Some Considerations on the Fluidized Carbonization of Coals

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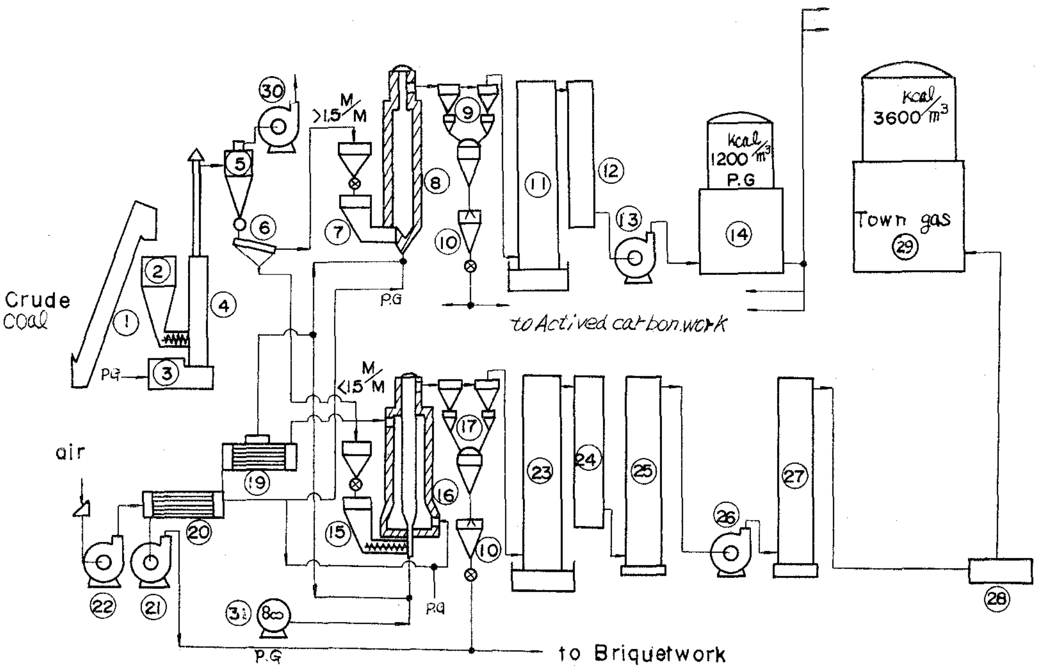
Abstract

It has been attempted to gasify non-cakicg coal, brown coal produced near Kushiro areas, Hokkaido, with the furnace both internally and externally heated by fluidized method on the commercial scale. On the basis of the results of commercial operation, some considerations have been made on relation between the nature of resulting products and the types of fluidized carbonization and on some problems encountered in the course of extending to the region of higher temperature, s. g. 900°C. From the material and heat balance for an inner retort, the heat input from the external chamber is estimated to be only 13.5% of the external heat value required. It seems most reasonable to conclude that in facilities, the heating area of an inner retort is enlarged and the waste heat is utilized as far as possible, in operations, heat transfer is concentrated at the lower parts of an inner retort and the temperature at the top of a combustion chamber is held as low as possible.



