# Development of Membrane Moisture Separator for BWR Off-gas System

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## ABSTRACT

In BWR plant off-gas treatment systems, dehumidifiers are used to maintain noble gas adsorption efficiency in the first half of the charcoal hold-up units. From the perspective of simplifying and reducing the cost of such a dehumidification system, Japanese BWR utilities and plant fabricators have been developing a dehumidification system employing moisture separation membrane of the type already proven in fields such as medical instrumentation and precision measuring apparatus. The first part of this development involved laboratory testing to simulate the conditions found in an actual off-gas system, the results of which demonstrated satisfactory results in terms of moisture separation capability and membrane durability, and suggested favorable prospects for application in actual off-gas systems. Further, in-plant testing to verify moisture separation capability and membrane durability in the presence of actual gases is currently underway, with results so far suggesting that the system is capable of obtaining good moisture separation capability.

## Membrane Moisture Separator System

### Principle of the gas separation membrane

Figure 1 shows the principle of the gas separation membrane. When a non-porous membrane is placed between high-pressure and low-pressure sides, the components contained within the gas will pass from the high-pressure side through the membrane, after which they are diffused into the low-pressure side. The ability of the gas separation membrane to allow the gas to permeate is the product of the total pressure and the molar fraction, meaning that it is dependent on the partial pressure differential of the permeating gas. The capability of the gas to permeate the membrane differs depending on the materials from which the membrane is constructed and the nature of the components contained within the gas. The gas separation membrane used in the off-gas system has a moisture ($H_2O$) permeability coefficient that is hundreds of times greater than its air ($O_2$ and $N_2$) permeability coefficient, a characteristic which makes it well-suited for separating moisture.

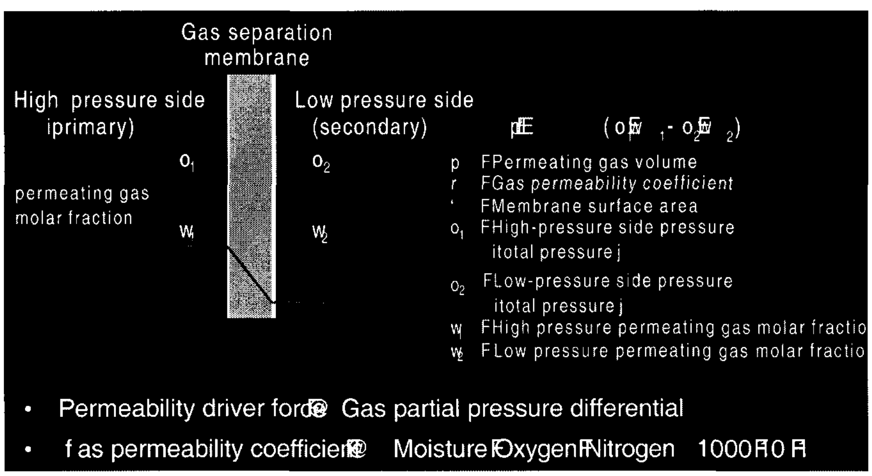


Figure 1 Principle of the gas separation membrane

### The moisture separation membrane module

The moisture separation membranes are configured to have a large surface area and offer excellent pressure tightness. Several thousand of these are bundled within a stainless steel container to form a single membrane module.

Figure 2 shows a conceptual outline of the membrane module. When humid gases pass through the moisture separation membrane, the partial pressure differential of the permeating gas allows the moisture contained within it to pass to the outside of the membrane, thereby dehumidifying the gas. In order to prevent gas separation performance degradation caused by the increasing molar fraction of the moisture that has passed through to the outside of the membrane, the system is designed to introduce a portion of the dehumidification gas obtained at the discharge port in order to purge the moisture that has passed through the membrane.

Also, applying this method within the off-gas system of BWR plants demands not only good moisture separation capability at the moisture separator discharge port, but also high membrane durability in terms of resistance to heat, water, radiation and chemicals, all of which durability requirements are fulfilled by the membrane in question.

