar and most of the carryover solids are removed. Residual vapors and gases leave the air-cooled condenser at about 340° F. and pass through a tube condenser, where they are cooled to about 150° F. The gases then pass through the steam-jacketed electrostatic precipitator, where the remaining tar is recovered, and leave the precipitator at 150° F. From here the gases go to the second indirect tube condenser to remove water and the last traces of condensable light oil. They leave the second tube condenser at 60°F. and are pumped into the recycle system or through the meter to storage or disposal.

Only one North Dakota lignite - Garrison lignite - was included in the series of coals tested in the pilot plant. Yields from the carbonization of this lignite are compared in table 22 with those from Sandow lignite from Texas processed under various operating conditions. Sandow lignite will be used to produce char for the Texas Power & Light Co. and the Aluminum Co. of America at Rockdale, Tex. $(\frac{1}{4})$.

Garrison lignite yielded approximately 70 percent of the amount of tar and light oil obtained from Sandow lignite under comparable processing conditions. As previously shown, North Dakota lignites in general yield less tar and light oil than Texas lignites.

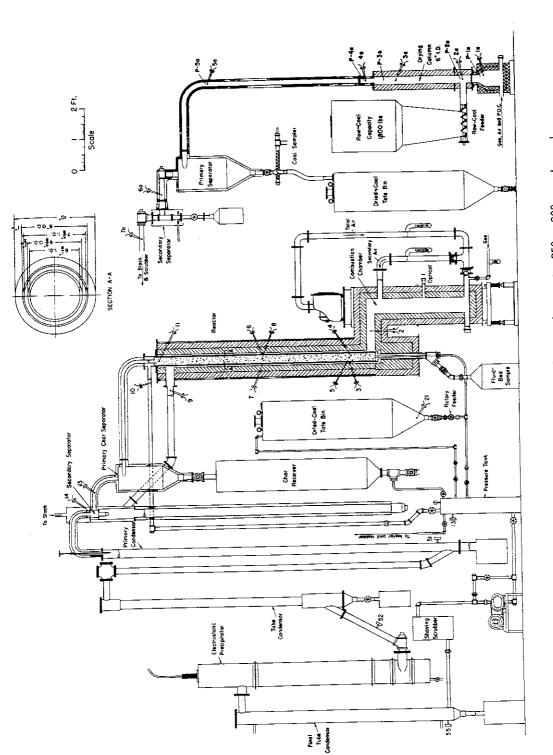


Figure 15, - Pilot plant for fluidized carbonization of noncoking coal-capacity 250 to 300 pounds per hour.

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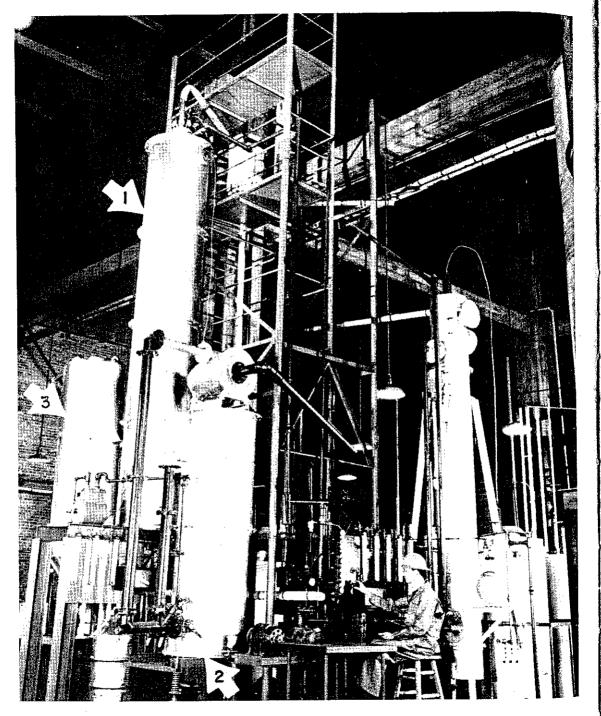


Figure 16. - Eight-inch fluidized carbonizer, Denver, Colo. (Arrow 1 points to retort and housing, arrow 2 points to combustion chamber, and arrow 3 points to dried coal hopper.

TABLE 22. -- Yield of products from lighties carbonized in 8-inch fluidized carbonizer 1/

^{2/} Helds shown represent yields of products obtained from the lignite only. The transport air used in run 36 is not included.

^{3/} Gas and water yields unreliable; data not reported.

^{1/2} Includes hydrogen and carbon gasified by transport air.