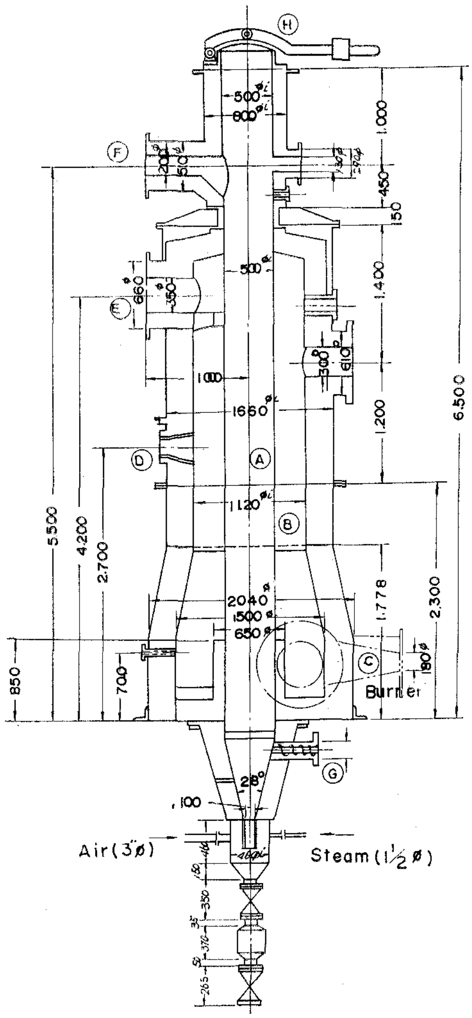
③, ④ : Flash dryer
⑥ : Vibrating screen
⑧ : Internal heating gasification furnace
⑭ : Gas holder
⑯ : Internal and external heating carbonization furnace
⑲ : Waste heat boiler
⑳ : Heat exchanger (air preheating)
㉑ : Waste gas blower
㉙ : Gas holder
㉛ : Roots blower

Fig. 1. Commercial plant (Kushiro, Hokkaido) with both fluidized gasification furnace and fluidized carbonization furnace

1. Introduction

The results of both a pilot and an industrial demonstration plant regarding the fluidized carbonization of non-caking coal, brown coal produced near Kushiro areas, with a furnace both internally and externally heated had been reported before[2),3)], as shown in Fig. 1.

In this paper, avoiding overlap, on the basis of the results of commercial operation, some considerations have been made on relation between the nature of resulting products and the types of fluidized carbonization and on problems occurred in the course of extending to the region of higher temperature.



(A): Carbonizing inner tube
(B): Combustion chamber
(C): Gas burner
(D): Auxiliary burner
(E): Combustion gas outlet
(F): Town gas outlet
(G): Screw feeder
(H): Safety valve

Fig. 2. Fluidized carbonization unit with the furnace both internally and externally heated

2. The nature of resulting products and the types of fluidized carbonization

It is always necessary to get the products that meet the requirement, selecting the types of furnace according to the variety of applications. The furnace structure, operation details and materials used have been reported.

As given in Table 1, the internal heating furnace of type 1 (hereinafter referred to as P_{500}) consists of the cylindrical carbonzing chamber made of refractory bricks (500 mm in inner diameter, 3,500 mm in height) and operating temperatures are the range of 500–600°C (hereinafter referred to as $P_{500,L}$). The internal and external heating furnace of type 2 and 3 (T_{500}) consists of the inner carbonizing retort made of particular stainless steel (6 mm in thick, 500 mm in inner diameter and 5,140 mm in height) encircled with the outer combustion chamber (1,120 mm in inner diameter, 4,300 mm in height). Operating temperatures are the range of 500–600°C (hereinafter referred to as $T_{500,L}$) and the range of 700–850°C (hereinafter referred to as $T_{500,M}$). In either cases, the pulverized coal (0.4–3 mm in size) dried by a flash dryer is introduced into the retort from the lower end by the screw feeder, as shown in Fig. 2 and 3.

The heat value required for carbonization per kg of wet feeding coal, H_c, is divided into 2 parts, namely the heat value generated within the chamber by a fluidizing air, H_i, and the heat value transmited from the outer combustion chamber to the inner retort, H_e. The expression, H_i/H_c is referred to as the coefficient of internal heat, H_e/H_c the coefficient of external heat and H_i/H_e, the ratio of internal heat to external heat respectively. And also the total heat value obtained as gas per kg of wet coal is expressed as (the amount of gas generated Nm^3/kg)×(the combustion heat of gas $kcal/Nm^3$).

For the purpose of producing the coalite having nearly equal calorific value these three types of fluidized furnace described above are operated commercially. It is recognized that there is a certain relation between the nature of products and these types of furnace as illustrated in Table 1. The results are summerized as follows.

1) Although the coalites obtained from these three furnaces are nearly the same in calorific values, significant differences are noticed among the data of proximate analysis. The volatile matter chiefly depends on both carbonization temperatures and the average holding time of granules, independent of the ratio of H_i to H_e. However increasing this ratio resulted in a decrease of the fixed carbon and an increase in the ash content.

