

[72] Inventors Akira Toyama
Kobe-shi;
Yukio Nakako, Nishinomiya-shi; Sakayuki
Nakanishi, Kobe-shi; Hiroetu Miki, Akashi-
shi, all of, Japan

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[73] Assignee Kobe Steel, Ltd.
Kobe-shi, Japan

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[33] Japan

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Primary Examiner—Reuben Friedman
Assistant Examiner—R. W. Burks
Attorney—Craig, Antonelli, Stewart and Hill

ABSTRACT: The air refining separation procedure, comprising, by using both the moisture-carbon dioxide absorbing apparatus consisting of the multiple fixed beds, which have two layers of a moisture absorbing layer and a carbon dioxide absorbing layer, where four processes of absorbing, heating regenerating, and cooling are repeated in turn, and the nitrogen absorbing apparatus consisting of the multiple fixed beds, where two processes of absorbing and regenerating are repeated in turn: (a) supplying the raw air, to obtain the refined air, into the fixed beds, which is in process of adsorption, of the said moisture-carbon dioxide absorbing apparatus; (b) subsequently supplying the refined air, to obtain the oxygen of low purity, into the fixed bed, which is in process of adsorption, of the said nitrogen absorbing apparatus; (c) supplying a part of the product oxygen of low purity into the fixed bed, which is in process of regeneration, of the said moisture-carbon dioxide absorbing apparatus, and subsequently recovering it as the product oxygen of low purity from the air refining separation apparatus; (d) supplying the rest of the oxygen of low purity into the fixed bed, which is in process of cooling, of the said moisture-carbon dioxide absorbing apparatus, where it is cooled and dehydrated, and then joining it with the low purity oxygen flow issued from the nitrogen adsorbing apparatus; and (e) supplying the nitrogen which desorbs from the fixed bed, which is in process of regeneration, of the said nitrogen adsorbing apparatus, after heating, into the fixed bed, which is in process of heating, of the said moisture-carbon dioxide absorbing apparatus, and subsequently discharging it out from the air refining separation apparatus.

[54] **REFINING SEPARATION PROCEDURE OF OXYGEN FROM AIR**
1 Claim, 10 Drawing Figs.

[52] U.S. Cl. 55/33, 55/68

[51] Int. Cl. B01d 53/03

[50] Field of Search 55/28, 29, 33, 68, 74, 76, 179, 180

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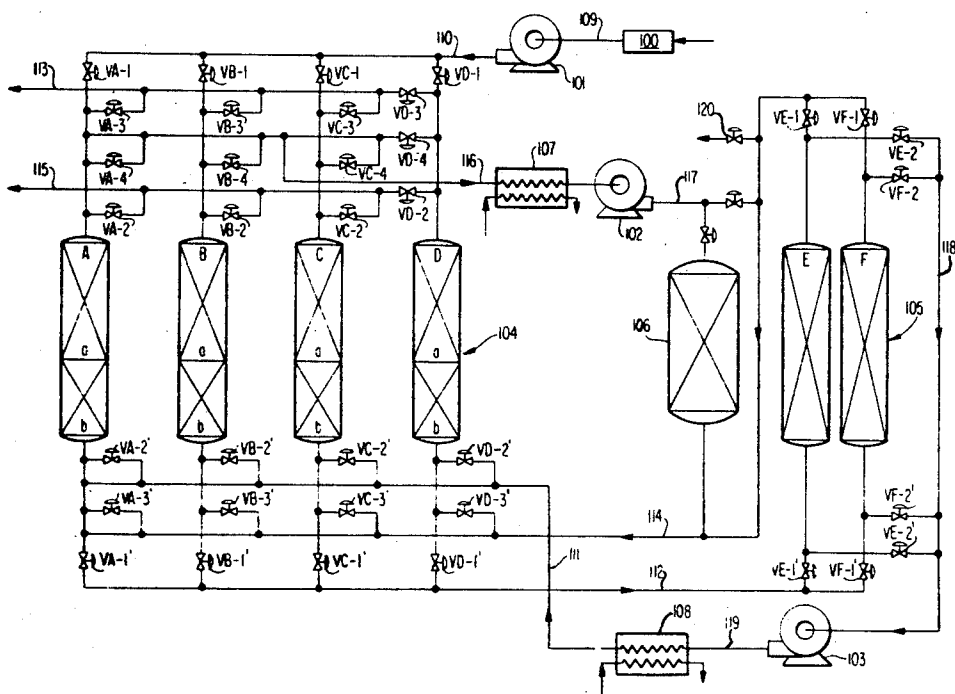
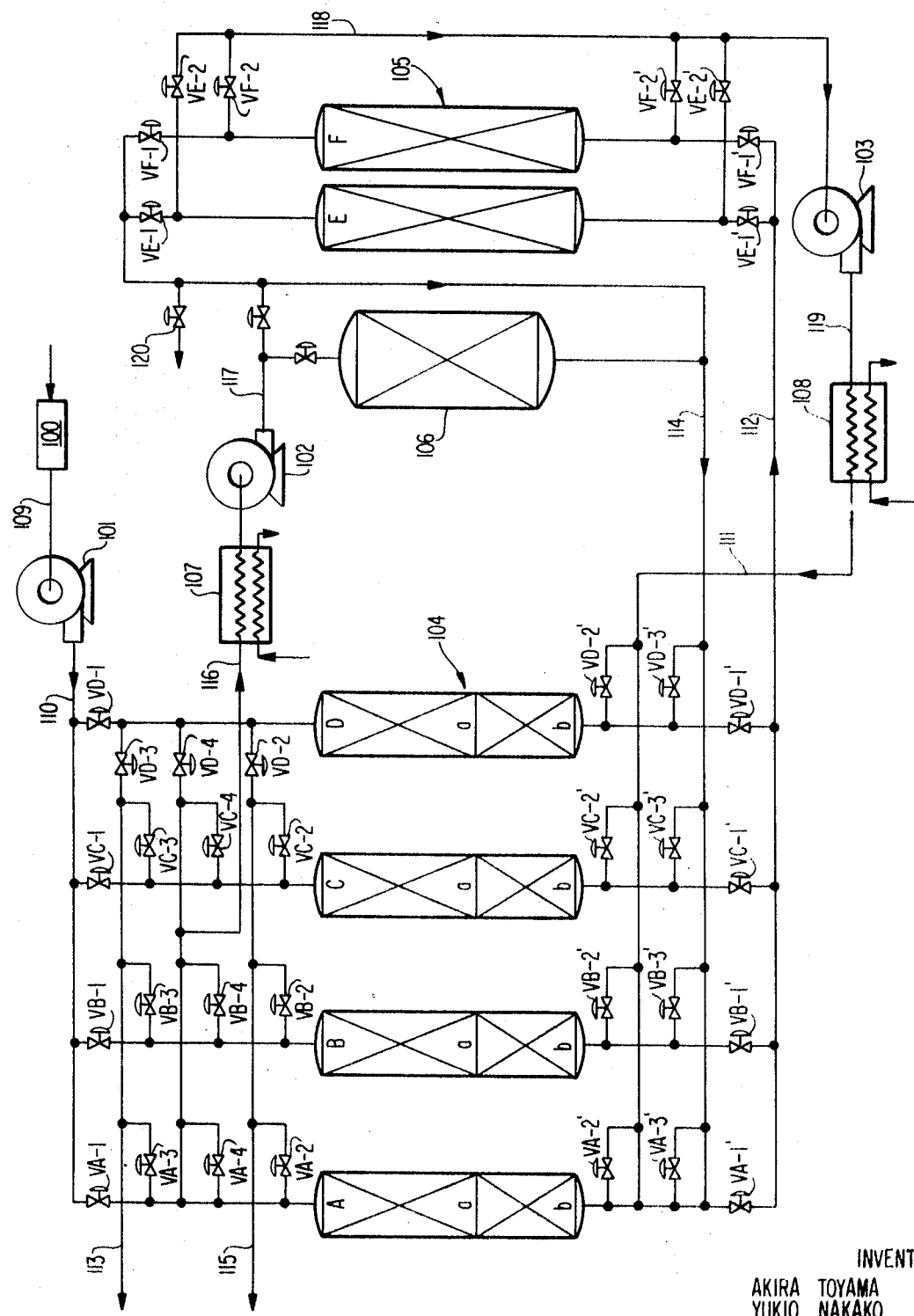


