Study on gas migration behavior through bentonite buffer material

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Abstract

Gasses generated due to the corrosion of metals contained in the waste packages and engineered barrier materials will accumulate in the Engineered Barrier System (EBS) of a radioactive waste repository, and may affect long-term stabilities of the EBS. To evaluate gas migration behavior through the bentonite buffer materials in conjunction with a reasonable design model is one of the challenges to be solved for the performance assessment of a radioactive waste repository.
A series of studies have been conducted for more than 10 years under the leadership of the Radioactive Waste Management Funding and Research Center (RWMC) in Japan to better understand gas migration behavior through bentonite. In this report, the outline of the framework of the gas migration study by RWMC as well as some of the results from laboratory gas injection tests are presented.
The laboratory test results focusing on the interfaces in bentonite materials (bentonite/bentonite interfaces) showed that water flow through the interface was blocked (self-healed) due to the swelling of the bentonite. In addition, the gas injection test after full saturation, indicated no significant differences in breakthrough pressure compared to the intact (e.g. no interface) specimen. These findings suggest that the interface is unlikely to become a dominant gas migration pathway in a saturated buffer.
It is expected that all findings acquired through this decade of study are summarized and integrated within this fiscal year (2017) taking into consideration the parallel studies on gas scenario development and integration of coupled modelling, and contribute to a reliable advanced repository design and performance assessment.

1. Introduction

The long-term stabilities of radioactive waste repository are generally secured by a multibarrier system consisting of Natural Barrier and Engineered Barrier System (EBS). A certain amount of gas will be generated by corrosion of metals contained in the waste packages and EBS materials. Such gases will accumulate in the EBS of a radioactive waste repository and may affect the long-term stability of the EBS. To evaluate gas migration behavior through the bentonite buffer materials, in conjunction with a reasonable design model, is one of the challenges to be solved for the performance assessment of a radioactive waste repository [1].

Extensive studies on gas migration behavior have been carried out. One of the famous international study is the FORGE (Fate Of Repository GasEs) project funded by EC. The following common challenges for the understanding of gas migration properties have been identified through the FORGE project [2]:

- Treatment of anisotropic micro phenomena within a macro model.
- Treatment of coupled phenomena considering both EBS stability and nuclide migration over time.
- Up scaling from laboratory to realistic scale with regard to phenomena understanding.

Based on the issues identified from the findings of the in-situ large-scale Gas Migration Test (GMT, 1997~2007) at Grimsel Test Site (GTS) [3], RWMC has conducted a series of studies from 2008 to better understand gas migration behavior through bentonite.

In order to find solutions to the internationally identified issues with regard to gas migration in EBS, research and development focused on the following three items: obtaining better date through laboratory testing, advancement of analysis tools and scenario development [4].

In this report, the outline of the framework of the gas migration study by RWMC, as well as some of the results from laboratory saturation and gas injection tests, are presented.