2) In the lower temperature carbonization, the considerable amount of tar is produced and combustive properties of resulting gas are not good. An increase of the carbonization temperature and of the holding time will naturally cause the corresponding cracking of the primary resulting tars. The larger the coefficient of external heat, the higher the calorific value of resulting gas.

Table 1. Some types of commercial fluidized carbonization plant and the properties of their products

Results of operation				Types		
				1 $P_{500,L}$	2 $T_{500,L}$	3 $T_{500,M}$
Carbonization condition		Feed rate	kg/hr	900	515	415
		Carbonization temp.	°C	550	560	800
		Mean holding time,	min	2.5	18	43
		Air coal ratio	—	0.367	0.265	0.265
		Steam coal ratio	—	0	0	0.120
Results of carbonization		Gas generated	Nm³/kg	0.406	0.365	0.670
		Coalite	kg/kg	0.62	0.62	0.51
		Tar	,,	0.17	0.17	0.087
		Acquired heat value as gas	kcal/kg	520	600	2,320
		External heat value required	,,	0	350	1,780
		Carbonization heat	,,	302	316	476
		Internal heat	,,	330	236	236
		Input heat	,,	—	80	240
			%	—	23	13.5
		Heat efficiency	,,	85.2	82.7	68.4
Properties of products	Gas	Calorific value	kcal/Nm³	1,270	1,650	3,460
		Specific wt. (air=1)	—	1.03	1.01	0.750
	Coalite	Ash	%	17.7	20.2	21.5
		V. M.	,,	20.9	16.0	6.5
		F. C.	,,	61.4	63.8	72.0
		Calorific value	kcal/kg	6,350	6,410	6,380
	Tar	Acidic oil	%	38.0	—	6.5
		Basic oil	,,	1.0	—	3.5
Note		Per kg wet coal, 0°C base It is given the content % in distillate up to 300°C $P_{500,L}$: Internal heating fluidized furnace at low tempreature $T_{500,L}$: Internal and external heating fluidized furnace at low temperature (inside diameter 500 mm respectively) $T_{500,M}$: Internal and external heating fluidized furnace at intermediate temp. (inside diameter 500 mm)				

3) Increasing the ratio of H_i to H_e resulted in the increase of heat efficiency. Since the average holding time of granules are possible to shorten, the capacity of these furnaces will increase.

3. Some problems occurred in the course of extending to the region of higner temperature

The fluidized gasification operation is a modification of the operation of P_{500}

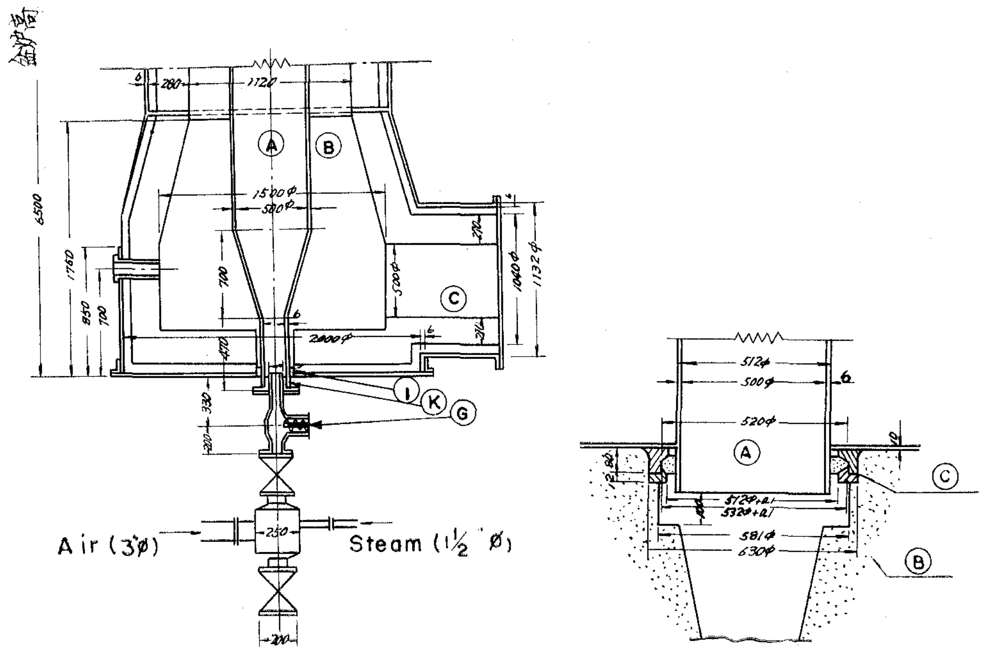
extending to the region of higher temperature and can be obtained a small amount of tar and an activated carbon as by-products. In the purpose of minimizing the tar trouble, practicing the continuous operation for a long time and increasing the heat value obtained as gas, some considerations have been made on some problems occurred in the course of extending to the region of higher temperature, comparing the operation of $T_{500,L}$ with that of $T_{500,M}$.