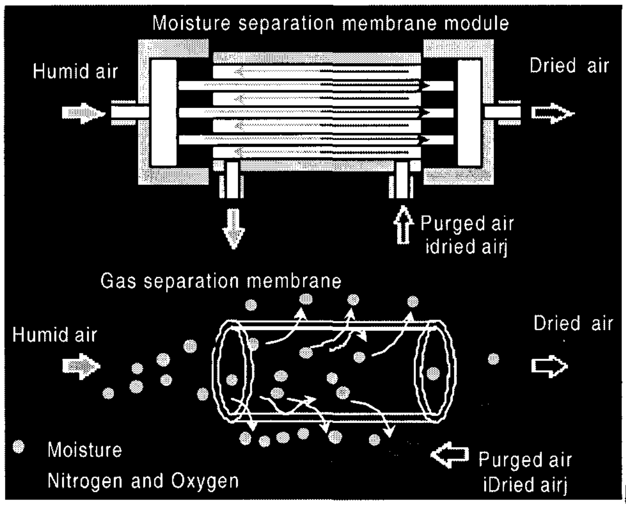
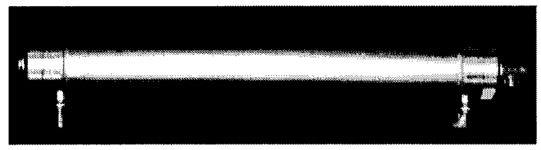


Figure 2 Moisture separation membrane module


Exterior view of Moisture separation membrane module
Diameter: 70mm , Length: 1,100mm

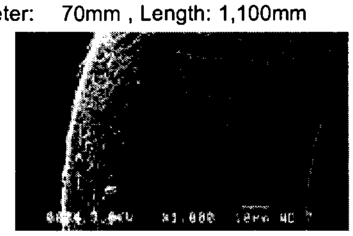

Cross-sectional view of the moisture separation membrane

Photo 1 Moisture separation membrane exterior module photo and cross-sectional views

**Study on application of the membrane moisture separator to the off-gas system**

While membrane moisture separators have found general use in fields such as medical and precision measuring apparatus, their application to BWR plant off-gas systems requires consideration of two additional aspects, including ‘operating pressure’ and ‘purge gas handling.’

- Operating Pressure

Among the general operational conditions for moisture separation membranes, the design must be such that the system maintains a high pressure differential, including introducing gas to the primary (high-pressure) side at a pressure of approximately 0.5-0.7MPa, followed by discharge into the atmosphere after lowering the pressure of the purge gas on the secondary (low-pressure) side. Application to off-gas systems, however, requires that the primary-side operating pressure be very near to atmospheric pressure, and this presents considerable difficulty in terms of maintaining a high pressure differential.

- Purge Gas Handling

While in general applications purge gas (secondary side) is simply released into the atmosphere, when applied to off-gas systems the radioactivity of the purge gas means that such release into the atmosphere is not an option.

Having taken the above two points into consideration, a study of how to apply a membrane moisture separator in an actual off-gas system resulted in the system construction shown in Figure 3.

