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Process Study and Exergy Analysis of a Novel Air Separation Process Cooled by LNG Cold Energy

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In order to resolve the problems of the current air separation process such as the complex process, cumbersome operation and high operating costs, a novel air separation process cooled by LNG cold energy is proposed in this paper, which is based on high-efficiency heat exchanger network and chemical packing separation technology. The operating temperature range of LNG cold energy is widened from 133K-203K to 113K-283K by high-efficiency heat exchanger network and air separation pressure is declined from 0.5MPa to about 0.35MPa due to packing separation technology, thereby greatly improve the energy efficiency. Both the traditional and novel air separation processes are simulated with air handling capacity of $20t \cdot h^{-1}$. Comparing with the traditional process, the LNG consumption is reduced by 44.2%, power consumption decrease is 211.5 kWh per hour, which means the annual benefit will be up to 1.218 million CNY. And the exergy efficiency is also improved by 42.5%.

Keywords: LNG, air separation, cold energy, exergy analysis, heat exchanger.

Introduction

Liquefied natural gas (LNG) is a kind of low-temperature (111K) and clean energy liquified by the lowtemperature process, and its effective utilization not only can improve the energy efficiency, ease the intensive energy-using pressure, but also produce huge economic benefits^[1-2]. The air separation system process is considered as the most reasonable way to use the LNG cold energy. Because the temperature of air separation process (90-100K) is lower than LNG (111K), and the LNG cold energy can achieve the maximum efficiency in the air separation system compared with being applied in refrigeration (133K), the low-temperature power generation (193K), preparation of dry ice (233K), rubber cryogenic grinding (253K) and other occasions^[3-4].

Using the LNG cold energy in air separation is a cost-effective, environmental and reasonable way. It's mainly because the introduction of LNG can reduce a large number of cold energy produced by a power-driven mechanical refrigeration, and the cold energy is used to maintain a certain low temperature and supply the required cold energy for producing liquid products in the air separation system. Therefore, the power consumption is decreased from 1.000kWh·kg⁻¹ (the conventional air separation process) to 0.333-0.500kWh·kg⁻¹ and the process can save more than 50% of power consumption^[5-8]. Additionally, the combination of the LNG re-gasification and air separation process, not only can provide the air separation unit with a large amount of cold energy within a very short time, which greatly shortens the starting time and improves the device production efficiency, but also

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simplify the air separation process, reduce construction expense, achieve a better efficiency and minimize the energy consumption and the cost.

In recent years, LNG cold energy air separation system has been widely and gradually used and sets off a trend of application in the world because of its significant advantages. This process was put into operation in Tokyo by Japan LOX company in 1971^[9], another LNG cold energy air separation project was put into operation and seven similar projects were under planning and construction by the end of 2012 in China. Nowadays the LNG cold energy air separation technology advances together with the development of these projects. However, the current mature air separation system still has shortcomings: the operating pressure of air separation column of 0.55MPa and the highest operating pressure of the system of 2.6MPa are still high. Therefore, a novel air separation system cooled by LNG cold energy is designed, which can reduce the operating pressure of air separation column and the highest operating pressure efficiently and cut down the total energy consumption.

Process analysis of traditional air separation process cooled by LNG cold energy

The current status of the LNG cold energy air separation process in application

At present, the LNG cold energy air separation technology has been put into operation in Japan, South Korea, France, Australia and other countries. There are also many air separation systems cooled by LNG cold energy which are under the planning and construction in Shenzhen, Fujian, Zhejiang LNG project in China. To sum up applications of this air separation technology home and abroad, some typical examples are shown in Table 1^[10-12].

It can be seen from Table 1: LNG cold energy utilization technology in air separation has realized the Industrial development in Japan and South Korea, and some LNG received stations in Putian, Dapeng, Ningbo in China are also setting up air separation factories to use LNG cold energy in China. The scale of the factories is growing larger, so is the LNG consumption. Besides, the power consumption is reducing

The improvement of air separation technology is aiming to develop the air separation factory, and it can't be separated with the hard-working scientific spirit and unremitting efforts of the scholars.