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Introduction of air with the lignite during run 36 increased the capacity of the unit appreciably, since I cubic foot of air, by internal combustion produces approximately 100 B.t.u., of which approximately 70 B.t.u. is available for carbonization after correction for the sensible and latent heats of the products of combustion at 900° F. The yield of tar remained essentially the same whether or not air was used, and the B.t.u. in the gas per pound of lignite remained constant. Introduction of air, of course, resulted in lowering the B.t.u. per cubic foot of gas because of the dilution effects of the products of combustion. The chief effect of air appeared to be reduction of the yield of char and increasing the yields of total gas and water. The yield of tar actually increased in run 36, when air was introduced, as the result of a new shipment of Sandow lignite, which yielded 13.2 percent tar and light oil in the assay test on moisture- and ash-free basis, as compared with 12.2 percent of the Sandow lignite used during run 14.

Analyses of the chars obtained from runs 11, 10, 14, and 36 are given in table 23. Physical properties of Garrison lignite and char are listed in table 24.

TABLE 23. - Chemical analyses of char

Carbonization test No	11	10	14	36½/
	840	840	900	900
	Garrison	Sandow	Sandow	Sandow
	North Dakota	Texas	Texas	Texas
Proximate analysis: H20percent Volatile matterdo. Fixed carbondo. Ashdo.	0	0	0	0
	28.7	30.2	26.0	24.1
	56.9	53.8	55.6	57.4
	14.4	16.0	18.4	18.5
Ultimate analysis:	3.6 66.8 1.3 13.3 0.6 14.4	3.5 66.6 1.5 10.9 1.5 16.0	3.3 65.9 1.4 9.1 1.9 18.4	1.5 8.9 1.5
Heating valueB.t.u./lb.	11,160	11,140	10,950	10,970

1/ Air introduced, 2.5 cu. ft. per lb. MAF lignite.

TABLE 24. - Physical properties of Garrison lignite and char

	As mined	Dried	Char
Average particle sizeinches	0.082	0.041	0.019
Densitylb./cu.ft.	41.4	40.0	35.9
Hardgrove grindability index		76.1	94.1

Average particle size was considerably reduced during processing, and grindability index was improved.

Composition of gases for the test runs of table 22 are presented in table 25.

TABLE 25. - Composition of carbonization gases, 8-inch fluidized carbonizer

Carbonization test No	11 840 Garrison North Dakota	10 840 Sandow Texas	14 900 Sandow Texas	36 900 Sandow Texas
Source	North Dakota	0	0	2.5
Air per lb. MAF lignitestd.cu.ft. Volume, percent:	1.2	0.7 51.6 4.9 0.3 17.3 4.7 14.2 4.0 2.3 389	4.1 42.4 5.7 1.4 14.0 10.9 17.8 2.4 1.3 465	1.1 23.2 2.6 0.2 6.3 3.2 6.5 2.9 54.0 218

Increasing the carbonization temperature from 840° to 900° F. reduced the concentration of carbon dioxide and carbon monoxide and increased hydrogen and methane. Application of internal heating by introduction of air with the lignite primarily increased the nitrogen content of the gas, thereby lowering the heating value; however, because of the presence of hydrocarbons, heating value was still higher than that of conventional producer gas.

Results of assay tests have shown that about 80 percent of the heating value of the lignite was retained in the solid residue. Pilot-plant data are similar, as shown in table 26.

TABLE 26. - Distribution of potential heat per pound of lignite as charged, 8-inch fluidized carbonizer

Ondai Sout			_	
Carbonization test No	90 Sand	low	36 <u>.</u> 900 Sand Tex	low
Name of lightee	Text B.t.u.	Percent		Percent
Lignite, as charged Char Tar + light oil	7,100 5,666 1,143	100.0 79.8 16.1	7,000 5,082 1,253	
Gas	348 43 7,100	4.9 8 100.0	441 224 7,000	3.2
Total	1 1 1		1,000	1 200

1/ Air introduced, 2.5 cu. ft. per lb. MAF lig

The available data did not allow a heat balance to be calculated for the Garrison lignite from North Dakota. When Sandow lignite was carbonized at 900° F., the char, which was 49.3 percent of the weight of the original lignite, contained 79.8 percent of its heating value. The char and the liquid byproducts (tar and light oil) contained 95.9 percent of the heating value of the lignite. When additional internal heating by admission of air was used, B.t.u. recovered in the char were reduced to 72.6 percent whereas B.t.u. in tar and light oil did not change

materially. The authors have therefore assumed that the air reacts primarily with the char and does not burn the liquid products of carbonization.

Since the economic success of the process depends on credits realized by sale of the liquid products, their recovery in the carbonizing equipment should approach that of the assay test, which is considered to give approximately optimum results.

