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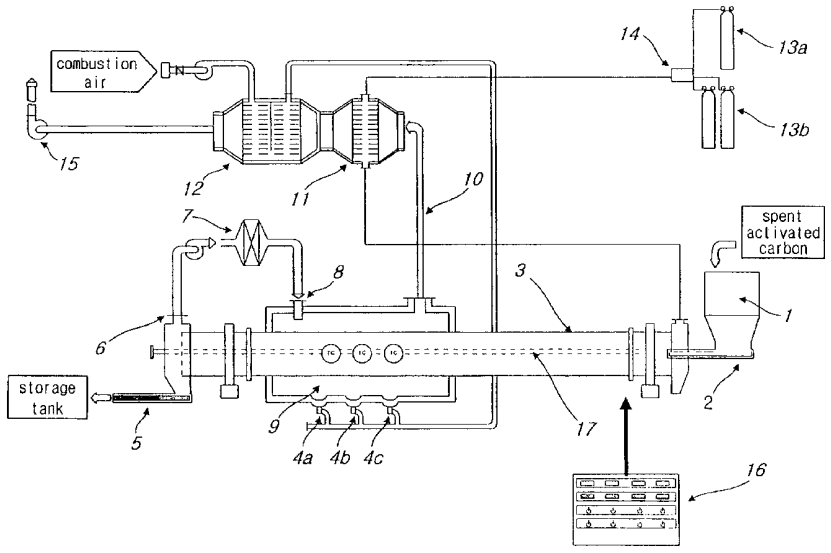
(54) Title: REGENERATION PROCESS AND FACILITIES OF SPENT ACTIVATED CARBONS



(57) Abstract: This invention relates to a regeneration process and facilities of spent activated carbons, the process being characterized by pyrolyzing in the low temperature range of between 200 °C and 800 °C, the facilities by comprising 1) a feeding section; 2) a reaction section in which pollutants attached to the spent activated carbons are removed by heat treatment, and which comprises a rotary kiln and a heating chamber, the heating chamber being located in the middle or downstream part of the rotary kiln and able to indirectly heat up the spent activated carbon traveling through the rotary kiln by heating its shells from outside; 3) a cooling section; and 4) CO_2 or CO_2 mixture gasstorages/supply section. The invention will prevent the loss and adsorption capacity reduction of activated carbon, will facilitate the improvement of hardness, recovery rate, and restoration of the adsorption capacity, and will enable the simplification of the facilities.

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REGENERATION PROCESS AND FACILITIES OF SPENT ACTIVATED CARBONS

FIELD OF THE INVENTION

This invention relates to a method and apparatus for regenerating spent activated carbon spent and discharged from water purification process, waste water treatment process, or petrochemical process where the activated carbon is used to recover solvent or high priced chemicals and so on, and more particularly, to a method and apparatus involving indirect heating and pyrolysis at the temperature range between 200℃ and 800℃ utilizing carrier gas containing carbon dioxide.

BACKGROUND OF THE INVENTION

In most of the conventional regeneration of activated carbon, the production process of new activated carbon was utilized involving high pressure steam and very high temperature exceeding 1000℃ in rotary kilns or multi-hearth furnaces. Moreover, regeneration was done without differentiating adsorbates characteristics, which results in such serious problems as the weight loss of the activated carbon, reduced hardness and adsorption quality of the activated carbon both caused by excessive oxidation and subsequent destruction of surface pores. To remedy such problems in thermal regeneration, this inventor previously applied for a patent for a method and apparatus for

regenerating spent activated carbon involving indirect heating utilizing heated air (Korean Patent Application No.: 10-1999-0021994) and followed it up with another patent application after verifying that using carbon dioxide as a carrier gas for regeneration of spent activated carbon improves the regeneration efficiency (Korean Patent Application No. 10-1999-43104). This invention intends to improve over the two previous applications and improve the efficiency in commercial application.

In order to rectify the weight loss and the drop of adsorption capacity in the process of thermal regeneration of spent activated carbons, there has been a lot of research and development in the field of biological regeneration and chemical regeneration. In the case of biological regeneration, however, it turned out that due to the long duration time required for the cultivation of proper microbes and their stabilization and also due to the fact that matters that can be decomposed by microbes are limited, it is neither economically viable nor technically practicable. In the case of chemical regeneration, there has been quite a few attempts for preliminary research. However, due to the cost of solvents, the trouble associated with the disposal of the residual waste organic solvent and the reduced adsorption capacity caused by the adsorption of the solvent during the regeneration process, these efforts could not be commercialized.