FIG. 1



INVENTORS
AKIRA TOYAMA
YUKIO NAKAKO
SADAYUKI NAKANISHI
HIROETU MIKI

BY
Craig, Antonelli, Stewart & Hill

ATTORNEYS

AD-SORBER	CYCLE	FIRST CYCLE				SECOND CYCLE			
	ORDER OF SWITCHING	1	2	3	4	1	2	3	4
	ELECTRO-MAGNETIC VALVE NO.								
A	VA-1	AD-				AD-			
	VA-1'	SORBING				SORBING			
	VA-2		HEATING				HEATING		
	VA-2'								
	VA-3			REGEN-				REGEN-	
	VA-3'			ERATING	COOLING			ERATING	COOLING
	VA-4								
B	VB-1		AD-				AD-		
	VB-1'		SORBING				SORBING		
	VB-2			HEATING				HEATING	
	VB-2'								
	VB-3				REGEN-				REGEN-
	VB-3'	COOLING			ERATING	COOLING			ERATING
	VB-4								
C	VC-1			AD-				AD-	
	VC-1'			SORBING				SORBING	
	VC-2				HEATING				HEATING
	VC-2'								
	VC-3	REGEN-				REGEN-			
	VC-3'	ERATING	COOLING			ERATING	COOLING		
	VC-4								
D	VD-1				AD-				AD-
	VD-1'				SORBING				SORBING
	VD-2	HEATING				HEATING			
	VD-2'								
	VD-3		REGEN-				REGEN-		
	VD-3'		ERATING	COOLING			ERATING	COOLING	
	VD-4								