In figure 17, yields obtained in the 8-inch pilot plant processing Sandow lignite at various temperatures are compared with tar-plus-light-oil yields from the 500° C. assay test.

The lower curve indicates that approximately 95 percent of tar plus light oil determined by the assay test was recovered in the pilot plant at carbonizing temperatures ranging from 900° to 950° F. The crosshatched area shows that some residual tar and light oil were left in the char, which, when added to the recovered tar and light oil, would exceed the assay yield. Low-temperature carbonization in the entrained and fluidized state in the equipment developed by the Bureau of Mines provides efficient recovery of liquid products. A high percentage of the original heating value of the lighte is recovered in a greatly reduced weight of char, which constitutes a high-B.t.u. boiler fuel.

Investigations to Produce Coherent Coke From Lignite

Char obtained from normal carbonization of United States lignites is of a loose, noncoherent nature, not suitable physically for blast-furnace use.

Laboratory studies have been made by the division of mines and mining experiments of the University of North Dakota in an effort to improve the coking characteristics of North Dakota lignite. Studies were made of the influence of the following variables on carbonization:

- 1. Addition of inorganic materials before carbonization.
- 2. Blending with bituminous coals and pitches before carbonization.
- 3. Mechanical pressure during carbonization.

It has been known for some time that addition of inorganic salts to either coking or noncoking coals influences both the structure of the resulting coke and its reactivity. In the tests with North Dakota lignite, various anhydrous and hydrated salts were added to samples of crushed, air-dried lignite, lignite was extracted with hydrochloric acid or sodium hydroxide solutions, and steam-dried lignite, and the mixture was then carbonized (9, 18, 19). Char, gas, and tar yields were measured and the physical structure of the char noted.

Hydrated aluminum salts, notably Al₂(SO₄)_{3.18H₂O and AlCl_{3.6}H₂O, caused enough cementation of char particles to produce coherent pseudocoke having, in some instances, a bright metallic luster resembling metallurgical coke and in others a black luster, as with petroleum coke. Hardness of selected cokes equaled that of petroleum coke but was less than that of metallurgical coke.}

Removal of a portion of the lignite ash by acid treatment increased the effectiveness of the aluminum hydrates but did not affect char, tar, and gas yields. Prior treatment with sodium hydroxide to remove the alkali-soluble humates destroyed the effectiveness of $\text{Al}_2(\text{SO}_4)_3.18\text{H}_2\text{O}$ in producing coherent coke and decreased the char, tar, and gas yields compared to carbonization of unextracted lignite with equal additions of aluminum hydrate.

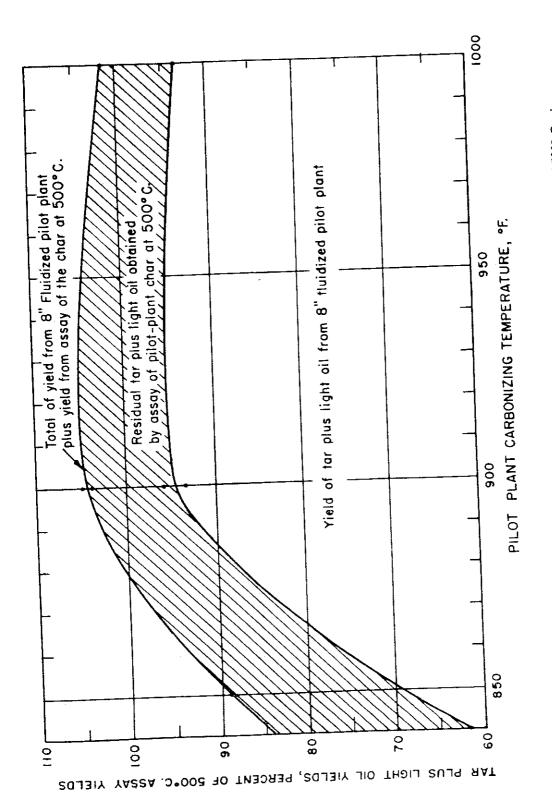


Figure 17. - Comparison of tar plus light-oil yields from 500° C. Sandow-lignite assay with pilot plant and 500° C. char assay.

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Addition of inorganic salts to steam-dried lignite did not yield as satisfactory a coke as the nondried lignite, and tar yields were generally lower (18).

Chars produced from lignites treated with anhydrous salts, including calcium carbonate, sodium carbonate, and aluminum sulfate or chloride, were similar to untreated chars. Some additional hydrous salts investigated, such as ferric chloride hexahydrate and magnesium chloride hexahydrate, were not effective in promoting formation of coke.