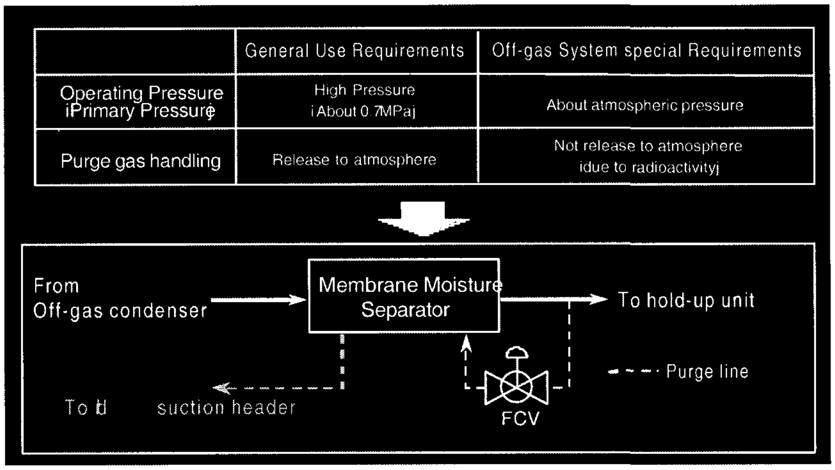



Figure 3 System layout for applying membrane moisture separator system within an off-gas system

As shown in the figure above, by making the connection to the purge line at the main condenser and nearby to it (SJAE intake header), the purge gas pressure is maintained in a state of high vacuum, resulting in a maintained high pressure differential, while at the same time ensuring that the gas is not released into the atmosphere. Further, even if the purge gas is returned to the main condenser, as long as a suitable purge volume (purge flow volume/dehumidifier discharge flow volume) is chosen, there will be no detrimental effect on the degree of the vacuum.

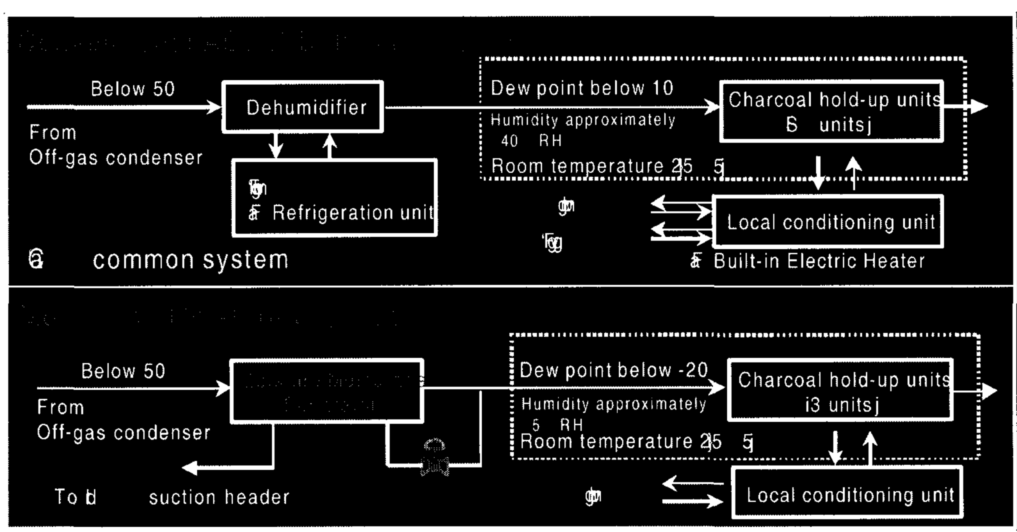



Figure 4 ABWR plant off-gas moisture separation system diagrams

Figure 4 outlines the application of the membrane moisture separator system to the ABWR plant off-gas system. The dehumidifier removes the moisture contained in the gas in order to maintain the noble gas adsorption efficiency of the active charcoal. In ABWR plant the dehumidifier is used to keep the dew point of the gas to 10 or less, and local air conditioning is used to control the air temperature in the charcoal hold-up units in order to maintain the performance of the active charcoal. In contrast, when the dehumidifier is replaced by the membrane moisture separator system, in addition to simplifying the system, the dew point at the intake to charcoal hold-up units decreases, and the resulting increases in active charcoal adsorption efficiency make it possible to reduce the volume of active charcoal. Also, the fact that strict control of the hold-up unit temperature is not necessary promises simplification of the air conditioning system. Design requirements for a system employing the membrane moisture separator are detailed in Table 1.