FIG. 2

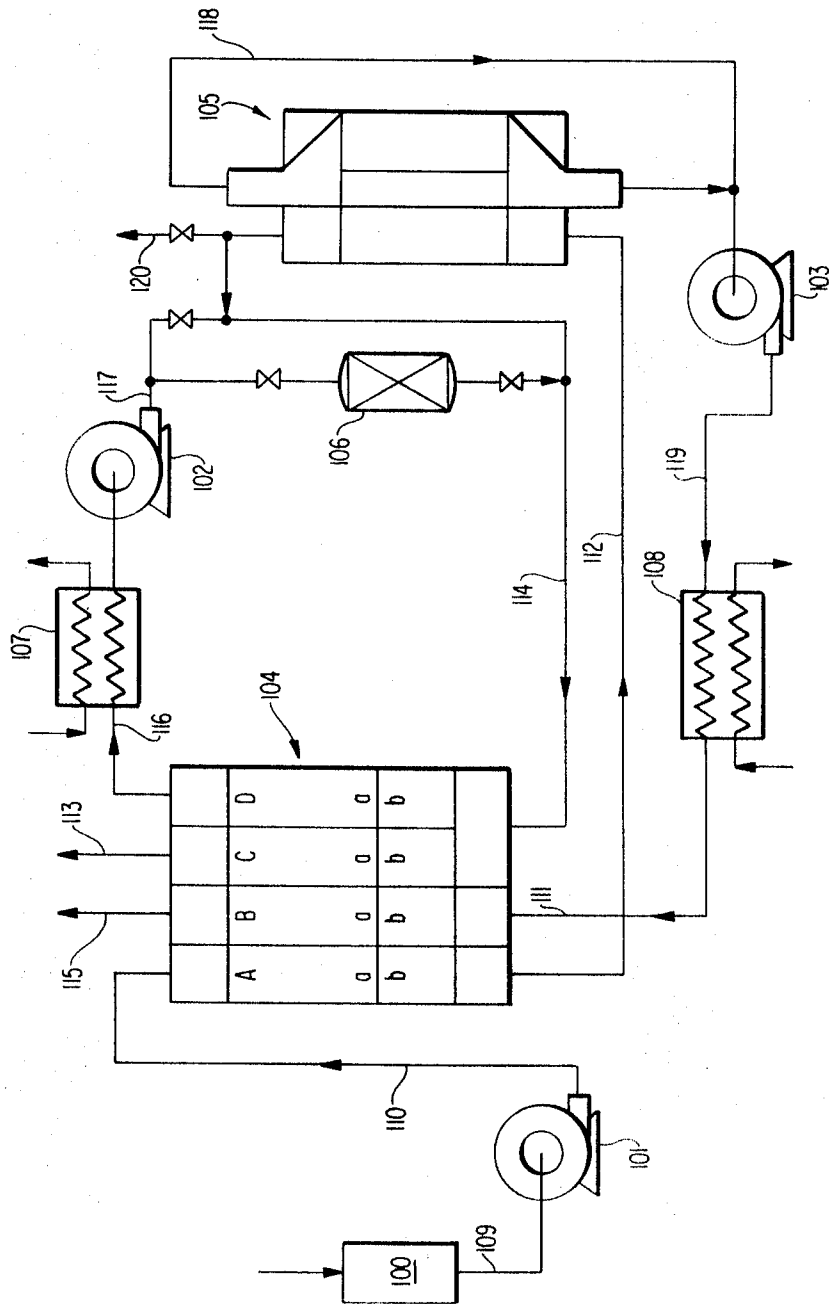


FIG 3

INVENTORS
AKIRA TOYAMA
YUKIO NAKAKO
SADAYUKI NAKANISHI
HIROETU MIKI

BY
Craig, Antonelli, Stewart & Hill

ATTORNEYS

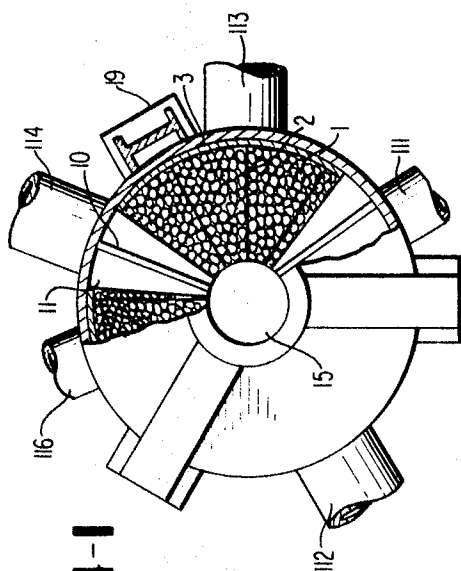


FIG 4-1

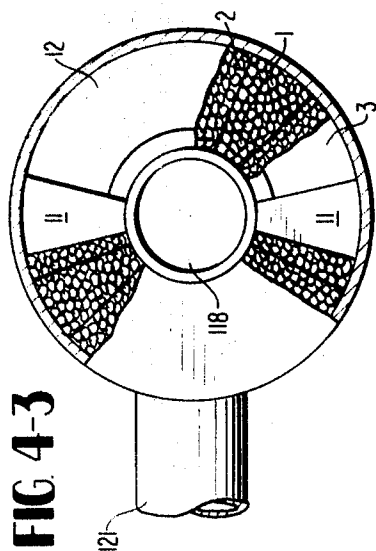


FIG 4-3

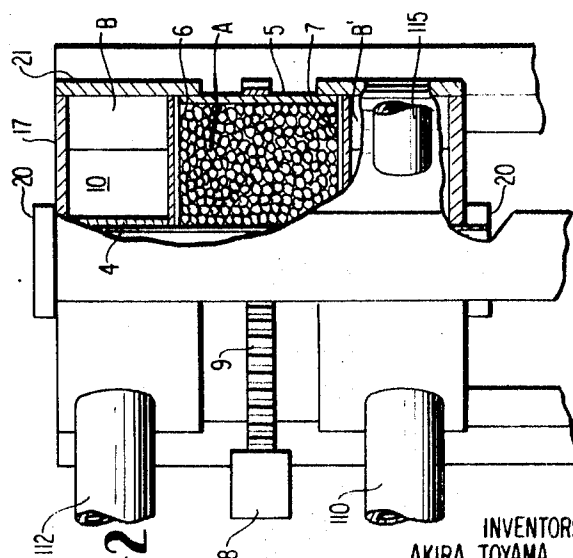


FIG 4-2

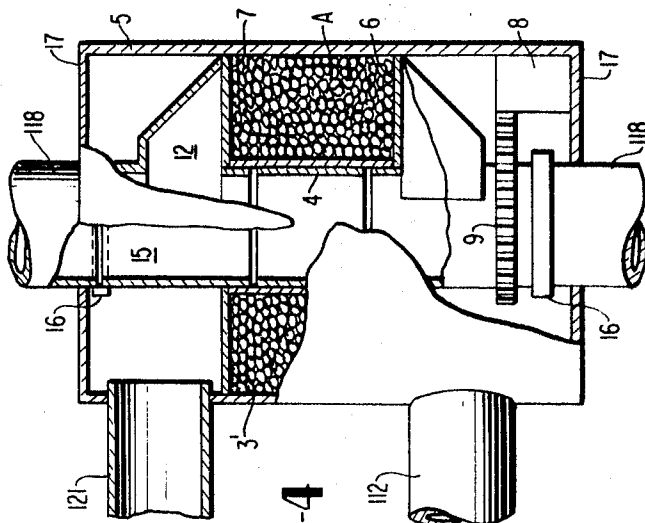


FIG 4-4

INVENTORS
AKIRA TOYAMA
YUKIO NAKAKO
SADAYUKI NAKANISHI
HIROETU MIKI

BY

Craig Antonelli, Stewart & Hill

ATTORNEYS

FIG 5-1

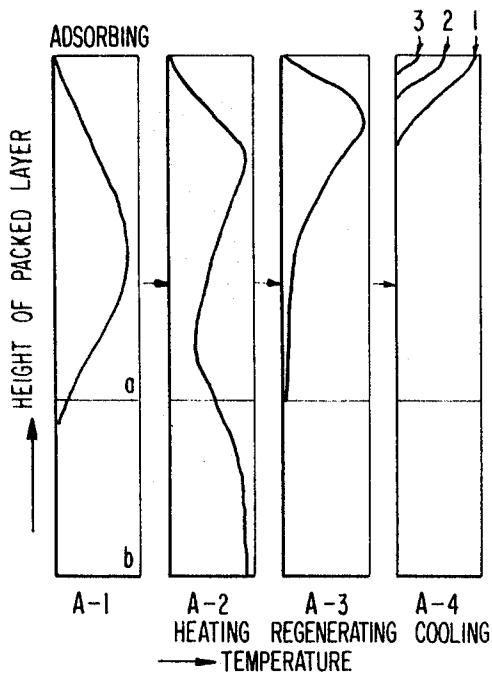


FIG 5-2

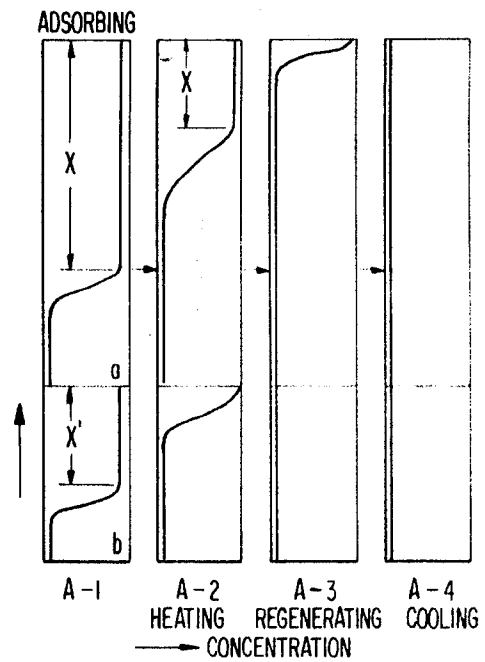
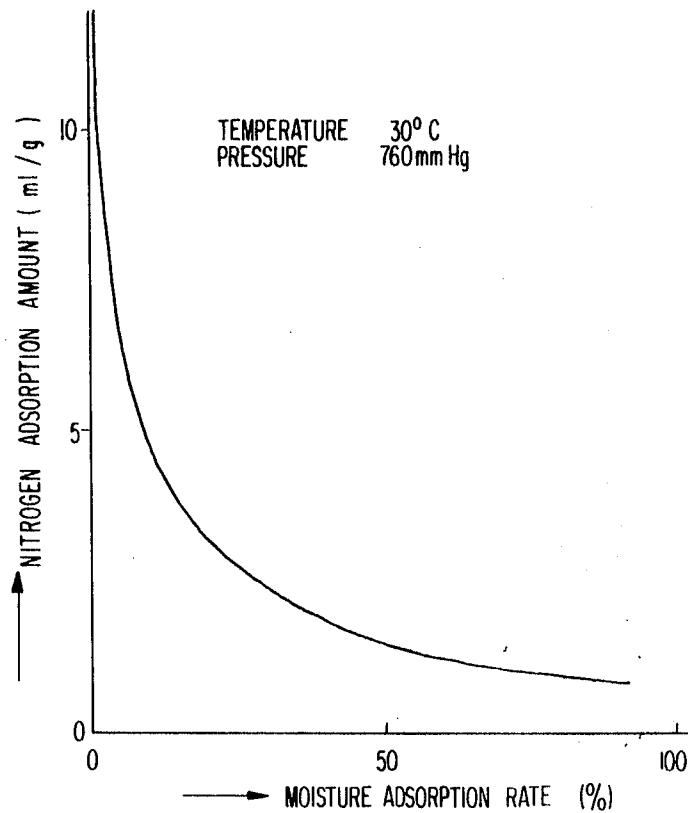


FIG 6



REFINING SEPARATION PROCEDURE OF OXYGEN FROM AIR

This invention relates to a refining separation procedure of oxygen from air. Particularly saying, this invention relates to a procedure for refining separation of oxygen from air by the so-called adsorption method using an adsorbent for removing moisture, carbon dioxide and nitrogen, which are included in air. In details further, this invention relates to a procedure for refining separation of air to obtain continuously oxygen in low purity by adsorbing moisture and carbon dioxide in air with an adsorbing apparatus which includes double layers of adsorbents and then nitrogen with natural or synthetic zeolite.

Hitherto, oxygen has been produced from air by the cryogenically liquefying rectification method or the adsorption method. Since moisture and carbon dioxide in air bring, in the former method, obstructive occlusion into the apparatus owing to their high melting points and reduce, in the latter method, the adsorbability of an adsorbent for nitrogen, they have usually been removed previously in the foretreatment process by the cryogenically solidifying method, the washing method and the adsorption method. Among them, the adsorption method is effected using the adsorption apparatus with either the fixed bed or the moving bed of the adsorbent. In the fixed bed adsorption apparatus, the size of the adsorber increases proportional to the interval of the adsorption-regeneration cycle. When the heating regeneration method is employed, it takes a relatively long time to heat up or cool down the adsorbent, the cycle period gets longer and the size of the apparatus is naturally enlarged, yielding troubles in construction of the apparatus having a large capacity and an unavoidable expensive cost. The moving bed adsorption apparatus, where adsorption and regeneration can continuously proceed, is suitable for treating a large amount of gas with a relatively small amount of the adsorbent, compared with the fixed bed adsorption apparatus. However, it has disadvantages in requiring a high degree of technique to design the stable apparatus and consuming a large amount of the adsorbent due to pulverization during movement in the apparatus.