The mechanism by which the pseudocoke forms is apparently one in which the melted salt dissolves in its water of crystallization and wets the surface of the lignite particles. The hydrocarbons evolved during carbonization pass through the surface film and in the presence of the inorganic matter are decomposed or cracked, forming a hard graphitic layer, which cements the carbonized particles (9).

Generally, yields of char and gas were increased somewhat and tar yield decreased when the inorganic salts were added. However, the process was not promising for commercial application because of the large amounts of hydrate required, ranging from 10 to 40 percent, by weight, of the lignite carbonized.

Carbonization properties of various blends of dried or precarbonized lignite with bituminous-coal tar, petroleum, and wheat-straw pitches or bituminous coals have been investigated $(\underline{10})$. Only blends of Pocahontas coal and lignite char containing at least 40 percent by weight of the coal gave a satisfactory coke. Addition of hydrated inorganic salts improved the coking characteristics of the various blends.

Bituminous coal-tar pitch was not satisfactory. Petroleum pitch gave a satisfactory coke if inorganic hydrates were added. Addition of 10 percent petroleum pitch reduced the requirement of aluminum chloride hexahydrate to produce coke by a factor of 5. Pitch was prepared from straw by steam distillation or pyrolysis with yields of about 4 percent. Blends with 16 to 20 percent of straw pitch and small amounts of inorganic hydrate produced a good coke upon carbonization (10). When mechanical pressure was applied to the material undergoing carbonization, improved coking characteristics were obtained. A coherent residue could be made without adding binder or inorganic salts. However, carbonizing pressures were high, being initially some 20,000 pounds per square inch, with the final pressure somewhat less and inversely related to the carbonization temperature. The size of particles subjected to thermal treatment influenced the coke characteristics - minus-20-mesh material gave the best results. Additions of inorganic hydrate or blending material required to obtain comparable cokes were reduced when the carbonization was carried out under pressure. Again, the pressure requirements and salt additions were such that such processes could not be considered for commercial use under present technological and economic conditions.

German Processes for Carbonizing Lignite and Brown Coal

The main purpose of brown-coal carbonization in Germany was to recover tar and light oil as a source of wax, fuel oil, diesel oil, wood preservatives, and motor fuel. The resultant char was used as boiler fuel, as a source of hydrogen for hydrogenation of brown-coal tar, and to a minor extent for domestic fuel. Yields of tar and light oil as high as 17 to 22 percent on a moisture- and ashfree basis are typical for Central German brown coals, which differ significantly in composition and properties from American lightes.

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Both external and internal heating were used to supply heat for the carbonization process. External heating requires that the bed of brown coal be thin to obtain adequate rates of heat transfer and retort capacity. Provision is made to deflect the descending stream of brown coal from the externally heated wall to secure uniform heating. Because of the narrow carbonization space, the previously dried brown coal must be crushed to approximately 0.04 to 0.12 inch to maintain free flow. Advantages of external heating are: Higher heating value of the undiluted carbonization gas, smaller gas volume to be processed, and higher concentration of light-oil vapors in the gas, which facilitates light-oil recovery. However, the capacity of the carbonizers is lower than that of internally heated units. Because of the small initial size of the brown-coal particles and the further breaking up of the particles during carbonization, the char particles are very small and require careful handling to prevent spontaneous heating or dust explosion.

In conventional German practice internal heating is carried out by burning the carbonization gas in a combustion chamber and introducing the products of combustion into the carbonizer. With this method of heating, the capacity of a carbonizer unit can be very large, the size being limited by the required uniform distribution of the products of combustion through the charge. It is necessary to use a uniformly sized feed, usually briquets made from previously dried brown coal. Lignites of a consolidated nature, like North Dakota lignite, can be carbonized by internal heating in as mined condition and need not be briquetted. Disadvantages of internal heating are: Low heating value of the gas because of dilution of products of combustion, large gas volumes, which must be processed, and the low concentration of light oil vapor in the gas, which does not permit efficient recovery by scrubbing.

Examples of externally heated carbonizers that were used commercially are the Rolle, Kosag-Geissen, and Borsig-Geissen retorts, shown in figure 18.