Table 1 @Membrane moisture separator design requirements

| parameters | requirements |
|---|---|
| Primary intake air volume flow rate | 80 $Nm^3/h$ { purge flow rate |
| Primary intake pressure | Atmospheric pressure or higher |
| Primary intake temperature | 50 or lower |
| Secondary discharge pressure | -690 mmHg or lower |
| Purge volume flow rate | 30 % or less of the intake flow |
| Dehumidifier pressure loss | 1000 $mmH_2O$ or lower |
| Primary discharge dew point | -20 or lower |

## Laboratory Testing

The moisture separation capability of the membrane moisture separator system is dependent upon factors such as transmembrane gas volume flow rate, gas temperature, and pressure differential, but when applying this system within the off-gas system, the system must be able to manifest moisture separation capability for all possible plant operation conditions (including startup, shutdown, normal operation). Further, the durability (deterioration characteristics) of the membrane itself must also be verified. Therefore, as a first step toward verifying these moisture separation performance and membrane durability requirements we conducted laboratory testing based on a simulation of the off-gas system.

## Laboratory testing apparatus

Assuming the introduction of the membrane moisture separator into the off-gas system, we developed testing apparatus designed to simulate the off-gas system. The operational flow of the apparatus used in this laboratory testing is shown in Figure 5, while Photo 2 shows an overview of the apparatus itself. The apparatus was comprised of the following components: a compressor to supply air; a pre-filter to remove oil and other contaminants; a bubbling tank and heater tubing assembly designed to supply heated, humidified gas; a canister used to hold the moisture separation membranes; vacuum pumps used to transfer both the gas and the purge gas; and sensors placed at various points to measure temperature, humidity, and dew point. Volume flow rate was adjusted using a flow control valve, while a thermal mass flow controller regulated the flow of aeration gas. A bubbling tank and heating tube assembly was used to raise the temperature as needed to maintain a constant temperature established for the purpose of the experiment. Pressure was adjusted using the vacuum pumps and the purge gas thermal flow controller in order to simulate the pressure in the off-gas system (moisture separator suction pressure near to atmospheric pressure, purge gas pressure in a high vacuum state).