DETAILED DESCRIPTION OF THE INVENTION

As stated above, this invention intends to provide a method and apparatus for

regenerating spent activated carbon facilitating the improvement of adsorption capacity and recovery rate, and the prevention of the loss in the hardness of the activated carbon overcoming the deficiency in the thermal regeneration of the spent activated carbon by conventional technologies.

On top of that, it is also intended to provide a method that can improve the regeneration efficiency by utilizing mixed carrier gases and regulating moisture content of the spent activated carbon before the regeneration.

In order to achieve the above mentioned goals, the method of regenerating spent activated carbon by this invention, in its regeneration process which includes drying step and pyrolysis step, is characterized by desorption of pollutants from activated carbon during the above drying step and pyrolysis step after being gradually heated by indirect heat transfer, and also by addition of cooling step wherein hot activated carbon regenerated in the preceding pyrolysis step is cooled.

Moreover, the regeneration apparatus of spent activated carbon by this invention is characterized by inclusion of feeding section of spent activated carbon (1,2); reaction section (3.9); and carrier gas supply section (13, 14). Above feeding section (1,2) introduces spent activated carbon into the apparatus. Above reaction section (3,9) wherein spent activated carbon is heated to desorb pollutants from it, is further characterized by inclusion of a rotary kiln (3) through which spent activated carbon is

transported and a combustion chamber (9) in which the lower middle portion of the above mentioned rotary kiln is externally heated to indirectly heat the spent activated carbon traveling inside the rotary kiln.

In the above mentioned regeneration method, the carrier gas supply section(13, 14) supplies carrier gases to the above mentioned reaction section.

In the above mentioned cooling section (5), activated carbon heated in the above mentioned reaction section is cooled down.

The above mentioned regeneration method and apparatus of spent activated carbon is also characterized by gradually heating the above mentioned spent activated carbon to the temperature between 200℃ and 800℃.

It is further characterized by utilizing carbon dioxide, mixture of carbon dioxide and oxygen, or mixture of carbon dioxide and nitrogen as its carrier gas.

It is further still characterized by regulating the moisture content of the spent activated carbon according to the characteristics of the adsorbates in the spent activated carbon when introduced at the feeding section.

It is further still characterized by treating the hot gas produced in the pyrolysis step by

directing it back to the combustion chamber.

It is further still characterized by utilizing the above mentioned hot flue gas to preheat the above mentioned carrier gas and the combustion air for the above mentioned pyrolysis section.

It is further still characterized by cooling down the spent activated carbon processed through the reaction section.

BRIEF DESCRIPTION OF THE DRAWING

Figure 1 illustrates the apparatus by this invention for regenerating the spent activated carbon.

<Description of Major Parts>

1: Spent Activated Carbon Input Tank

2: Screw Conveyor

3: Rotary Kiln

4a, 4b, and 4c: Burners

5: Cooling Section

6: Exhaust Outlet of Process Gas

7: Dust Removal Facility

8: Inlet of Process Gas into the Combustion Chamber

9: Combustion Chamber

10: Outlet of Flue Gas

11: Primary Heat Exchanger

12: Secondary Heat Exchanger

13a, 13b: Carrier Gas Vessels

14: Gas Mixing tank

15: Exhaust Blower

16: Control Panel

17: Temperature Sensor

BEST MODE FOR CARRYING OUT THE INVENTION

This invention relates to a method and apparatus for regenerating spent activated carbon utilizing carbon dioxide as the main carrier gas and pyrolysis process. The type of the regeneration furnace is rotary kiln. Instead of directly heating the inside of the furnace with burner flame as in the conventional processes, the external shell of the rotary kiln is heated by burners and spent activated carbon traveling within the kiln is indirectly heated by heat transfer. The apparatus by this invention involves a combustion chamber provided around the lower middle section of the rotary kiln. Carrier gas of low temperature containing carbon dioxide is introduced into the reaction section of the kiln,

which facilitates the gradual drying of the spent activated carbon over a certain period of time, which results in the prevention of the rapid temperature rise of the spent activated carbon, and thus minimization of adsorbates turning into char. Cooling facility is provided at the end of the sealed moving devices to prevent regenerated hot activated carbon from oxidation by contacting external atmosphere.