The Rolle retort is an improved and modified design of a carbonizer that was extensively used in the early days of brown-coal carbonization. To increase daily capacity and size of the char, the former operation on brown coal as-mined was changed to carbonization of briquets made from brown coal dried to approximately 14 percent moisture. The main parts of the Rolle retort are two cylinders hung in a cylindrical shell constructed of refractory with surrounding heating flues (30, p. 20). The upper cylinder, made of cast iron, is internally heated by gas to dry and heat the briquets to carbonization temperature in the shortest possible time. The annulus formed by the outer and inner cylinder is 12 inches wide. Water vapor and gases are withdrawn by a louverlike hood resting on top of the cast iron cylinder. Products of combustion leaving the cast-iron cylinder pass into the main flue-gas duct. The lower cylinder consists of a steel framework that supports refractory brick, each having a hole allowing the gases to escape from the annular space into the inner part of the cylinder. Gas is withdrawn at the bottom of the cylinder. Annular space in the carbonizing section is 7 inches wide. The retort has a carbonizing capacity of 12 tons of briquets per day. Tar recovery is 70 percent of the yield by Fischer assay. Because of their low capacity, only 3 plants, producing approximately 72,000 tons of tar per year, were equipped with Rolle retorts in Germany at the close of World War II.

The Kosag-Geissen and Borsig-Geissen retorts (fig. 18) also employ external heating of the carbonization space (30, pp. 22, 25). The basic design of the two retorts is similar, the latter being an improved modification of the first one. Both retorts are operated on dried and crushed brown coal, which descends between two concentric metal cylinders, the inner one of which rotates slowly and is heated by a large Bunsen-type gas burner. Baffles set in the carbonization space deflect

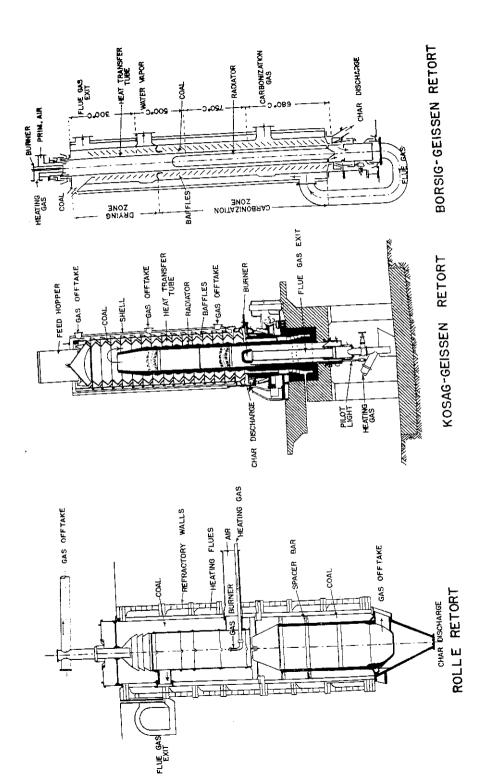


Figure 18. - Externally heated German carbonizers.

the stream of descending coal particles to secure uniform heating. Carbonization gases pass through openings between the louverlike baffles into a gas-collecting space from which they are withdrawn through several gas offtakes. In the Borsig-Geissen retort, provision is made for flue gases leaving at the bottom of the heating chamber to be returned to an annulus surrounding the gas-collecting space to prevent formation of deposits. The main difference between the two designs is the width of the annular carbonization space, which is 2 inches for the Kosag-the width of the annular carbonization space, which is 2 inches for the Kosag-Geissen retort and 3/4 to 1 inch for the Borsig-Geissen retort. Reduction of the width of the carbonization space accounts for a higher capacity of the Borsig-Geissen retort per unit area of heated surface or unit volume of carbonization space.

In table 27, comparative figures are given for test runs on the two types of carbonizers (30, pp. 24, 30, 31).

TABLE 27. - Comparative figures for Kosag-Geissen and Borsig-Geissen retorts

T of over	Borsig-Geissen	Kosag-Geissen
Type of oven Throughput/daytons Moisture of brown coalpercent Heated surfacesq.ft.	34.6 15.3 136	41.0 15.6 1,359
Capacity of heated surface	21.2 12.33	2.5 258
Capacity of carbonization space	23 ¹ 4 115.8	13.3 91.8

In addition to a number of Kosag-Geissen retorts, there were 22 Borsig-Geissen retorts in Germany at the close of World War II, the combined tar production of which was approximately 145,000 tons per year. A design of a large Borsig-Geissen retort, which supposedly would carbonize 120 tons of dried brown coal per day, was developed but not built.

The internally heated Lurgi-Spulgas retort (30, pp. 32-41), the same system that is in commercial use at Dickinson to carbonize North Dakota lignite, found the widest application in Germany. Basic design of the improved carbonizer is essentially like that previously shown. The capacity of a carbonization unit when using brown coal has been increased since the Dickinson plant was built. Two shafts, each approximately the size of the Dickinson carbonizer, were placed on each side of the central combustion chambers supplying the heating gases. As a result of improved distribution of heating gases, the capacity per unit volume of carbonization space was increased when brown-coal briquets were used; however, there was no substantial increase of the capacity per unit volume in direct carbonization of lignite similar to North Dakota lignite. Recovery of tar and light oil was approximately 90 percent of that obtained by Fischer assay. Heat consumption per pound of briquets containing 15 percent moisture was in the range of 900 to 1,100 B.t.u., leaving some excess gas, which was used to dry the brown coal before it was briquetted. The heating value of the stripped gas was from 180 to 215 B.t.u. per cubic foot, depending on the brown coal used. Gas obtained from carbonization was sufficient in all cases to supply heat for both carbonization and drying.