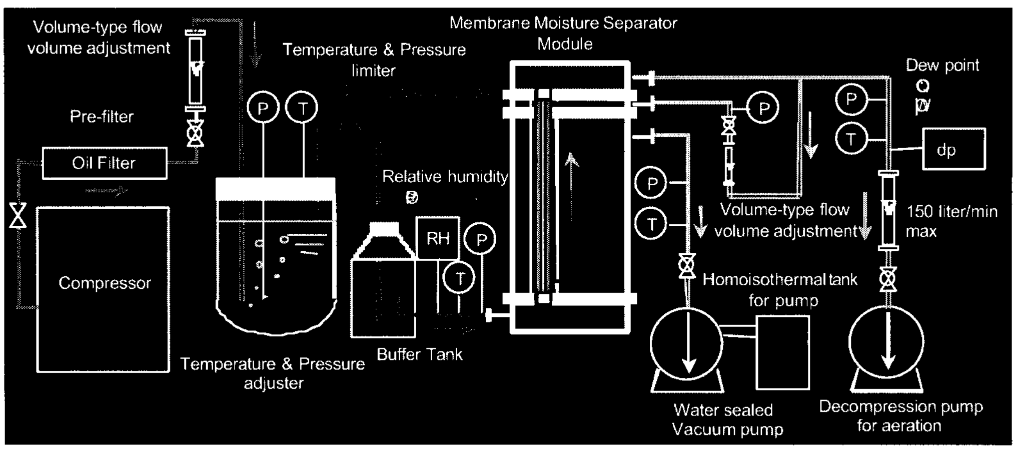



Figure 5 Testing apparatus flow diagram

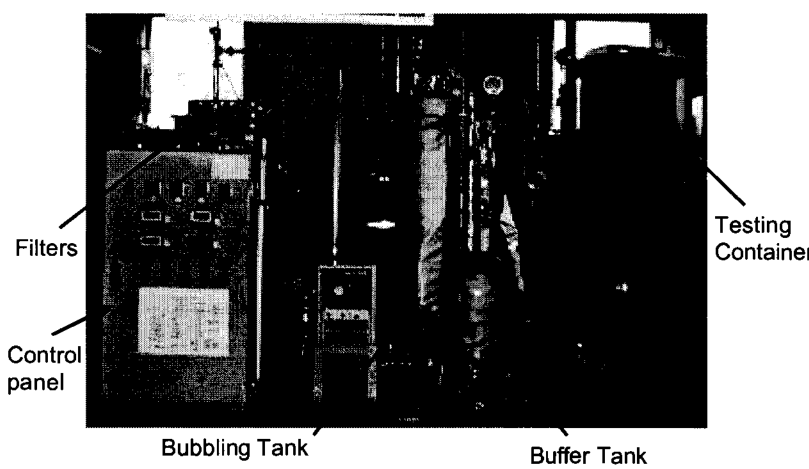



Photo 2 Laboratory testing apparatus

## Laboratory testing design

Tables 2 and 3 detail the items tested in the laboratory in order to verify the moisture separation performance and durability requirements needed for actual application. To test moisture separation performance we operated the laboratory testing apparatus, sending saturated vapor to the moisture separator and measuring the dew point at the moisture separator discharge. As an evaluation criteria we established that satisfactory moisture separation capability would be indicated by the achievement of a dew point of -20 or lower (the design dew point of the off-gas system) at the testing apparatus discharge. Durability testing included removing the moisture separation membranes from the module following the performance test, conducting tension tests on these, and comparing their tensile strength before and after the performance testing.

The testing parameters used during the laboratory testing are listed in Table 4, and the various tests were conducted based on these conditions. We optimized the aeration volume flow rate in consideration of the pressure loss typically found in off-gas systems, and we set the purge rate at the lowest possible rate capable of still maintaining high moisture separation capability. We also adjusted the moisture separator intake temperature to design temperature, and set the moisture separator intake pressure and purge gas pressure differential based on a condenser design vacuum that hypothesized rising coolant temperatures during summer operation.

Table 2 Moisture separation capability test

| Test Item | Content |
|---|---|
| Aeration flow dependence | @Varied the aeration flow to the module and verified the resulting moisture separation capability |
| Gas temperature dependence | @Varied the aeration gas temperature and verified the resulting moisture separation capability |
| Pressure dependence | @Varied the transmembrane pressure differential and verified the resulting moisture separation capability |
| Performance achievement testing | @Measured the amount of time required to satisfy performance requirements |
| Long-term stability | @Conducted extended long-running operation and verified the resulting dehumidification performance stability |
| Life span evaluation | @Verified transmembrane differential pressure increases in order to determine the life span of the membrane |
| Transient characteristics | @Simulated the transient changes in the off-gas system and verified the resulting moisture separation capability |

Table 3 Moisture separation capability test and durability test items

| Test items | Content |
|---|---|
| Resistance to radiation | Conducted irradiation hypothesizing presence of noble gases and verified the resulting changes in moisture separation capability and durability. |
| Resistance to chemicals | Introduced chemical contaminants hypothesizing those found in off-gas systems and verified the resulting moisture separation capability and durability. |
| Resistance to heat | Conducted testing under environmental temperatures in excess of those hypothesized for an actual system and verified the resulting moisture separation capability and durability. |
| Resistance to water | Wetted the moisture separation membrane and verified the resulting moisture separation capability and durability. |

Table 4 @Laboratory basic testing parameters

| | |
|---|---|
| Aeration flow volume | 35 Nl/min per tube |
| Purge rate | 10% |
| Dehumidification intake gas temperature | 50 |
| Transmembrane pressure differential | 700 mmHg |