In most of the conventional thermal regeneration processes, spent activated carbon is exposed directly to the bare flame from burners and gasification reaction in which highly oxidative steam is used. These result in the loss of hardness and recovery rate of the activated carbon. Gasification method creates additional pores in the activated carbon and causes the weakening of pore structure and fails to achieve complete removal of the adsorbates from the activated carbon.

Conversely, this invention takes care of these problems by utilizing carbon dioxide as carrier gas; by controlling the moisture content of the spent activated carbon before feeding them into the apparatus; and by regenerating spent activated carbon utilizing pyrolysis process that enables the selective disintegration and removal of pollutants from the spent activated carbon at a temperature of 800℃ or lower. This ensures high recovery rate preventing weight loss and reduction of hardness.

In the conventional apparatuses, air pollution prevention device like a secondary combustion chamber is necessary to treat the toxic flue gas, but not in the case of this

invention in which toxic flue gas from the reaction is introduced back into the combustion chamber and completely burnt in the direct contact with flame that heats up the shell of the kiln. And moreover, a heat exchanger is provided to retrieve energy from the flue gas and preheat the carrier gas and the combustion air. This makes this invention highly efficient in the field of energy saving, too. The regeneration apparatus by this invention distances itself from conventional ones that utilize direct heating method and steam as its oxidation agent, and has the advantage of efficient regeneration with the help of simplified and automatized facilities.

The regeneration method by this invention is characterized by improving regeneration efficiency by applying proper reaction temperatures and retention time according to specific adsorbates; using either carbon dioxide, mixture of carbon dioxide and oxygen, mixture of carbon dioxide and nitrogen, nitrogen or air as its carrier gas; maintaining the carrier gas velocity at Reynolds number 500 or less; and controlling moisture content in accordance with specific adsorbates in the spent activated carbon.

Reference is now made to the attached drawing to provide a detailed description of the method and apparatus by this invention for regenerating spent activated carbon.

Fig. 1 is a system diagram of the regeneration apparatus for spent activated carbon by this invention.

Compared with conventional ones, the regeneration apparatus by this invention places emphasis on, firstly prevention of loss and reduction of hardness of the regenerated activated carbon, secondly prevention of loss of adsorption capacity due to pore destruction, thirdly simplification of the facilities by the elimination of the secondary combustion chamber utilizing the primary combustion chamber for the prevention of air pollution, and fourthly energy saving through the utilization of heat exchangers.

Following is a detailed description of the system of the spent activated carbon regeneration apparatus by this invention

Spent activated carbon from the input tank (1) is fed into the shell of the kiln (3) by the screw conveyer (2). The shell of the kiln is heated by burners (4a, 4b, 4c) properly positioned to prevent rapid temperature rise of the spent activated carbon and maintain a gradual temperature curve in the kiln. Burners (4a, 4b, 4c) are controlled by temperature readings by the sensors (17) installed inside the kiln (3) shell. Heat indirectly transferred through the shell is used as the energy source to remove the moisture and volatile and involatile matters adsorbed on the spent activated carbon. Regenerated activated carbon in the kiln is then transported through cooling system to cool it to a temperature, low enough for contact with the atmosphere in the storage tank.

Gas generated in the process is discharged through the exhaust outlet (6) and introduced into the combustion chamber (9) through its inlet (8) opening after it gets dedusted by

the dust removal facilities (7). In this step, the process gas from the rotary kiln is exposed to the direct contact with the flame of the burners (4a, 4b, 4c) and completely burnt. Flue gas with its toxic ingredients completely removed is then transported to the primary heat exchanger (11) through the outlet (10) to heat up the carrier gas and then transported again to the secondary heat exchanger (12) to impart its heat to the combustion air for the burners (4a, 4b, 4c). Gas mixing tank (14) is used to mix carrier gases stored in the carrier gas vessels to such a proportion as to maintain selective oxidative atmosphere in the rotary kiln.

Temperatures of the burners (4a, 4b, 4c), feeding rate of spent activated carbon and r.p.m. of the rotary kiln are controlled by preset figures in the PID controller of the control panel(16), which enables the automatic operation of the apparatus.