At the end of World War II 98 Lurgi-Spulgas retorts were operating in Germany, carbonizing 39,000 tons of brown-coal briquets per day, of 11 to 15 percent moisture

content, in the production of 1,560,000 tons of tar and light oil per year. Each Lurgi-Spulgas unit thus had a carbonizing capacity of close to 400 tons of briquets per day. In addition, 80 Lurgi-Spulgas retorts were concentrated at 1 plant processing 25,000 tons of 35 percent moisture Czechoslovak lignite per day in the production of 900,000 tons of tar and light oil per year. This Czechoslovak lignite was similar in moisture content and in physical structure to North Dakota lignite. Each Lurgi-Spulgas unit had a capacity of approximately 310 tons of lignite per day.

Slightly more than 90 percent of the total low-temperature carbonization tar produced in Central Europe during World War II from briquetted brown coal and asmined lignite was obtained from Lurgi-Spulgas retorts. The low-temperature tar was almost exclusively used as feed stock for hydrogenation plants.

Properties of Low-Temperature Lignite Tars

The temperature of carbonization influences the characteristics of tar and light oil produced from coal. True primary tars (24) are produced by techniques in which the tar vapors are swept from the carbonizing chamber without secondary thermal decomposition. Primary tars generally have low specific gravity, ranging from 0.95 to 1.05 at 25°C. High-temperature tars have a higher specific gravity of 1.15 to 1.25 at 25°C. and are relatively dark. Low-temperature tar differs essentially in composition from high-temperature tar. The composition is not well known. It contains few simple aromatic substances; complex tar acids and hydro-aromatic and aliphatic substances are present. The majority of the aromatic compounds present in ordinary high-temperature tar are formed by pyrolysis during carbonization (16, p. 384).

Properties of typical low-temperature tars obtained by fluidized carbonization of lignite and subbituminous coals are compared with those from high-temperature carbonization of bituminous coals in table 28 (24).

Low-temperature tar obtained from lignite and subbituminous coals is characterized by a high concentration of tar acids, by lower yield of residue when distilled to equal temperature, and by absence of naphthalene, which is a product of rearrangement of the constituents of primary tar as a result of high temperatures.

The distribution of tar acids in tar obtained by fluidized carbonization of lignite and subbituminous coals is shown in table 29 (24).

Concentration of the commercially more desirable lower boiling acids is low. If lower boiling tar acids are desired, the high-boiling tar acids must be cracked. Development of catalytic cracking processes is now being attempted.

The method of distilling low-temperature-carbonization tars influences the relative yields of liquid distillate and solid residue. In industrial tar processing, tube-still distillation produces more liquid distillate and less pitch and fixed gas than conventional pot-still distillation. In the laboratory, distillation at reduced pressure increases the tar-distillate fraction boiling above 270° C. over and above that obtained by distillation at atmospheric pressure, with a simultaneous reduction of yield of solid residue. Since liquid products are, in general, more valuable than the solid pitch, careful consideration should be given to proper selection of the distillation processes to realize the high possible credit from the tar.

TABLE 28. - Properties of typical low-temperature tar obtained by fluidized carbonization of lignite and subbituminous coal compared with properties of byproduct tar from coke ovens

	Internally heated retort,	Koppers coke
Carbonization process	entrained + fluidized	oven <u>l</u> /(average)
Specific gravity of dry tar60°/60°	1.043	1.180
Benzene insoluble percent	8.3	4.6
Ash in dry tar do.	1.0 - 3.0	-
Solidifying point°C.	25 - 30	-
Maximum dist. temp do.	310	300
Distillate analysis, vol. % dry tar:		
0-170° C	2.5	0.7
170° - 200°C	4.5	0.4
200° - 210°C	3.5	0.7
210° - 235°C	8.0	5.3
235° - 270°C	12.0	11.1
270° - end point	24.5	8.1
Total distillate	55.0	26.3
Pitch residue and loss	45.0	73.7
Tar acidsvol. % dry tar	20.0	2.01
Tar bases do.	2.0	-
Neutral oil do.	40.0	-
Naphthalene in 170° - 235°C.		
fractionvol. % dry tar	0.0	7.45
Composition of neutral oil, vol. %		
dry tar:		
Olefins	12.0	_
Aromatics	20.0	-
Paraffins	8.0	-
Pitch residue:		
Melting pointcube in air°C.	110	-
Melting point ring and ball do.	-	48.5
Specific gravity 25°/25°	1.200	

1/ Rhodes, E. D., Chapter in Chemistry of Coal Utilization: Vol. II, John Wiley & Sons, 1945, pp. 1287-1325.