**Laboratory testing results**

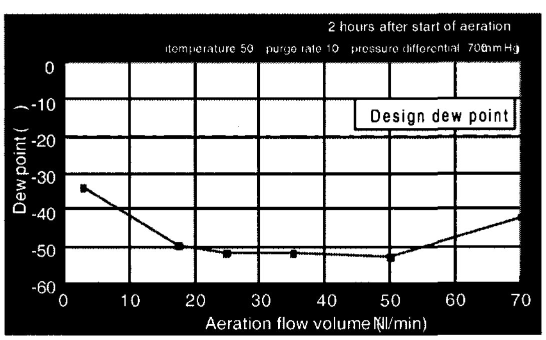
The results of all laboratory tests are summarized in Tables 5 and 6. The aeration flow dependence test and the pressure dependence test have been taken as representative examples of these and their results plotted on Graphs 1 and 2.

- Aeration flow dependence test

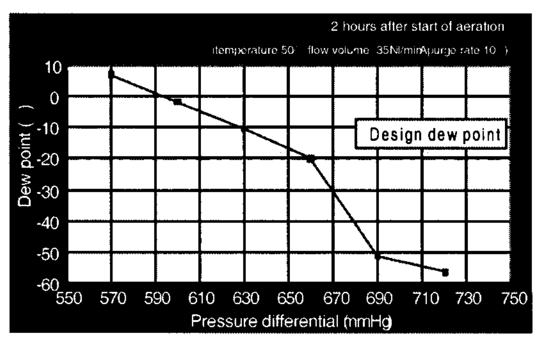
Graph 1 plots the moisture separator discharge dew point after two hours of aeration at a flow rate of 3-70 Nl/min. It may be seen from this that the dew point satisfies the design requirement of -20 or lower, and that the prescribed performance was obtained at all flow rates within the range covered by the test.

- Pressure differential dependence test

Results of the pressure dependence test show (Graph 2) that when the pressure differential is 660 mmHg or less, the dew point rises above the design dew point of -20 .




Graph 1 Aeration flow dependence test results




Graph 2 Pressure dependence test results

Table 5 Moisture separation capability test results

| Test Item | Test result |
|---|---|
| @Aeration flow dependence | Performance obtained for all volume flow rate. |
| @Gas temperature dependence | Design dew point requirements satisfied. |
| @Pressure dependence | Design dew point requirements satisfied if -660mmHg or less. |
| @Performance achievement testing | Performance obtained after approximately 10 minutes. |
| @Long-term stability | Stable operation verified for 1 month of continuous operation. |
| @Life span evaluation | No difference in performance observed before and after testing. |
| @Transient characteristics | Moisture separation capability fluctuates according to fluctuations in purge gas pressure. |

Table 6 Membrane durability test results

| Test items | Test result |
|---|---|
| Resistance to radiation | Almost no change in performance. |
| Resistance to chemicals | Almost no change in performance. |
| Resistance to heat | No change in performance. |
| Resistance to water | Tensile strength decreased by approximately 30%, but still maintained well within tolerance. |

## Evaluation

When the secondary side pressure is increased, the transmembrane pressure differential decreases and the dew point at the moisture separator discharge rises. Test results indicate that a dew point of -20 or lower can be maintained as long as the secondary side pressure is -660 mmHg or lower, however at -570 mmHg (equivalent to ANN of main condenser vacuum level Low Low) it rises to 8 . Given the assumed coolant temperatures for which the condenser is designed, a drop to this point is unlikely, but even hypothesizing that gas at a dew point of 8 flows into the charcoal hold-up units for one week, resultant in-flowing water consumes less than a mere 1 wt% of design margin charcoal volume. Consequently, there will be almost no decline in performance. Further, calculations based on the charcoal margin show that as long as the plant is not operated continuously for months at the 570 mmHg vacuum level, there will be almost no effect on the performance of the charcoal hold-up unit, and since such continuous operation is not expected in any case, this is not perceived as presenting a problem.

## In-Plant Test

### Test objectives

Since the results of laboratory testing suggested applicability of the membrane moisture separator to the off-gas system, we next organized the items to be subject to positive verification toward application in the off-gas system and conducted in-plant testing with the following objectives.