Following is a typical embodiment example of the spent activated carbon regeneration method using the apparatus by this invention:

Activated carbon used for solvent recovery for a period of 1 year in a petrochemical process was taken as the test sample. An apparatus as illustrated on Fig.1 was used. Internal temperature inside the rotary kiln (13) was controlled to maintain a temperature curve between 200℃ and 600℃ with the reaction time of 50 minutes. Mixture of carbon dioxide and oxygen was used in the following proportions by volume: $CO_2(9):O_2(1)$, $CO_2(8):O_2(2)$, $CO_2(7):O_2(3)$, $CO_2(5):O_2(5)$.

Table 1 shows the result of this test expressed in the iodine adsorption capacity, apparent density and hardness. Testing method of KSM 1802 was used. Comparison was made with new virgin activated carbon and regenerated activated carbon produced by a local regenerator using the same test sample. Their process uses steam at a high temperature (1000℃ or more) with direct firing.

Table 1

	New carbon	Activated carbon Regenerated according to the Example				Comparative Example(regenerated carbon by other company
		CO_2:O_2 =9:1	CO_2:O_2 =8:2	CO_2:O_2 =7:3	CO_2:O_2 =5:5	
Iodine Adsorption Capacity	1151	1072	1095	1120	1070	993
Apparent Density	0.45	0.44	0.42	0.41	0.41	0.47
Hardness(%)	99.0	99.0	98.2	98.2	95.2	84.4

As shown in the above table, regeneration by this invention produced much better results than the conventional process and mixture of carbon dioxide and oxygen at the rate of 7:3 proved to be the best. Therefore, it is obvious that using a mixture of carbon dioxide and oxygen at a proper rate as a carrier gas is more suitable for regeneration than the

conventional process using steam at a high temperature. However, as the oxygen content in the carrier gas increases, oxidation rate is increased and it results in the loss of hardness. Therefore, it is important to decide proper oxygen content suitable for different spent activated carbon.

Another test was made to verify the improvement of regeneration efficiency by regulating moisture. Its result is shown in the table 2.

The same apparatus as in the previous test was used. Activated carbon used for a period of 1 year for the treatment of chemical agents in a petrochemical process was taken as the test sample in this case. Temperature in the kiln (3) was controlled to maintain a curve between 200℃ and 600℃ with the retention time of 50 minutes. To study the effects of different moisture contents, carbon dioxide was selected as the lone carrier gas, with the moisture content varying at 20%, 30% and 40%. Regeneration efficiency expressed in iodine adsorption capacity, apparent density and hardness was tested in accordance with KSM 1802. Comparison was made with the results obtained from new virgin sample and the same spent activated carbon sample completely dried before the test.

Table 2

	New carbon	Dried sample by this Invention	Sample with 20% moisture	Sample with 30% moisture	Sample with 40% moisture
Iodine Adsorption Capacity	998	955	976	989	962
Apparent Density	0.51	0.52	0.51	0.51	0.52
Hardness(%)	97.8	96.5	96.2	96.0	96.0

As can be seen from the table 2 above, as the moisture content increases up to a certain range, regeneration efficiency improves, too. The moisture adsorbed on the activated carbon is thought to be transformed to steam, which in turn dissolves the pollutants in the activated carbon. Therefore, it is obvious that adding moisture to a proper content helps to improve the regeneration efficiency. However, in case the moisture content exceeds 30%, some residual moisture in the pores reduces the iodine adsorption capacity and increases the apparent density. Therefore, it is important to run enough tests to decide a suitable moisture content for the best regeneration.

As discussed and proven in the above, the regeneration method by this invention is different from the conventional gasification method wherein spent activated carbon is reactivated by directly exposing them to high temperature heat and steam. And more

particularly, it provides much better results than the conventional gasification method in the hardness, recovery rate and adsorption capacity of the activated carbon, as pyrolysis and indirect heating are used to selectively desorb only the pollutants from the spent activated carbon at a temperature range lower than 800℃ without causing the loss of activated carbon and reduction of the adsorption capacity, and as properly mixed carrier gas facilitates the selective oxidative atmosphere. And the secondary combustion chamber required for the prevention of air pollution in the case of conventional method can be eliminated by putting the pyrolysis gas back into the main combustion chamber.

What is claimed is :

1. A method for regenerating spent activated carbon including a drying step and a pyrolysis step, which is characterized by desorbing pollutants from spent activated carbon through gradual drying and pyrolysis by indirect heat transfer.