TABLE 29. - Distribution of tar acids in primary tars

Tar acids	Boiling range, OC.	Volume percent of tar-acid fraction	Volume percent of total tar
Phenol O-cresol m- and p-cresol Isomeric xylenols High boiling acids Acid residue		2.0 1.0 3.0 28.0 23.0 43.0	0.4 .2 .6 5.5 4.5 8.5

When lignite and subbituminous coal tars were subjected to atmospheric or vacuum distillation in the laboratory, it was noted that lignites produced the greater quantity of distillate rich in high-boiling fractions; the effect was more pronounced in the distillate produced by distillation under reduced pressure. Tar from subbituminous coals showed a greater spread of the distillation products by the two distillation techniques.

EL.

Preliminary characterization data on properties of lignite tar obtained from Texas and North Dakota lignite by the fluidized, entrained technique are given in table 30. It may be noted that the temperature of carbonization was 900° F. for attrital Sandow lignite, whereas it was 840° F. for woody garrison lignite. Both temperature and different petrographic composition may contribute to the different characteristics of the tars. Decomposition temperature of the Garrison tar is distinctly lower than that of Sandow lignite, which accounts for the lower yield of distillate. The concentration of tar acids is appreciably higher for the woody Garrison lignite than for Sandow lignite.

No analyses are available for the light oil that can be extracted from the gas in fluidized carbonization. Properties of light-oil extracted from the gas of a Lurgi-Spulgas retort processing German brown coal briquets are given in table 31 (30, p. 289)

Our present knowledge of the properties and potential uses of low-temperature lignite tar is of rudimentary nature because work on this subject is limited. Extensive work on chemical composition, products obtainable, and potential uses, which is now being carried out at various laboratories, should permit more accurate estimation of credits obtainable.

Uses for Products From Low-Temperature Carbonization of Lignite

The chief products from low-temperature carbonization of lignite are char, tar and light oil, and gas. Char is the most important product on a weight basis, being approximately 45 to 50 percent of the lignite as mined. The char contains about 80 percent of the original heating value of the lignite when external heating or internal heating by inert gases is used. Partial internal heating by introducing air with the lignite reduces this figure to 73 to 75 percent.

Because the yield of tar and light oil from North Dakota lighte is only about 4 to 6 percent of the weight of the lighte as mined, only processes that consume large quantities of char can make tar available in quantities sufficient for processing it for maximum realization.

The most logical large-scale outlet for char is utilization as boiler fuel for large power plants or industries that consume large quantities of process steam. To make utilization of char as a boiler fuel attractive, the cost per million B.t.u. at the point of consumption must be equal to or less than that of competitive industrial fuels. Where the power plant is close to the mine, char must compete with as mined lignite. Since the char contains only 80 percent of the heating value of the lignite, more lignite is initially required to make available equal amounts of B.t.u. than would be necessary when raw lignite is fired directly. One million B.t.u. is equivalent to 143 pounds of as mined lignite or to 91 pounds of char when the heating values are 7,000 B.t.u. per pound and 11,000 B.t.u. per pound, respectively. Since the yield of char is approximately 50 percent of the weight of the lignite, 182 pounds of raw lignite is required to produce the amount of char equivalent to 1 million B.t.u. Tar credit must pay for the cost of carbonization and for the additional raw-material requirement.

An additional small credit will be obtainable at the boiler plant when char is used because of the reduced tonnage of fuel to be handled and the increased efficiency of the boiler furnaces.