3-1 Fluidizing agents and the materials of inner retort

It should be selected from among fluidizing agents that are under our hand cheaply. The air is necessary to obtain the internal heat with the increase of carbonization temperature. Using the oxygen increases the calorific value of resulting gas. The steam play an important role in protecting an inner retort as well as controling the combustion of particles and promoting the decomposition of tar. Holding particles are settled at the bottom of inner retort and this part has a danger of becoming the higher temperature. The continuous introduction of steam prevents from rising the temperature owing to the water gas reaction. Perhaps the steam had not been introduced because of lower carbonization temperature in Parry process[4]. The carbonization heat may depend on the external heat alone, using the fluidizing agents such as inert gas, steam and producer gas instead of air. In this case, the quality and the yield of coalite increase, on the other hand, the amount of gas generated and the capacity of furnace decrease.

It is proved that the stainless steel tube (25Cr–12Ni), 6 mm in thick, are possible to practice over two years at the common commercial operation (800°C). Operating at the carbonization temperature of 900°C only for several days (1,150°C at the top of the outer combustion chamber), this tube become brittleness and things of practically no use.

Industrial tests on the inner retort made of carborundum brick (30 mm in thick) have been practiced at higher temperature of 900°C. It was recognized that this material was practicable at the pilot plant (100 mm in diameter of inner tube) before this. It is, however, inpossible to use in the industrial practice owing to such severe operating conditions as described in the next place, in spite of the careful consideration for design, production and operation. The temperature difference between the inside and the outside of this retort at the same cross section amounts to about 400°C and the retort is cracked in places owing to the expansion difference. And the average operating pressure, in practice, is 500 mm in water column within the retort and –50 mm in water column at the outer combustion chamber. The pressure difference between the inside and the outside of thr retort amounts to about 550 mm in water column. This is far beyond the allowable pressure for the circular brick structure. This material is inferior to the metal in heat transfer and moreover it is extremely difficult to seal for expansion, as shown in Fig. 3.



After reconstruction

Ⓐ: Inner retort
Ⓑ: Combustion chamber
Ⓒ: Gas burner
Ⓖ: Screw feeder
Ⓘ: Asbestine packing
Ⓚ: Metal packing

Before reconstruction

Ⓖ: Inner retort
Ⓑ: Brick
Ⓒ: Asbestine packing

Fig. 3. Mechanism of air tight at the lower end of this unit

3–2 The external heat value required Q_e and the calorific value acquired as gas Q_g

The effects of both carbonization temperatures and the rate of feed on the external heat value required and the calorific value acquired as gas (Q_g) are

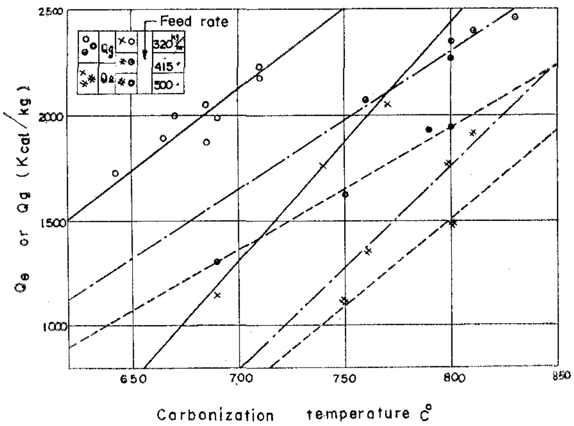


Fig. 4. Relation between carbonization temperature and acquired heat value (Q_g) as gas, external heat value required (Q_e)

illustrated at the operation for the furnace of type 3 in Table 1, as given in Fig 4. The results are summarized as follows.