The introduction of traditional air separation process cooled by LNG cold energy

With the increase of LNG imports in China recently, the scholars have made a lot of study on how to introduce the LNG cold energy to the air separation plant. At present, the comparatively more mature and simple air separation process is a LNG pre-cooling liquid air separation process with medium-pressure nitrogen cycle designed by Yan Na^[13], which is based on the processes of Chen Zeshao ^[14], Jin Tao ^[15] and Xiong Yongqiang's study ^[16]. The process simplified Chen's ^[14] internal and external cycle refrigeration system into internal circulation refrigeration, and cancelled high-pressure nitrogen compressor, nitrogen turbo expander and Freon refrigeration unit. It also solved the problem of Jin Tao's process ^[15] that the liquid nitrogen product could not be obtained, because it was too difficult to achieve low-temperature compression of the cyclic nitrogen. Besides, it reduced a compressor and simplified the process on the basis of Xiong Yongqiang's study ^[16], which made the system more energy-saving and the operation more convenient.

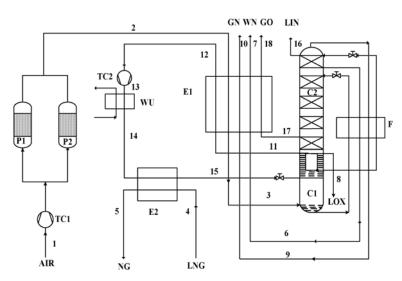
The process of traditional air separation system cooled by LNG cold energy is generally composed of three parts: the pretreatment system of feed air, distillation system and heat exchanger system, and the specific process is shown in Figure 1.

Air at 0.1MPa, 285K enters the air compressor, after compression and purification treatment it changes to 0.55MPa, 303K. Then it enters the main exchanger E1 to obtain the cold energy from the nitrogen and waste nitrogen out from the top of the upper column. The oxygen out from the top of the lower column goes into the bottom of the lower column to rectify again. The distillation system consists of upper column and lower column. And there are four streams (nitrogen, cyclic nitrogen, liquid nitrogen and waste nitrogen) coming out from the top of the upper column and two streams (oxygen and liquid oxygen) coming out from the top of the lower column.

The heat exchanger system is made up of two heat exchangers. There are four cold streams (oxygen, nitrogen, cyclic nitrogen and waste nitrogen) and a hot stream (air) in the heat exchanger E1, a cold stream (LNG) and a hot stream (cyclic nitrogen) in the LNG heat exchanger E2.

 Table 1
 Air separation equipment utilizing LNG cold energy

LNG	Negishi	Senboku	0,	Sodegaura	Pyeongtaek	Putian	Dapeng	Ningbo
Receiving bases	base	base	Chita base	base	base	base	base	base
LNG flow rate(t · h ⁻¹)	8	23	26	34	50	50	54	68
Power consumption $(kWh \cdot kg^{-1})$	0.67	0.5	0.475	0.45	_	0.49	_	_



TC1-Air Compressor; P1,2-Purifier; F-Filter; TC2-Cyclic Compressor; WU-Washing Unit; C1-Lower column; C2-Upper column; E1-Main Exchanger; E2-LNG Exchanger

Fig. 1 Traditional air separation process by using LNG cold energy

The cyclic nitrogen from the lower column enters the heat exchanger E1 directly, exchanges the heat with the air and cools it. After being compressed to about 2.6MPa by circulating compressor TC2 and cooled by washing unit WU, the cyclic nitrogen goes into the LNG heat exchanger E2. And its temperature is cooled to about 120 K by the LNG, pressure is released to 0.12MPa by the throt-tle. Then it goes through the throttle valve and its pressure is compressed to about 0.55MPa. After all of this it is back to the lower column and the cycle is completed.

In this paper, the Aspen plus software is used to simulate the traditional air separation process cooled by LNG cold energy with air handling capacity of $20t \cdot h^{-1}$. The components of the feed air and the LNG are shown in

Table 2.

We set the isentropic and mechanical efficiency of the air pre-treatment compressor and nitrogen compressor at 0.85 and 0.9, respectively. Peng-Robinson equation is chosen to calculate thermodynamic properties of the streams in the process. The simulation results of main streams in the traditional process are shown in Table 3.