- Moisture separation capability for actual gas

While testing in the laboratory used uncontaminated gas that had been passed through a filter, in actual application it is possible that the gases in question could be contaminated by anti-corrosion agents from auxiliary boilers and/or sludge from within the piping system. Consequently, it was necessary to verify whether or not such contaminants would effect performance.

- Influence of plant transients on moisture separation capability

While the laboratory testing simulated plant start-up and normal operation states, it was also necessary to verify how moisture separation capability is effected by realistic fluctuations in the vacuum of the main condenser.

- Evaluation of life span in presence of actual gases

While long-term operation stability was studied through the laboratory testing, it was also necessary to verify the moisture separation capability and durability of the membrane under long-term operation in an actual environment.

**In-plant test apparatus**

The in-plant testing apparatus is outlined in Figures 6 and 7 and shown in Photo 3. The in-plant test apparatus has been installed between the off-gas condenser and dehumidifier unit within the off-gas system of Kashiwazaki Kariwa #1 unit operated by Tokyo Electric Power Company. This apparatus is comprised of the membrane moisture separator, vacuum pump equipment, valves and other equipment used in the laboratory testing. Gas is introduced from the off-gas condenser discharge line, passes through the testing apparatus, and returns near the point from which it is first extracted. While the system in question essentially does not require vacuum pump equipment, such equipment is included only in this in-plant test alone as a means of generating additional pressure for the purpose of returning the discharged gas to a point in the off-gas system near the place from which it originally issued. The purge gas is bifurcated at the moisture separator discharge, passed through a flow control valve to be introduced into the apparatus and finally drawn from the apparatus discharge into the main condenser.