1) Both the external heat value required for carbonizing the coal to coalite and gas and the calorific value acquired as gas proportionally increase with the increase of carbonization temperatures at the fixed feeding rate. However the increase of Q_e is steeper than the increase of Q_g. The smaller the feed rate, the lower the carbonization temperature that Q_e exceeds Q_g. From these experimental results, in practice, keeping lower carbonization temperatures, making larger load of furnace and higher average concentration of particles are profitable to the operation. Q_e and Q_g proportionally decrease with the increase of the feed rate at the fixed carbonization temperature.

3-3 Analysis of the external heat value required, Q_e

The external heat value required, Q_e is 1,780 kcal per kg of wet coal at the carbonization temperature of 800°C and the feed rate of 415 kg/hr as given in Fig 4. Among these, the heat value transmited into the inner tube, H_e, is attempted to calculated from the heat and material balance and the several equations of heat transfer.

On the one hand, the temperature of several points within the furnace is measured at the normal operation and the overall coefficient of heat transfer to fluidized bed from the outer combustion chamber is estimated to be 41.5 kcal/m·hr·°C, H_e corresponding to this is estimated to be 240 kcal. On the other hand, from the material and heat balance for an inner tube as given in Table 2, H_e is estimated to be 240 kcal. Good agreement between these two methods is

Table 2. Material and heat balance for fluidized carbonization with the furnace both internally and externally heated

(Per kg of wet feed coal, 0°C base)

		°C	Nm³	kg	Sensible and latent heat kcal	%
Heat input	Coal (wet)	50	—	1	14.0	2.4
	Air	20	0.265	0.328	2.0	0.3
	Steam	105	—	0.120	78.0	13.7
	Combustion heat, H_i	—	—	—	236.0	41.4
	External heat, H_e	—	—	—	240.0	42.2
	Total	—	—	1.448	570.0	100.0
Heat output	Coalite	800	—	0.51	162.0	28.5
	Gas	800	0.67	0.648	198.0	34.7
	Tar	800	—	0.087	29.3	5.2
	Steam	800	—	0.183	180.7	31.6
	Unaccounted	—	—	0.020	—	—
	Total	—	—	1.448	570.0	100.0

Table 3. Analyse of external heat value required
(Per kg of wet feed coal, 0°C base)

Item	Kcal	%
Heat input from internal retort H_e	240	13.5
Heat loss from external wall	106	6.0
Other heat loss	1434	80.5
total	1780	100.0

recognized. The heat loss from the outer wall and the other heat loss are calculated respectively as shown in Table 3. According to these analyse, the heat input from the external retort, H_e, is estimated to be only 13.5% of the external heat value required, Q_e. This value for the furnace of type 2 in Table 1 is estimated to be 23% by a method similar to that described above. One may conclude that the heat input from the external retort, H_e, decreases with the increase of carbonization temperatures.

In view of the above facts it seems most reasonable to conclude that in facilities, the heating area of inner tube is enlarged and the waste heat is utilized as far as possible, in operations, heat transfer is concentrated at the lower parts of inner retort and the temperature at the top of combustion chamber is held as low as possible.

4. Conclusion

It had been attempted to gasify non-caking coal, brown coal produced near Kushiro areas, by fluidized method on the commercial scale. From a consideration of some problems encountered in the course of commercialization it may be concluded as follows.

1) On the basis of the operating results for three different types of fluidized carbonization, calorific value of resulting coalite being similar to each other, it is pointed out that there is a certain relation between the type of carbonization and the property of resulting products.