TABLE 30. - Properties of low-temperature primary tars obtained from fluidized carbonization of lignites (12)(24)

Carbonization run No	_	6	_	L1
Coal	Sandow, Tex. 900 1.062 8.1		Garrison, N. Dak. 840 1.054 19.1,	
Town of carbonization				
Spec gravity of dry tar 60°/60°				
Benzene-insoluble in dry tarpercent		Vacuum1/	Atmos 1/	Vacuum1/
	Atmos1/	Varcumi∓)	KUMUS <u>-</u>	* CCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCC
Meximum dist. temp. or initial decomp.		284	250	206
temp. of dry tar2/	310	360	286	280
temp. of dry tar2/do.	346	200	200	200
Distillate to decomp. temp., vol. 70 of	53.6	70.6	42.0	38.8
dry tar	51.7	10.0	TZ.0	JO .U
Distillate analysis, vol. % of dry-				
solids-free tar:4/	4.0	2,3	1,9	1.0
To 170° C	1	3.8	3.1	1.4
170° - 200° C	2.7	3.3	3.9	2.3
200° - 210° C		5.8	9.4	9.2
210° - 235° C	7.5	10.7	13.9	14.4
235° - 270° C	9.2	40.2	5.0	6.0
270° - decomposition	2.9	40.2	4 8	4.5
Residue and loss	2.9	7.	,,,,	
Composition of distillate, vol. %:	29.8	32.2	66.8	64.8
Acids	3.4	4.0	3.2	6.0
Bases	66.8	63.8	30.0	29.2
Neutral oil	00.0	03.0	30.0	
Distillate yield, vol. % of dry-solids-	Ì			
free tar:	15.4	22.7	28.1	25.2
Acids	1.8	2.8	1.3	2.3
Bases	34.5	45.1	12.6	11.3
Neutral cil	J+•/			į
Composition of neutral oil, vol. %:	31.1	59.1	43.3	47.8
Olefins	32.4	24.1	35,4	29.2
Aromatics	36.5	16.8	21.3	23.0
Distillate yield, vol. % of dry-solids-				
Distillate yield, vol. p of dry bolids				
free tar: Olefins	10.7	26.7	5.4	5.4
Aromatics	11.2	10.9	4.5	3.3
Paraffins	12.6	7.5	2.7	2.6
Pitch residue, melting point:	- 1	= /		
Cube in air	2/114	2/147	118	92
Ring and balldo.	110	135	-	01
Specific gravity	1.169	1.160	1.204	1.184

Atmospheric distillation at 620 mm.; vacuum distillation at 40 mm. absolute.

Atmospheric distillation at 620 mm.; vacuum distillation at 40 mm. absoluted 2/ Indicated vapor temp. at decomp. point in first distillation separation.

3/ Second distillation (Hempel) made at 620 mm. on both type distillates.

4/ By Hempel distillation.

5/ In glycerol.

TABLE 31. - Properties of light oil from German brown-coal briquets

010	
Specific gravity at 20° C.	0.851
tar acidsweight	10.1
Distiliate analysis (Engler) vol 4.	
To 60° C.	_ ,
60° - 80° C.	1.4
80° - 100° C.	6.3
100° = 120° a	14.9
100° - 120° C.	29.7
120° - 140° C.	42.5
±+0 - 100 C	60.5
mo = 100 C	73.7
±00 = 200- C	80.0
200°- 220° C	98.1
	30.1
Composition of tar acids, wt. 4:	
Phenol	32.0
o-cresot	17.3
m-cresol	10.4
p-cresol	
Isomeric xylenols	18.0
	22.3

Where char is to be shipped, the question of freight charges enters the picture because of reduced tonnage on an equal B.t.u. basis. However, freight rates applying to char would first have to be known before any shipping advantage can be finally evaluated. Special handling to avoid spontaneous ignition would also have to be considered.

Large-scale consumption of char in form of briquets for domestic fuel use does not seem promising in the light of declining utilization of solid fuels for domestic heating under present conditions.

Other potential applications for lignite char are the manufacture of electrodes and the production of calcium carbide; the latter requires cheap power and availability of limestone. Because of its high reactivity, lignite char should be an efficient agent for reducing ores in processes not requiring a physically coherent coke. It might be used for partial reduction of nonmagnetic taconite as the first step of the magnetic concentration.

Char from brown coal, which is somewhat similar to that from lignite, has been extensively used in Germany to produce synthesis gas and hydrogen. Possible utilization of lignite char is more a question of cost than of quality. Therefore, the future development of low-temperature carbonization of lignite depends to a large extent on profitable use of byproducts, chiefly the tar.

Lignite tar is a suitable source material for manufacturing aromatic hydrocarbons and liquid fuels by the hydrogenation process. Hydrogenation of tar is simpler and requires less hydrogen than direct hydrogenation of coal. Tar acids recovered from processing low-temperature tar are suitable wood preservatives (25). If the more valuable lower boiling tar acids are desired, the high-boiling acids must be catalytically cracked. These acids have been used in lacquers and plastics in Germany (28, p. 37).

The gas produced in internally heated, low-temperature-carbonization processes is commonly consumed in the process itself, which seems to be the most

desirable utilization. Because of its low heating value, pipeline transportation would be uneconomical. Heating value of carbonization gas from internally heated processes approaches that of producer gas.

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