The first object of this invention is, in the refining separation procedure of air by the adsorption method, to shorten remarkably the cycle in the apparatus consisting of adsorbing, heating, regenerating and cooling by using a usual fixed bed adsorber in a specific arrangement as the adsorption apparatus for adsorbing moisture, carbon dioxide, and nitrogen, whereby the adsorbent amount to be used can be reduced and the size of the adsorption apparatus is minimized.

The second object of this invention is to make enable the adsorption apparatus to operate continuously for a long time by using a usual fixed bed adsorber in a specific arrangement as the adsorption apparatus, whereby the highly efficient procedure can be offered to refining separation of air.

The third object of this invention is to simplify the system of the air refining separation apparatus by using a usual fixed bed adsorber in a specific arrangement as the adsorption apparatus, whereby the maintenance and operation of the apparatus can be easily done.

The fourth object of this invention is to avail of the whole adsorption heat, which is generated during the adsorbing operation, as the desorption heat, by using a usual fixed bed adsorber in a specific arrangement as the adsorption apparatus, whereby the heat to be supplied from the outside for heating regeneration can be greatly reduced.

The air refining separation procedure of this invention is characterized by using both the moisture-carbon dioxide adsorbing apparatus comprising multiple fixed beds, which have two layers of a moisture adsorbing layer and a carbon dioxide adsorbing layer, where four processes of adsorbing, heating, regenerating and cooling are repeated in turn, and the nitrogen adsorbing apparatus comprising multiple fixed beds, where two processes of adsorbing and regenerating are repeated in turn, and comprises:

- a. supplying the raw air into the fixed bed, which is in the adsorption step of the said moisture-carbon dioxide adsorbing apparatus, to obtain the refined air;
- b. supplying the refined air into the fixed bed, which is in the adsorption step of the said nitrogen adsorbing apparatus, to obtain the oxygen of low purity;
- c. supplying a part of the product oxygen of low purity into the fixed bed, which is in the regeneration step of the said moisture-carbon dioxide adsorbing apparatus, and subsequently recovering it as the product oxygen of low purity from the air refining separation apparatus;
- d. supplying the rest of the oxygen of low purity into the fixed bed, which is in the cooling step of the said moisture-carbon dioxide adsorbing apparatus, to cool and dehydrate and then joining it with the low purity oxygen flow from the nitrogen adsorbing apparatus; and
- e. supplying the nitrogen which desorbs from the fixed bed, which is in the regeneration step of the said nitrogen adsorbing apparatus, after heating into the fixed bed, which is in the heating step of the said moisture-carbon dioxide adsorbing apparatus, and subsequently discharging out the same from the air refining separation apparatus.