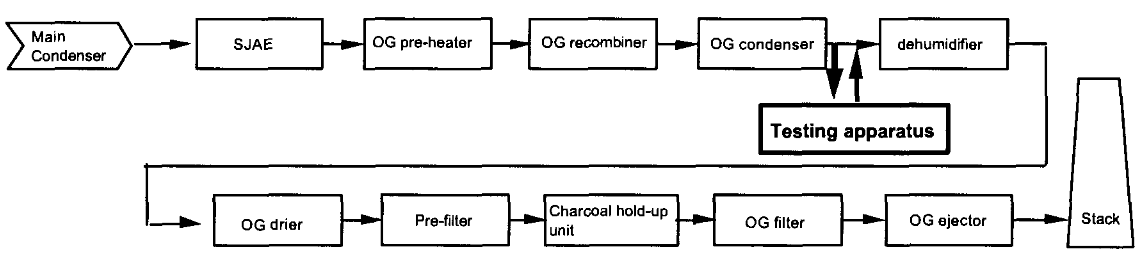



Figure 6 In-plant testing system diagram

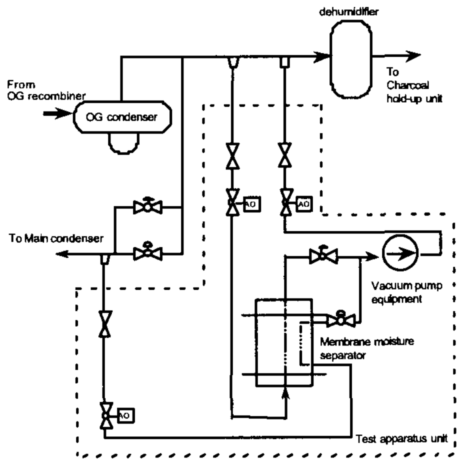


Figure 7 Outline of testing apparatus

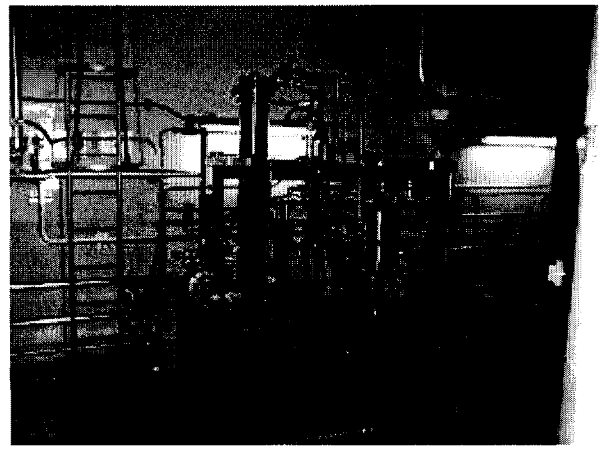


Photo 3 Overall view of testing apparatus

**In-plant test process**

We are in the process of conducting long-term operation testing. Plans for test implementations include two operational cycles (conducted over approximately two years) designed to test membrane durability.

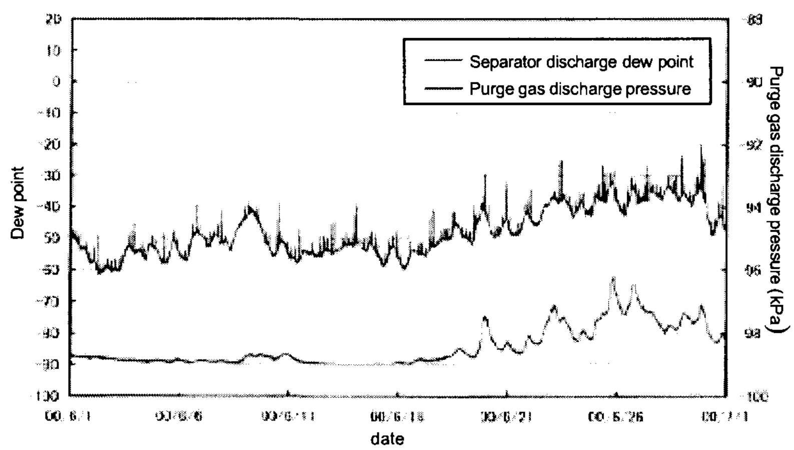
Continuous operation testing is being conducted using two moisture separation membrane modules installed in the testing apparatus, with the goal of verifying moisture separation capability and durability. Following the completion of the first operation cycle, the first membrane module will be replaced with a new module, and the used module will be dismantled and analyzed to determine the tensile strength and other factors pertaining to the membrane. The remaining module will be further used for the second operation cycle, after which it will be subjected to the same durability testing and comparative study as for the module used in the first round operation cycle.

**In-plant testing results**

In-plant test commenced in April 2000 and is currently in progress. Here we introduce some of the moisture separation capability results obtained so far. Evaluation of membrane durability is scheduled to be studied as the ongoing test makes the relevant data available.

Graph 3 shows fluctuations in the purge gas pressure and moisture separation capability during plant operation. These results indicate that moisture separation capability changes in accordance with fluctuations in purge gas pressure. With the exception of short period after the start-up of testing apparatus, dew point is within a range of -37 and -80 , thereby satisfying the -20 design requirements. Because of decreasing condenser vacuum levels accompanying rising seawater temperatures during the summer months, there are decreases in the transemembrane pressure differential, and these are accompanied by slight changes in moisture separation capability.

The moisture separation capability under various plant operation condition satisfies the design dew point of -20 , and the startup transition fluctuations have no significant effect on moisture separation capability.




Graph 3 Moisture separation capability during plant operation

**Conclusions**

With the objective of developing an off-gas system membrane moisture separator system, we first conducted laboratory testing, with results indicating that the membrane moisture separator is able to meet the performance and durability requirements needed for operation within an off-gas system, and therefore shows promise for plant application.

Further, we are currently implementing in-plant testing toward positive verification of applicability in actual systems, and so far this testing has been achieving favorable results for moisture separation capability. In the future we will evaluate aspects such as durability, and plan to conduct studies toward the introduction of this membrane moisture separator system in the off-gas system.