The characteristics of the process are as follows: (1) LNG heat exchanger is introduced to replace the freon refrigeration unit of the traditional process (reference [14]), which can recover LNG cold energy, save energy and lower production costs effectively; (2) The nitrogen external circulation system (reference [14]) is abolished and two compressors (the nitrogen turbo expander and

Ingredient	N_2	O ₂	CH_4	C_2H_6	C_3H_8	C ₄ H ₈ -1	C ₄ H ₈ -2
Air	0.791	0.209	0	0	0	0	0
LNG	0.0011	0	0.8939	0.0576	0.033	0.0066	0.0078

 Table 2
 Components of air and LNG (mole ratio)

Table 3	The major stream	is simulation	results of traditional	air separatior	n process using LNG	cold energy
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	U U	Temper-	Pressure/	Flow			Temper-	Pressure/	
Stream	Ingredient	ature/ K	MPa	$/ t \cdot h^{-1}$	Stream	Ingredient	ature/ K	MPa	Flow / $t \cdot h^{-1}$
1	air	285	0.1	20	10	nitrogen	282	0.1	1.3
2	air	303	0.55	20	11	nitrogen	103	0.55	13.75
3	air	239	0.55	20	12	nitrogen	190	0.55	13.75
4	LNG	113	3	4.98	13	nitrogen	309	2.6	13.75
5	NG	300	3	4.98	14	nitrogen	303	2.6	13.75
6	Waste nitrogen	79	0.13	1.01	15	nitrogen	120	2.6	13.75
7	Waste nitrogen	282	0.13	1.01	16	Liquid nitrogen	79.5	0.13	2.13
8	Liquid oxygen	92	0.15	2.95	17	oxygen	89	0.1	1.21
9	nitrogen	81	0.1	1.3	18	oxygen	283	0.1	1.21

the booster compressor) are eliminated, which not only reduce the maximum operating pressure of the system and ensure safety, but also simplify the process; (3) The introduction of the LNG cold energy decreases the precooling temperature of cyclic nitrogen, reduces the power consumption largely and realizes the energy conservation effect more obvious. However, there still exists some drawbacks that the operating pressure of air separation column of 0.55MPa and the highest operating pressure of the system of 2.6MPa are still high, so as the air consumption which is 0.358kWh·kg⁻¹.

Technical research of the novel air separation process cooled by LNG cold energy

A new process is proposed on the foundation of drawing on the virtue and weakness of the above researches, which can widen the operating temperature range of LNG cold energy from 133K-203K to 113K-283K, recover the LNG cold energy and save the energy more effectively at the same time. Besides, it also solves the problem of low-temperature compression, reduces the highest operating pressure of the system from 2.6 MPa to 1.5MPa and enhances the security of the system.

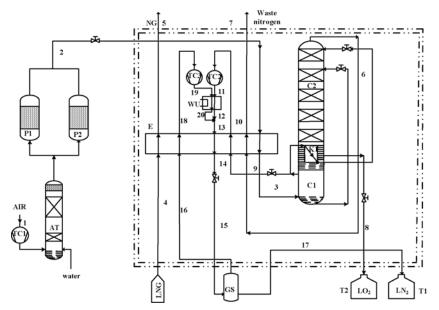
As it is shown in Figure 2, comparing with the traditional process, the differences between the novel method and the traditional one are that the initial state of the air is 0.12MPa, 288K and the pressure enters the heat exchanger is 0.35MPa. The entire distillation column is the distillation part, and the use of the packing technology decreases the operating pressure to 0.35MPa. The specific operational procedures of distillation part are as follows: the cyclic nitrogen comes out from the top of the lower column. And it enters the heat exchanger E to cool down, then it is compressed to about 1.5MPa by nitrogen compressor TC2, washed by water unit (stream 12), liquified and cooled to 110K by LNG cold energy, overheat waste nitrogen and cyclic nitrogen out from the tower (stream 9) in the heat exchanger E. Then the cyclic nitrogen's pressure is released to 0.12MPa by the throttle. After all, the gas-liquid separator GS separates all the stuff. The liquid nitrogen goes directly into the liquid nitrogen tank T1 for storage, and gas enters the heat exchanger E heated to about 298K, then it is compressed to 1.5MPa and cooled to 303K (stream 20) by the nitrogen compressor and the washing unit, respectively. At last, stream 20 mixes with stream 12 (stream 13) enters the heat exchanger. The cycle is completed.

The Aspen plus software is used to simulate the novel air separation process cooled by LNG cold energy with air handling capacity of $20t \cdot h^{-1}$. The components of the feed air and the LNG are shown in Table 2, property method and device parameters are the same as the traditional process. The simulation results of main streams in the novel process are shown in Table 4.