Although the adsorbing apparatus to be used in this invention is a conventional fixed bed adsorber, its arrangement and usage are substantially different from those of the conventional one so that the remarkable effect is obtained as hereinafter described in details.

In the air refining separation procedure of this invention, the moisture-carbon dioxide adsorbing apparatus and the nitrogen adsorbing apparatus, consisting of conventional multiple adsorbers as shown in FIG. 1, comprise a construction so as to repeat each process in turn by the valve operation according to the usage form. In addition, in this invention, the adsorber so constructed in a body with the multiple fixed beds shown in FIG. 4 as to repeat each process in turn by rotating operation can be used.

FIG. 1 is a flow sheet showing an embodiment of this invention;

FIG. 2 is a chart showing the interrelation between the process of the moisture-carbon dioxide adsorbing apparatus and the valve-switching operation in the embodiment shown in FIG. 1;

FIG. 3 is a flow sheet showing another embodiment of this invention;

FIG. 4 is a plane figure and a side view illustrating the adsorbing apparatus to be used in the embodiment shown in FIG. 3;

FIG. 5 is a profile showing the temperature distribution and the concentration distribution of the adsorbate in the adsorbing bed of the moisture-carbon dioxide adsorbing apparatus in the embodiments shown in FIGS. 1 and 3; and

FIG. 6 is a graph showing the influence of the moisture on the nitrogen adsorbent.

As described above, the characteristics of the air refining separation procedure of this invention lie principally in peculiarity of the usage of the adsorber, which will be clarified by the following illustration with the accompanying drawings.

The moisture-carbon dioxide adsorbing apparatus of the embodiments shown in FIG. 1 consists of the combination of four groups of the conventional fixed bed adsorbers A, B, C, D of the same kind, each adsorber being fitted with the supplying duct 110 for the feed air, the supplying duct 111 for the heated gas, the discharging duct 112 for the refined air, the discharging duct 113 for the product gas, the supplying duct 114 for the product gas and the circulating cooled gas, the discharging duct 116 for the circulating gas for cooling and the discharging duct 115 for the heated gas. Between each adsorber and the ducts there are set up seven electromagnetic valves, which are so contrived together with the timer that each adsorber can continuously remove moisture and carbon dioxide by repeating the cycle of four processes, i.e. adsorbing, heating, regenerating and cooling. Illustrating the example of the valve switching shown in FIG. 2 (wherein the square

indicates the opening of the electromagnetic valve) for the process where an adsorber is used for each of the four groups of the moisture-carbon dioxide adsorbing apparatus as shown in FIG. 1, the adsorber A plays, on the opening of the electromagnetic valves VA-1 and VA-1', the adsorbing operation of the feed air flowing in through the duct 110 and the valve VA-1, and the refined air is sent through the valve VA-1' to the duct 112. In the adsorber B, on the opening of the valves VB-3' and VB-4, the cooling operation proceeds by means of the cooling gas flowing in through the duct 114 and the valve VB-3', and the said cooling gas then flows through the valve VB-4 into the discharging duct 116 for the circulating gas for cooling. In the adsorber C, on the opening of the valves VC-3 and VC-3', the regenerating operation proceeds by means of the regenerating gas flowing in through the duct 114 and the valve VC-3', and the said regenerating gas is discharged as the product gas through the valve VC-3 and the duct 113. In the adsorber D, on the opening of the valves VD-2' and VD-2, the heating operation proceeds by means of the heating gas flowing in through the duct 111 and the valve VD-2', and the said heating gas is discharged through the valve VD-2 and the duct 115. After a given period of time elapses, the respective electromagnetic valves are switched, and each adsorber continuously repeats in turn the processes of adsorbing, heating, regenerating and cooling by means of the on-off operation of the electromagnetic valves with the timer operation, like that way as the adsorber A from adsorbing to heating, the adsorber B from cooling to adsorbing, and so on. In these operations, the gas flowing in from the one side of the piping issues to the other side of the piping at the corresponding location through the adsorber. During this process, the adsorbing of the adsorbate on the adsorbing bed with the gas to be treated, the heating and partially regenerating of the adsorbing bed with the heating gas, the desorbing of the adsorbate and the regenerating of the adsorbent with the regenerating gas, and the cooling of the adsorbing bed with the cooling gas proceed.

The nitrogen adsorber 105 employs a conventional fixed bed adsorber as shown in the flow sheet of FIG. 1 and it so contrived that adsorbing and regenerating in vacuum continuously proceed by means of the combination of the electromagnetic valves and the timer. Illustrating the operation, the refined air flows into the adsorber E and the adsorption of nitrogen proceeds when the electromagnetic valves VE-1 and VE-1' are opened and the valves VE-2 and VE-2' are shut. On the other hand, in the adsorber F, on the opening of the valves VF-2 and VF-2' and the shutting of the valves VF-1 and VF-1', the desorption of the adsorbed nitrogen as well as the regeneration of the adsorbent proceed by means of the vacuum pump reducing the inside pressure. The electromagnetic valves continuously change these operations with each other by the timer operation, whereby the adsorbers E and F repeat alternately the adsorbing and regenerating.

In the embodiment of FIG. 3, there is shown the continuous fixed bed adsorber served as the moisture-carbon dioxide adsorber and the nitrogen adsorber and consisting of the multiple continuous fixed-bed adsorbent-packed layers, which are adjacent to each other by means of partition walls, and the gas distributing sections, which distribute the gases to be supplied to and discharged from the said adsorbent-packed layers. It is so constructed that either of the said adsorbent-packed layers and the gas distributing sections can rotate.