2. The method for regenerating spent activated carbon according to claim 1, wherein the drying step and pyrolysis step are characterized by the gradual heating of the spent activated carbon to the temperature range between 200℃ to 800℃.

3. The method for regenerating spent activated carbon according to claim 1, which is characterized by the fact that gas containing carbon dioxide is utilized as a carrier gas.

4. The method for regenerating spent activated carbon according to claim 3, wherein the gas containing carbon dioxide is one selected from the group consisting of carbon dioxide, mixture of carbon dioxide and oxygen, and mixture of carbon dioxide and nitrogen.

5. The method for regenerating spent activated carbon according to claim 1, which is characterized by the fact that moisture content of the spent activated carbon introduced to the drying step is regulated according to the characteristics of adsorbates.

6. The method for regenerating spent activated carbon according to claim 1, which is characterized by further comprising a flue gas treatment step for the combustion of the hot flue process gas produced in the pyrolysis step by means of the indirect heating of the drying and pyrolysis steps.

7. The method for regenerating spent activated carbon according to claim 6, that is characterized by further comprising a step that preheats the carrier gas and combustion air of the pyrolysis step by using the hot flue gas processed through the flue gas treatment step.

8. The method for regenerating spent activated carbon according to claim 1, which is characterized by further comprising a cooling section that cools down the spent activated carbon processed through the reaction section.

9. An apparatus for regenerating spent activated carbon, which is characterized by comprising:

a) a feeding section through which spent activated carbon is introduced;

b) a reaction section that desorbs pollutants by heating the introduced spent activated carbon, which is characterized by comprising a rotary kiln through which spent activated carbon travels and a combustion chamber that indirectly heats the spent activated carbon

traveling through the rotary kiln by heating the outside of the rotary kiln in the middle and lower part; and

c) a carrier gas supply section that supplies carrier gas to the reaction section.

10. The apparatus for regenerating spent activated carbon according to claim 9, wherein the reaction section is heated gradually to the temperature range between 200℃ and 800℃.

11. The apparatus for regenerating spent activated carbon according to claim 9, which is characterized by the fact that the carrier gas is one selected from the group consisting of carbon dioxide, a mixture of carbon dioxide and oxygen, and a mixture of carbon dioxide and nitrogen.

12. The apparatus for regenerating spent activated carbon according to claim 9, which is characterized by the fact that the moisture content of the spent activated carbon at the feeding section is regulated according to the nature of adsorbates.