Comparison of the novel and traditional technology and exergy efficiency analysis of system

Simulation result analysis of the novel and traditional technology

In order to verify the new technology with the charac-



TC1-Air Compressor; TC2-Nitrogen Compressor 1; TC3-Nitrogen Compressor 2; E-Exchanger; T1-Liquid Nitrogen Tank; T2-Liquid Oxygen Tank; C1-Lower column; C2-Upper column; K1-Condensate Evaporator; AT-Washing Tower; WU-Washing Unit; GS-Gas-liquid Separation

Fig.2 The novel air separation process by using LNG cold energy

Stream	Ingredient	Temper- ature/ K	Pressure/ MPa	$Flow / t \! \cdot \! h^{-1}$	Stream	Ingredient	Temper- ature/K	Pressure /MPa	$Flow \ / \ t \cdot h^{-1}$
1	air	288	0.12	20	11	nitrogen	468	1.5	3.5
2	air	303	0.35	20	12	nitrogen	303	1.5	3.5
3	air	98	0.35	20	13	nitrogen	303	1.5	5.528
4	LNG	113	0.12	2.778	14	nitrogen	110	1.5	5.528
5	NG	283	0.12	2.778	15	nitrogen	79	0.12	5.528
6	Waste nitrogen	81	0.12	12.25	16	nitrogen	79	0.12	2.028
7	Waste nitrogen	283	0.12	12.25	17	Liquid nitrogen	79	0.12	3.5
8	Liquid oxygen	92	0.13	1.2	18	nitrogen	298	0.35	2.028
9	nitrogen	96	0.35	3.5	19	nitrogen	642	1.5	2.028
10	nitrogen	298	0.35	3.5	20	nitrogen	303	1.5	2.028

Table 4 The major streams simulation results of the novel air separation process by using LNG cold energy

teristics of high-efficiency heat exchange and low energy consumption, this paper analyzes operating pressure of system, energy consumption of equipment and unit liquid products, heat exchange condition of heat exchangers and exergy efficiency of system of both the novel and traditional processes, respectively. Aspen Plus is used to simulate the processes, and the simulation results are shown in Table 5.

It can be seen from Table 4 and Table 5 that a novel air separation process with air handling capacity of $20t h^{-1}$ consumes LNG of $2.778t \cdot h^{-1}$, it has a 44.2% reduction of LNG compared with the traditional process. The liquid products are LN₂ (99.99mol %) of $3.5t \cdot h^{-1}$ and LO₂ (99.80mol %) of $1.2t \cdot h^{-1}$. The total energy consumption is 1469kW•h per hour, and the unit power consumption of the liquid air separation products is 0.313kWh·kg⁻¹, which can save 211.5kW per hour compared with the traditional system of 0.358kWh·kg⁻¹. If the system operates 24 hours every day, 300 days every year, and electricity price is 0.8 CNY per kWh, and the annual benefit will be 1.218 million CNY. The unit power consumption of the liquid air separation products is 12.6% lower than the traditional process, which might be explained by the following three reasons: (1) The high-efficiency heat exchanger network is adopted in this process which can

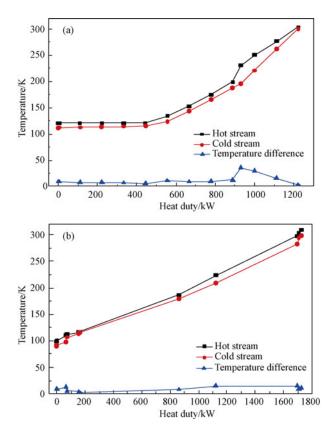
match the heat transfer between LNG and nitrogen better. The latent heat of the two streams are used efficiently; (2) The heat exchanger pressure of cyclic nitrogen drops from 2.6MPa to 1.5MPa; (3) The operating pressure of air separation tower drops from 0.55MPa (traditional process) to 0.35MPa because of the packing separation technology, which greatly decreases the energy consumption of compressed nitrogen.

The heat exchange case of LNG heat exchanger by the traditional process is shown in Figure 3 (a), and the novel process is shown in Figure 3 (b). The less difference of temperature between the hot and cold streams, the higher efficiency of the heat transfer. On the contrary, the greater difference of temperature between the hot and cold streams, the lower efficiency of the heat transfer.