2) In the external and internal heating fluidized carbonization of coal, extending to the region of higher temperatures resulted profitably in reducing the tar trouble and also increasing the heat amount acquired as gas. However, on the other hand, the increase of Q_e is more than the increase of Q_g with increasing of carbonization temperatures. It is recognized that the fluidized carbonization at the higher temperature is at a disadvantage thermally.

3) In view of the above results it seems most reasonable to conclude that in facilities, the heating area of inner tube is enlarged and the waste heat is utilized as far as possible, in operations, heat transfer is concentrated at the lower parts of inner tube and the temperature at the top of combustion chamber is held as low as possible.

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For reference

In order to calculate the overall coefficient of heat transfer, the following equation are used.

1) Coefficient of heat transfer h_1 to the outside wall of inner tube from outer combustion chamber

$$h_1/C_p G\cdot(C_p\mu/\lambda)^{2/3} = 0.0234/(D_e G/\mu)^{0.2}$$

h_1: coefficient of heat transfer		kcal/m²·°C·sec
C_p: specific heat under constant pressure		0.302 kcal/kg·°C
G: mass velocity $(=\rho u)$		0.546 kg/m²·sec
λ: thermal conductivity		1.77×10^{-5} kcal/m·sec·°C
μ: viscosity coefficient		4.70×10^{-5} kg/m·sec
D_e: equivalent diameter D_2-D_1	D_2: inside dia. of outer tube	1.12 m
	D_1: outside dia. of inner tube	0.512 m

$h_1 = 3.51$ kcal/m²·hr·°C

2) Coefficient of radiant heat transfer to the outside wall of inner tube from outer combustion chamber h_2

Non-luminus flame radiation encircled by the insulated reflective surface

$$Q = 4.96\left[(T_G/100)^4-(T_1/100)^4\right]F \qquad F_{R_1} = A_1/A_R \text{ angle factor}$$

$$A_1 F = \frac{x}{\frac{1}{A_1}\left(\frac{1}{\varepsilon_1}-1\right)+1\Big/\frac{E_G}{x}\left(A_1+\frac{A_R}{1+\frac{\varepsilon_G/x}{1-\varepsilon_G/x}\cdot\frac{1}{F_{R_1}}}\right)}$$

$x = E_G^2/(2\varepsilon_G-\varepsilon_{2G})$ 0.187

ε 0.165

A_1: surface area heated		6.93 m²
A_R: insulated reflective surface		15.1 m²
F: coeficient of radiant heat transfer (overall)		0.1402
E_1: blackness of surface heated 0.8	$Q=12{,}600$ kcal/m²·hr	
E_G: blackness of non-luminous flame 0.123	$h_2=12{,}600/(1120-910)=60$ kcal/m²·hr·°C	

3) Coefficient of heat transfer h_3 to fluidized bed from inside wall of inner tube (Walton and Levenspiel equation)

$$h_3/C_p G = 0.6(D_p G/\mu)^{-0.7}$$

D_p: mean particle size	0.5 mm
h_3: coeficient of heat transfer	kcal/m²·sec·°C
D: inside diameter of inner tube	0.5 m
λ: thermal conductivity of gas	1.87×10 kcal/m·sec·°C
G: mass velocity of gas	0.499 kg/m²·sec
μ: viscosity coefficient of gas	4.04×10^{-5} kg/m·sec
C: specific heat under constant pressure	0.411 kcal/kg·°C

$h_3=124$ kcal/m²·hr·°C

4) Overall coefficient of heat transfer h_c to fluidized bed from the outer combustion chamber

$$1/h_c = 1/(h_2+h_1)+d/\lambda+1/h_3$$

d: thickness of stainless tube	6 mm
λ: thermal conductivity of stainless tube	24.5 kcal/m·hr·°C

$h_c=41.5$ kcal/m²·hr·°C

5) Heat amount transmitted to the inside of inner tube per kg of wet feed coal (H_e)

feed rate 415 kg/hr

load of furnace per heated surface m^2 $415/6.93 = 60$ kg/m^2·hr

$H_e = 41.5\,(1120-800)/60 = 220$ kcal/kg

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