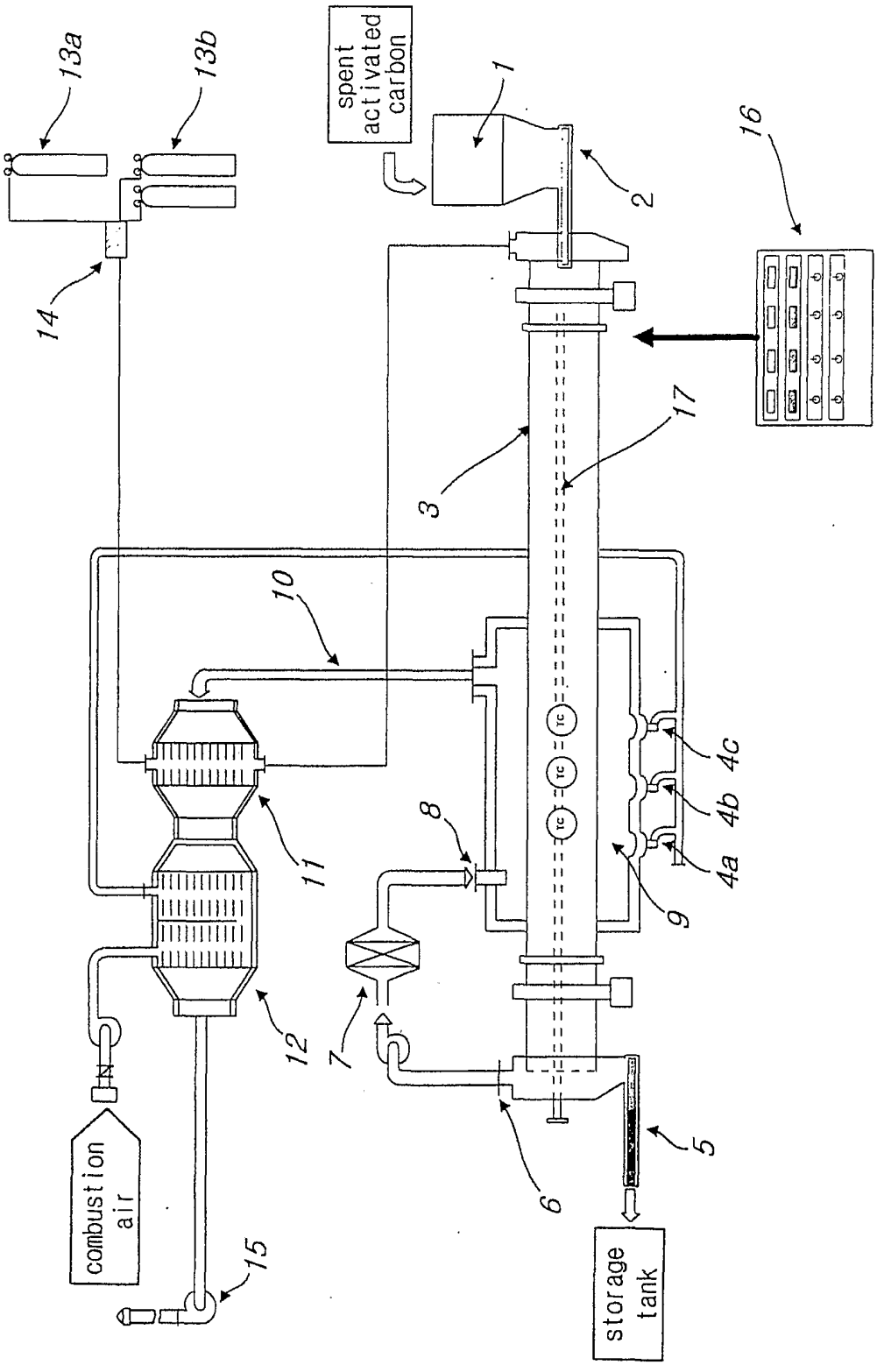
13. The apparatus for regenerating spent activated carbon according to claim 9, which is characterized by further comprising a flue gas treatment section which treats the flue process gas discharged from the reaction section by putting the gas back into the combustion chamber for combustion.

14. The apparatus for regenerating spent activated carbon according to claim 13, which is characterized by further comprising a heat exchange section that preheats the carrier gas and combustion air using the heat of the flue gas processed through the flue gas treatment section.

15. The apparatus for regenerating spent activated carbon according to claim 9, wherein the rotary kiln of the reaction section is equipped with temperature sensors inside.

16. The apparatus for regenerating spent activated carbon according to claim 9, which is characterized by further comprising a cooling section that cools down the spent activated carbon processed through the reaction section.

Fig.1


13a
13b
spent activated carbon
1
2
16
14
3
17
10
11
8
4c
4b
4a
9
12
7
6
5
combustion air
15
storage tank

INTERNATIONAL SEARCH REPORT

International application No.
PCT/KR01/01818

A. CLASSIFICATION OF SUBJECT MATTER

IPC7 B01J 20/30, B01J 20/34

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC7 B01J 20/30, B01J 20/34, C01B 31/08

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched
Korean Patents and applications for inventions since 1975 Korean Utility models and applications for Utility models since 1975
Japanese Utility models and applications for Utility models since 1975

Electronic data base consulted during the intertnational search (name of data base and, where practicable, search terms used)
NPS, PAJ, USP "activated carbon, reproduction, regeneration, rotary kiln"

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	JP 52-37597 A (HITACHI PLANT ENG & CONSTR CO., LTD.) 23 MARCH 1977 See Claim; Page 2, line 7 - line 12 (the right side)	1, 2, 3, 4
Y	JP 11-188259 A (UNION SERVICE:KK, KASAHARA TAKASHI) 13 JULY 1999 See Abstract; Claims; Figure 1	1, 2, 6
Y	JP 51-119697 A (NIPPON FURNACE KOGYO KK) 20 OCTOBER 1976 See Claims	1, 6, 8
A	JP 52-792 A (ASAHI DOW CO., LTD.) 06 JANUARY 1977 See Claim; Page 2, line 8 - line 18 (the left side); Figure 2	1, 2, 3
A	KR 90-4396 A (CHOE, WAN-SUN) 12 APRIL 1990 See Claims; Figure 2	1, 6

☐ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

* Special categories of cited documents:
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"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
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