The moisture-carbon dioxide adsorber 104, shown in FIG. 3, consists of, as shown in FIG. 4-1 and FIG. 4-2, three portions, i.e. the adsorbing bed A, which is fixed on the freely rotatable central axis 15 settled on the bearings 20 fixed at the centers of the upper and lower surfaces of the apparatus body, and the distributing sections B and B', which are so fixed to the frame 19 as to contact on the upper and lower surfaces of this adsorbing bed. The adsorbing bed A is divided with the partition walls 2 welded to the central axis 15 and the outer cylinder 5 into the radially ordered small chambers, on the upper and lower surfaces of which are settled either the metal gauzes or the perforated plates 6 and 7 having the adequate

opening area required for the adsorbents 1 not to leak and for the gas to flow freely, and in the space of which are the adsorbents packed. On the outer cylinder 5 of this adsorbing bed A, the driving devices 8 and 9 are settled, and the adsorbing bed A is rotated by this driving device toward the definite direction at the constant speed. The distributing sections B and B' settled at the upper and lower positions of this adsorbing bed A are divided respectively with the parting strips 10 into four divisions for adsorbing, heating, regenerating, and precooling at the corresponding positions of B and B', respectively. When the same gas is used for both regenerating and precooling, either one of the distributing sections is divided into three portions. In the embodiment of FIG. 3 and also in FIG. 4-1 and FIG. 4-2, the distributing section B is divided into three portions and the distributing section B' is divided into four portions.

The said parting strip 10 is fixed to the outer cylinder 21, the upper and lower plates 17 and the central tube 4, respectively, by means of such as to keep airtight. Each contact portion of the parting strip 10 to the adsorbing bed A is fitted with the sealing plate 11 which has the size greater than at least one of the adsorbing chambers radially ordered, and it is so constructed to seal the contact portion between the sealing plate 11 and the end surface of the partition wall 2. Also, the contact portion between the outer cylinder 5 of the adsorbing bed and the outer cylinder 21 of the distributing section has an airtight construction. Thus, the distributing chambers are kept to be perfectly airtight to each other and to atmosphere. Each of the distributing sections B and B' is provided with the raw material air supplying duct 110, the heating gas supplying duct 111, the refined air discharging duct 112, the product gas discharging duct 113, the supplying duct 114 for the product gas and the circulating gas for cooling, the discharging duct 116 for the circulating gas for cooling and the heating gas discharging duct 115. The gas flowing in from the one side of these ducts, goes through the adsorbing bed A into the other side of the ducts at the corresponding position. During this process, the adsorbing of the adsorbate on the adsorbing bed with the gas to be treated, the heating and partially regenerating of the adsorbing bed with the heating gas, the desorbing of the adsorbate and the regenerating of the adsorbent with the regenerating gas, and the cooling of the adsorbing bed with the precooling gas take place.

In the nitrogen adsorber 105 shown in FIG. 3, each adsorbing chamber carries the adsorbing-regenerating cycle out at a definite period, by means of rotation of the distributing sections, which are settled on the upper and lower surfaces of the multiple adsorbing chambers adjacent to each other by means of the partition walls, as shown in FIG. 4-3 and FIG. 4-4. To illustrate the construction, the outside of the apparatus consists of the cylinder 5 and the upper and lower covers 17, and, at the middle of the inside, the adsorbing section A consisting of a series of the fixed bed adsorbing chambers 3 packed with the adsorbent 1. These chambers 3 are divided by means of the outer cylinder 5 of the apparatus, the inner cylinder 4 having the concentric axis therewith, the partition walls 2 to prevent the mingling of gases, which are welded firmly to both of the said cylinders, and the upper surface 7 and the lower surface 6 formed with the perforated plate or the metal gauze having the adequate opening area required for the gas to flow freely and for the adsorbent not to leak. At the central portion of the apparatus, the central tube 15, which is rotatable and internally touched to the inner cylinder 4, is settled, being connected with the duct 118, which is fixed to the upper and lower covers 17 by means of the airtight joint 16. To this central tube 15, the duct 12 leading to the adsorbing chamber 3 is fitted at the upper and lower sides, whereby the desorbed gas from the adsorbing chamber can be discharged to the outside of the apparatus through the tube 15 and the duct 118. At the front and rear of the duct 12, the sealing plates 11 are fitted, and the respective contact portions between these sealing plates 11 and the upper and lower end surfaces of the parting walls 2 are furnished with the sealing devices, to prevent the

gas leakage at the adsorbing and regenerating sections. It is so contrived that the central tube 15, which is fitted with the duct 12 and the sealing plates 11, is made revolved by revolving the revolving body 9 at a constant speed by means of the driving devices 8. In FIG. 4-3, a single duct 12 is settled, respectively, at the corresponding positions to the upper and lower sides, but it is possible to set up the duct in two portions at the symmetrical positions for good balance. For such a case, the sealing plates 11 must be settled at the front and rear of the respective ducts 12. The outer cylinder 5 is fitted with the ducts 112 and 121, respectively, at the upper and lower sides and the gas flows in and out through these two ducts. Namely, the raw gas supplied at the duct 112 flows in the adsorbing chamber 3 of the adsorbing operation section, where the adsorbate is removed by adsorption, and goes out from the duct 121 to the outside as the product gas. On the other hand, in the adsorbing chamber, which contacts at the upper and lower surfaces 6 and 7 to the duct 12, the regenerating operation is carried out at a lower inside pressure by employing a vacuum pump (not indicated in FIGURES) connected with the duct 118. As the ducts 15 and 12 rotate by means of the revolving device 8, the process in the adsorbing chamber changes from adsorbing to regenerating in vacuum, in turn. During this operation, the adsorbing chamber is sealed for a moment with the sealing plate 11, the raw gas is interrupted to flow in. The said adsorbing chamber gets to connect with the duct 12 by rotation of the ducts 15 and 12, and to carry the regenerating process out. The sealing plates 11 should be those having such width as to cover at least one of the adsorbing chambers for carrying the adsorbing and regenerating processes out smoothly.

As understood from the above descriptions, the adsorbent in the adsorbing chamber is fixed as in the fixed bed adsorber. On the other hand, the movement of the ducts relative to the adsorbing chamber makes it possible to subject the adsorbent to adsorption and regeneration in continuation. Therefore, the adsorbing apparatus in this invention can treat a great amount of gas with only a small amount of the adsorbent, not consuming the adsorbent caused by pulverization as seen in the moving bed adsorbing apparatus.

According to this invention, the air refining separation is done by the following new process, based on the specific usage form of the adsorbing apparatus as described above.