Compared (a) with (b) we can see that the difference of temperature between the hot and cold streams of traditional process is too large, one of the largest differences of temperature is 35K. However, the difference of temperature of 10K between the hot and cold streams of novel process is much smaller, and the grips temperature (minimum difference of temperature) is only 1.9K. All of these certify that the utilization of highly efficient heat exchanger network can make full use of cold energy and show a significant energy saving effect.

 Table 5
 Simulation results of the traditional and novel nitrogen expanding refrigeration system

Technical Specifications	Traditional process	Novel process
handling capacity of air/ t · h ⁻¹	20	20
LNG outlet pressure/MPa、Temperature/K	3.0, 300	0.12, 285
LNG consumption/ $t \cdot h^{-1}$	4.98	2.778
flow ratio of cyclic nitrogen and processing air	0.6875	0.1014
compression work of air /kW	1580	641.5
compression work of nitrogen/kW	462.6	377.6
other consumption/kW	600	450
The maximum operating pressure/MPa	2.6	1.5
Unit energy consumption of liquid products/kWh·kg ⁻¹	0.358	0.313



The heat exchange profile of exchanger E of the tradi-Fig. 3 tional (a) and novel (b) process

System exergy analysis of novel and traditional air separation processes cooled by LNG cold energy

Exergy balance analysis ^[17] is used to analyze the exergy of all the parts and the whole system of both the novel and traditional air separation processes cooled by LNG cold energy at the same condition in this paper. System gray box analysis model is applied to analyze the exergy ^[18]. The process is a physical change process of steady flow, which isn't involved in the chemical reaction. Therefore, only the physical exergy of each stream is calculated in this article, and the exergy of streams is calculated by the formula (1), air contains oxygen of

0.209, nitrogen of 0.791. And 0.1MPa, 298.15K is set as the environmental state, which is called the zero exergy. The system exergy efficiency is calculated by the formula (2)^[19]. The results of each stream are shown in Table 6.

$$E_{x,h} = (H - H_0) - T_0(S - S_0)$$
(1)

$$\gamma = E_{x,ef} / E_{x,in.}$$
 (2)

E_{x,h}——The enthalpy exergy of working fluid, refers to the working ability of the working fluid while it deviates from the equilibrium state at the atmospheric environment.

H, S--The enthalpy and entropy of the working fluid at the end state, respectively.

 H_0 , S_0 —The enthalpy and entropy of the working fluid at 0.1 MPa, 298.15K, respectively.

η-System exergy efficiency, characterizes the effective utilization ratio of the exergy in the system.

 $E_{x,ef}$ —The net output exergy of the system. $E_{x,in}$ —The net input exergy of the system.

Table 6 shows the exergy analysis results of the traditional and novel air separation processes on the basis of cyclic nitrogen gas expansion refrigeration. As it can be seen from Table 6, the novel process need less exergy input than the traditional one at the same air handling capacity, and the former exergy efficiency of the whole system is 1.7 times of the latter one. The reasons can be explained as follows: (1) The high-efficiency heat exchanger network is adopted in this process which can match the heat transfer between LNG and nitrogen better. The latent heat of the two substances is used efficiently; (2) The novel process uses LNG cold energy to substitute the nitrogen external cycle expansion refrigeration, cancel the external cyclic nitrogen, and decrease the cycle pressure to 1.5MPa. Meanwhile the introduction of LNG can eliminate some equipment such as expander and booster, which will lower the energy consumption of system; (3) The operating pressure of air separation tower in the novel process drops to 0.35MPa. In this way, energy consumption of nitrogen compressor is reduced greatly.

 Table 6
 Results of the exergy analysis of the traditional and novel processes

Analysis condition		Traditional process		Novel process		
	compression work of air	1497		605.6		
input exergy/kW	compression work of nitrogen	444.3	3563	353.5	2358	
	LNG cold exergy	1622		1399		
	LN_2	100.5		727.2		
Outrust an and A-W	LO_2	128.4	12(2	243.0	15(7	
Output exergy/kW	Cyclic nitrogen	1057	1363	525.7	1567	
	Waste nitrogen	76.60		70.57		
System exergy efficie	ncy/ %	38.2		66.4		

Characteristic and potential application of the novel process

Characteristics of the novel process

The characteristics of the novel process are reflected in the following aspects:

1) Design of the high-efficiency heat exchanger network gathers all hot and cold streams together and widens the operating temperature range of LNG cold energy from 133K-203K to 113K-283K. The high-efficiency heat exchanger network will use the LNG cold energy, save the energy consumption and reduce the investment cost effectively at the same time.