To illustrate the air refining separation procedure of this invention with the representative example in FIG. 1, the natural air including moisture and carbon dioxide, after solid components such as dust are removed by a simple filter 100, flows through the duct 109 into the blower 101 for raising pressure up to the necessary pressure for the apparatus, and is further introduced into the moisture-carbon dioxide adsorbing apparatus through the duct 110. The raw air flows then into the adsorbing bed consisting of two layers, i.e. a moisture adsorbing layer *a* and a carbon dioxide adsorbing layer *b*. The majority of moisture is removed during passing through the adsorbing layer *a*, and the remaining moisture in trace and carbon dioxide are removed on passing through the adsorbing layer *b*. When the adsorbing bed gets to adsorb moisture to 40-60 percent of the saturated adsorption capacity, it is transferred by the valve switching operation to the heating as the next process. The adsorption heat evolved at the adsorbing layers accumulates in the adsorbent in the adsorbing bed, and the refined air goes out at a temperature nearly close to the inlet temperature. Accordingly, at the end of the adsorbing operation, the adsorption profile in the adsorbing bed gets to the pattern of A-1 in FIG. 5-2, showing the concentration distribution of the adsorbate in the adsorbing layer, where the parts indicated by X and X' show the saturated state or nearly for moisture and carbon dioxide, respectively. The temperature distribution at this state affords as shown by A-1 of FIG. 5-1, the mountain-shaped curve having the nearly same temperatures at the air inlet and outlet portions and the highest at the adsorption zone. This means that the adsorption heat evolved by adsorption is all retained in the adsorbing layers. In

the adsorbing layer *a*, the adsorbent (e.g. silica gel, alumina gel) having a large adsorption capacity in the high relative humidity region is used. In the adsorbing layer *b*, the adsorbent (e.g. natural zeolite, synthetic zeolite) having a strong adsorption ability in the low relative humidity region and an adsorption ability of carbon dioxide at normal temperatures.

The desorbing nitrogen, which is heated at the heater 108, flows through the duct 111 into the adsorbing bed, which has been transferred to the heating section A-2. After heating a part of the inlet side of the adsorbing layers *b* and *a*, removing the adsorption heat accumulated in the adsorbing bed to the moisture adsorbing section formed in the air inlet side of the adsorbing layer *a*, desorbing a part of the adsorbed moisture and getting to the nearly same low temperature as of the raw air simultaneously with retention of moisture and carbon dioxide, the nitrogen flows out. The majority of the adsorbed carbon dioxide in the adsorbing layer *b* is desorbed off during the said process. Accordingly, as shown in FIG. 5-1, by means that the adsorbing layer *b* is heated with the heated desorbing nitrogen, the temperature of the said adsorbing layer increases, and the majority of carbon dioxide are desorbed as shown in FIG. 5-2, while in the adsorbing layer *a*, by means of the flowing of the heated desorbing nitrogen, the high temperature zone moves upward, and thereby, a part of the adsorbed moisture is desorbed, and the height of the saturated adsorption section X becomes low. By further switching the valves, the adsorbing bed is transferred to the regenerating section A-3. In the regenerating operation, the product oxygen coming through the duct 114 flows through the adsorbing bed, moving further the high temperature zone, which has been formed in the adsorbing layer by the adsorption heat in the previous adsorbing operation A-1, and by the flowing of the hot desorbing nitrogen in the heating operation, to the moisture adsorbing section of the adsorbing bed at the air inlet side, and regenerating the adsorbent. The product oxygen flow issues out of the apparatus at the nearly same temperature as of the inlet air, containing the desorbed moisture and carbon dioxide. On the other hand, the adsorbing bed is made almost free from the adsorbed moisture and carbon dioxide in the regenerating section A-3. The high temperature zone caused by adsorption heat in the adsorbing section A-1 of the adsorbing bed is removed by desorption heat nearly equal thereto in the absolute value of calory to remain in the adsorbing layer *a* the high temperature zone formed in the heating section A-2. Therefore, in the adsorbing bed, the temperature distribution as shown in FIG. 5-1 is established, and moisture in the adsorbent is mostly desorbed, as shown in FIG. 5-2. The adsorbing bed of this state is transferred to the cooling operation. In the cooling section A-4, a part of the product gas to be circulated as the cooling gas, flows through the adsorbing bed as in the regenerating section A-3, removing completely the said high temperature zone, which has been retained in the adsorbing layer, to the outside of the adsorbing bed, and accordingly, the high temperature zone is completely eliminated as the result of the changing of the temperature distribution along the course of ①, ② and ③ in turn, as shown in FIG. 5-1. During this process, the remaining moisture is completely desorbed, and the regenerating and cooling operations of the adsorbing bed are completed. This regenerating of the adsorbent can be carried out more completely, due to the passing of the high temperature zone, which has been formed in the heating section A-3, through the adsorbing bed during the regenerating and cooling operations. On the other hand, the cooling gas issues out at the relatively high temperature, accompanied by a very small amount of moisture, being cooled at the cooler 107 after passing through the duct 116, being dried sufficiently at the desiccator 106 after passing through the duct 117, and being furnished through the duct 114 for the cooling operation. Because of extremely low relative humidity of this cooling gas, natural zeolite or synthetic zeolite is suitable for the desiccant of the desiccator. The air refined in the moisture-carbon dioxide adsorbing apparatus 104 is introduced into the nitrogen adsorbing apparatus 105 through

the duct 112. The refined gas introduced flows into the one of the fixed bed adsorbers, which have the similar construction to the moisture-carbon dioxide adsorbing apparatus. Each adsorber is packed with natural zeolite or synthetic zeolite as the adsorbent, which can adsorb larger amounts of nitrogen than oxygen at the temperature close to the normal temperature. During the passing of the refined air, which has flowed in, through this adsorbing bed, nitrogen in the air is adsorbed, and oxygen in the rest air is enriched, issuing out as the product oxygen through the duct 114, being supplied to the regenerating operation of the said moisture-carbon dioxide adsorbing apparatus 104, and finally, flowing out as the product oxygen through the duct 113 to the outside of the apparatus. On the other hand, the nitrogen, which has been adsorbed in the adsorbing layer of the nitrogen adsorber, is desorbed adiabatically under the reduced pressure by the vacuum pump 103, and discharged through the duct 118, being supplied to the heating operation of the moisture-carbon dioxide adsorbing apparatus 104 through the duct 119, the heater 108, and the duct 111, and finally, being exhausted as the waste nitrogen through the duct 115 to the atmosphere.

In addition, in the procedure of this invention, if it is required to obtain the clean low purity oxygen having no moisture and carbon dioxide, a part of it can be taken out from the duct 120. According to the procedure of this invention, the processes were carried out under the conditions as indicated in Table 1, by employing the moisture-carbon dioxide adsorbing apparatus 104, which consists of the eight adsorbers, where the moisture adsorbing layer *a* is packed with alumina gel, the carbon dioxide adsorbing layer *b* is packed with natural zeolite, and the adsorbing layer of the nitrogen adsorbing apparatus 105 is packed with natural zeolite, and thereby, about 27 Nm³/hr of the low purity product oxygen, which contains about 35 percent oxygen, were obtained from 50 Nm³/hr of the raw air. In the moisture-carbon dioxide adsorbing apparatus of this case, the numbers of the adsorber were so assigned to the respective processes as follows; three to adsorbing, one to heating, two to regenerating, and two to cooling.

TABLE 1

Condition	Operation			
	Adsorbing (air)	Heating (nitrogen- removing)	Regener- ating (oxygen- producing)	Cooling (oxygen- producing)
Superficial velocity (m./s.)	0.5	0.6	0.6	0.6
Reaction time (min.)	15	5	10	10
Temperature (° C.)	13-25	170-180	13-25	13-25

The resulting amounts of the removed moisture and the removed carbon dioxide are those shown in Table 2.

TABLE 2

Moisture			Carbon dioxide		
Adsorber inlet (percent)	Adsorber exit (p.p.m.)	Regenerat- ing and cooling product oxygen (p.p.m.)	Adsorber inlet (p.p.m.)	Adsorber exit (p.p.m.)	Regenerat- ing and cooling product oxygen (p.p.m.)
0.35-0.58	15	16	310-380	0	0
	10	12		0	0
	6	8		0	0
0.8-0.9	11	12	310-380	0	0
	8	10		0	0
	7	8		0	0
1.5-1.6	10	11	310-380	0	0
	8	10		0	0
	7	9		0	0

Although it is possible as described before to use synthetic zeolite as the adsorbent, nitrogen adsorbing ability of the both being listed in Table 3, cheaper natural zeolite is excellent in

adsorbing ability and was the preferably adequate adsorbent for that use.

TABLE 3

Adsorbent	Pressure (mm. Hg)	Tempera- ture (° C.)	Equilibrium amount of adsorption (ML/g.)	
			Oxygen	Nitrogen
N.Z. A.	760	9-13	6.5	19.5
N.Z. B.	760	30	4.5	11.0
N.Z. C.	760	30	4.6	10.2
M.S. 4A.	760	20	2.2	6.2
M.S. 5A.	760	20	3.2	10.9
M.S. 13X.	760	20	2.1	6.7

N.Z. ... natural zeolite

M.S. synthetic zeolite (means Molecular Sieves)

The nitrogen adsorbability of the said adsorbent for nitrogen is influenced by moisture and carbon dioxide in the gas. The influence by moisture is especially remarkable as shown in FIG. 4, and it must be completely removed in the moisture-carbon dioxide adsorbing apparatus. Thus, it was necessary to dry the cooling gas up to the value (a dew-point below -55° C) as shown in Table 3, by means of such adsorbents suitable for drying the gas of low relative humidity as natural zeolite or synthetic zeolite.

As specified above, according to the procedure of this invention for producing the low purity oxygen, the following effects can be obtained:

1. Conventional fixed bed adsorbers are employed as the moisture-carbon dioxide adsorbing apparatus and the nitrogen adsorbing apparatus. Because of the feature particularly in the adsorbing and regenerating procedures of the moisture-carbon dioxide adsorbing apparatus, however, the cycle of adsorbing, heating, regenerating and cooling can be markedly shortened whereby the amount of the adsorbent as well as the size of the apparatus can be minimized to more or less 1/10 of the conventional one and further the operation of the process can be made continuous;
2. Due to employing the continuous fixed bed adsorbing apparatus, the flow-system is very simple, and the operation and maintenance are quite easy;
3. Since the adsorption heat evolved in adsorbing moisture is effectively utilizable as desorption heat, only a little amount of heat to be supplied from the outside is needed for heating and regenerating;
4. Since the product oxygen and the desorbing nitrogen, both of which have no moisture and carbon dioxide, are employed as the regenerating gas for the adsorbent of the moisture-carbon dioxide adsorbing apparatus, the cost for power consumption can be markedly saved; and
5. Since the high temperature zone, which is formed by the desorbing nitrogen heated in the process of heating operation of the moisture-carbon dioxide adsorbing apparatus, moves through the adsorbing layer during the regenerating and cooling operations, the adsorption of moisture and carbon dioxide can be accomplished completely.

What we claim is:

1. The air refining separation procedure, comprising, the use of both a moisture-carbon dioxide adsorbing apparatus consisting of the multiple fixed beds, which have two layers of a moisture adsorbing layer and a carbon dioxide adsorbing layer, where four processes of adsorbing, heating regenerating, and cooling are repeated in turn, and a nitrogen adsorbing apparatus consisting of the multiple fixed beds, where two processes of adsorbing and regenerating are repeated in turn, a. supplying raw air, to obtain the refined air, into the fixed bed, which is in process of adsorption, of the said moisture-carbon dioxide adsorbing apparatus,

- b. subsequently supplying the refined air, to obtain the oxygen of low purity, into the fixed bed, which is in process of adsorption, of the said nitrogen adsorbing apparatus,
- c. supplying a part of the separate oxygen of low purity into the fixed bed, which is in process of regeneration, of the said moisture-carbon dioxide adsorbing apparatus, and subsequently recovering it as the product oxygen of low purity from the air refining separation apparatus, 5
- d. supplying the rest of the oxygen of low purity into the fixed bed, which is in process of cooling, of the said 10 moisture-carbon dioxide adsorbing apparatus, where it is

- dehydrated and cooled, and then joining it with the low purity oxygen flow issued from the nitrogen adsorbing apparatus, and
- e. supplying the nitrogen which desorbs from the fixed bed, which is in process of regeneration, of the said nitrogen adsorbing apparatus, after heating, into the fixed bed, which is in process of heating, of the said moisture-carbon dioxide adsorbing apparatus, and subsequently discharging it out from the air refining separation apparatus.

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