2) Simulated the traditional and novel air separation processes cooled by LNG cold energy with air handling capacity of $20t \cdot h^{-1}$. The LNG consumption is reduced by 44.2% compared with the traditional process, and the power consumption decreases 211.5kWh per hour, which means the annual benefit is up to 1.218 million CNY. And the exergy efficiency is also improved by 42.5%. All of these prove that the novel air separation process cooled by LNG cold energy has a great energy saving effect and economic benefits.

3) The maximum operating pressure of the system is reduced from 2.6MPa to 1.5MPa so that the safety is insured.

4) The cyclic nitrogen flows into the gas-liquid separation directly after the throttle instead of entering the distillation tower, and the liquid nitrogen is collected as product after separation. The low-temperature nitrogen gas enters the heat exchanger again to recycle cold energy.

5) The problem of low-temperature compression of cyclic nitrogen has been solved. The outlet temperature of cyclic nitrogen from the heat exchanger rises from 95K, 190K and 282K in the literature [14], [15] and [13] respectively to 298K. The rising of outlet temperature not only avoids the technical difficulty brought by the low-temperature compression of cyclic nitrogen in the literature [14-15], but also lowers the outlet temperature of some other streams which are used to cool the feed air in the literature [13]. This part of cold energy can transfer heat with the cyclic nitrogen before it enters the compressor, in order to reduce the energy consumption of the cyclic nitrogen compressor and improve energy efficiency.

6) The characteristics of system process are simple, miniature, skid-mounted and open-stop feature.

Potential application of the novel process

LNG satellite stations which locate in the remote and scattered areas are highly fragmented and unstable for providing the LNG cold energy. And these features lead the traditional air separation system not to be implemented in the LNG satellite station. These satellite stations have a low LNG flow rate and big cold energy fluctuations, which limits the use of LNG cold energy. However, when the broad distribution of LNG satellite station inosculates with its industrial development layout, its cold energy will have a great utilization space, which can achieve a less investment and quicker payback for the cold energy utilization projects. Therefore, the large number of small-scale LNG satellite stations has a great potential development. This paper designs a novel air separation process cooled by LNG cold energy, with characteristics of simple, miniature, skid-mounted and open-stop feature. The novel process can realize full use of the cold energy and develop itself with the development of LNG satellite station.

Summary

After studying the traditional and novel air separation processes cooled by LNG cold energy, this paper analyzes the processes from the cold energy and exergy consumption. In condition, the novel process has a wide prospect in application. The main conclusions are as follows:

1) The high-efficiency heat exchanger network is designed to widen the operating temperature range of LNG cold energy from 133K-203K to 113K-283K. At the same time, the air separation pressure is reduced from 0.5MPa to about 0.35MPa. It has a significant energy saving effect such as the unit power consumption of the liquid air separation products is reduced by 12.6%. Raising the temperature of the cyclic nitrogen pressure-outlet from the heat exchanger to approximately 298K can avoid the technical difficulty caused by the low-temperature compression. On one hand, it can enhance the security of this system by reducing the heat transfer pressure of the cyclic nitrogen to 1.5MPa; On the other hand, the energy consumption can be lowered at the same time.

2) Simulated the traditional and novel air separation processes cooled by LNG cold energy with air handling capacity of $20t \cdot h^{-1}$, the LNG consumption of the novel process is reduced by 44.2% compared with the traditional one. And the power consumption decreases 211.5kWh per hour, which means the annual benefit will be up to 1.218 million CNY. The exergy efficiency has also a 42.5% improvement. Above all, it proves that the novel air separation process cooled by LNG cold energy has a great energy saving effect and economic benefits.

3) Designed a novel air separation process cooled by LNG cold energy, according to characteristics of LNG satellite stations such as huge fluctuations of cold energy, dispersive cold source and limited number. And this set of equipment with miniature, skid-mounted and openstop feature can fulfill the full utilization of LNG cold energy in LNG satellite station in remote areas. Comparing with the traditional air separation process, the new one designed in this paper has characteristics of low energy consumption (unit power consumption of liquid air separation is only 0.313kWh·Kg⁻¹), relatively low system operation pressure (0.35MPa), flexible adjustment, security system, etc.

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