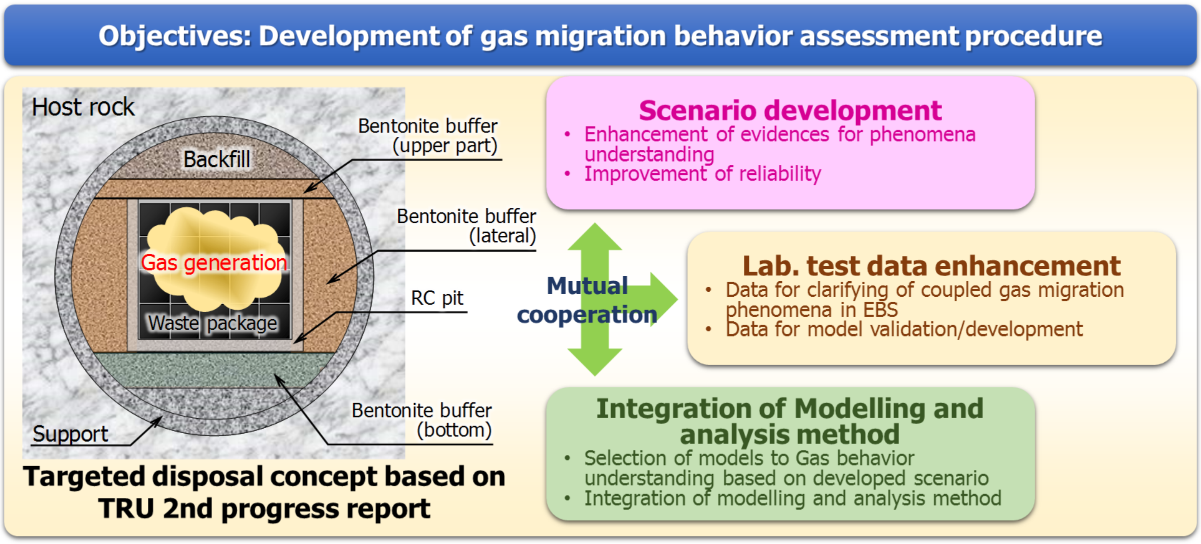


Fig. 1 Overview of RWMC's study on Gas migration

2. Studies on gas migration by RWMC

The studies on gas issues, e.g. gas generation, gas migration in EBS, and effect on long-time repository stabilities, are being conducted by JAEA and NUMO for Japanese HLW and Transuranic (TRU) waste repositories [5], [6], [7]. Especially in Japanese TRU waste repository, the following phenomena or critical events have been considered in migration of gases generated in the waste packages, that could significantly influence the performance of the disposal system:

- Gas transport and diffusion to the outside of the EBS in the form of dissolved gas in groundwater.
- Gas pressure pushing contaminated water out from the waste package and disposal cavern into the geosphere.
- Excess gas/water pressure causing stress on the EBS components leading to mechanical failures and preferential pathway creation, allowing gas/water to seep out.

These phenomena have been considered in the groundwater advection scenario as increased nuclide migration due to the expulsion of contaminated water resulting from gas generation in the EBS. The performance assessment has been conducted with the basic expulsion model. It showed that the maximum dose calculated was not significantly different from the results of the reference case.

Due to the background mentioned above, the still ongoing study on gas migration in Japanese TRU waste repository was initiated in 2008 by RWMC with the purpose of reducing the uncertainties in the performance assessment of a multibarrier system. Fig. 1 shows the interrelationship of each activity of RWMC's gas study project.

RWMC's study on gas has been ongoing for over ten years in two phases. In the first phase (2008~2012), studies on gas migration behavior were conducted that take into consideration the issues identified in the TRU 2nd progress report [6], specifically laboratory gas injection tests for understanding gas/water permeability in bentonite material, development of integrated modeling and analysis method, and scenario assessment with consideration of the transition of repository. While substantive findings relevant to gas migration in bentonite were acquired through the first research phase, several issues, which arose from uncertainties of gas behavior, remained.

In the subsequent second phase (2013~2017), laboratory tests targeted at not only bentonite materials but also cementitious materials have proceeded to enhance data related to gas/water permeability, which contribute to reduce the uncertainties of gas migration behavior. In addition, modeling approaches to apply the parameters estimated from laboratory test results have also been investigated to trace laboratory test sequence with higher reliability.

Through a series of studies, it has been foreseen that the interfaces between bentonite materials may induce preferential pathways for gas generated in a waste package (cf. Fig. 2). RWMC has therefore

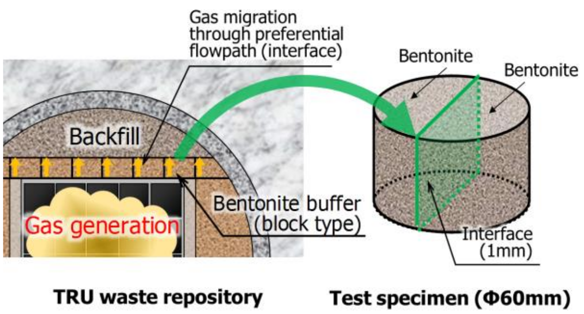


Fig. 2 Preferential gas pathways due to interfaces

initiated a laboratory gas injection test program to evaluate the effect of interfaces (bentonite/bentonite interfaces), which are expected to exist in bentonite materials, on gas migration behavior. The gas injection tests have focused on three phenomena, (re-)saturation, gas injection, and gas breakthrough (sudden flow increase), in a potential gas migration scenario.

The latest findings through the gas injection test will be discussed in the next chapter.

3. Laboratory tests focusing on the existence of interface in bentonite

(1) General idea and condition of the test

The laboratory equipment consisting of gas/water injection system, gas/water outflow measurement system, test column, and operation panel was set up prior to initiation of the gas injection test. Two types of Na bentonite (Kuni-gel V1) column specimens, with and without interface were prepared respectively. Both specimens had a diameter of 60mm and a height of 25mm and were placed into the column of the test equipment as shown in Fig. 3. The specimen was compression-molded as two layers to reach the required dry density. The specimen without interface was named Case-1 and the one with interface was named Case-2. With respect to the initial condition of both specimens, the dry density of 1.36 Mg/m^3 was set as referred to in the concept of the TRU 2nd progress report. The water content of 32.4% (saturation ratio of ca. 90%) was set to reduce the duration of saturation (cf. Tab. 1). The specimen was compression-molded statically (compression speed of 1mm/min) by two layers to reach the required dry density. As for the Case-2, a plate of 1mm thickness was set at the center of the column container which was removed after compression.

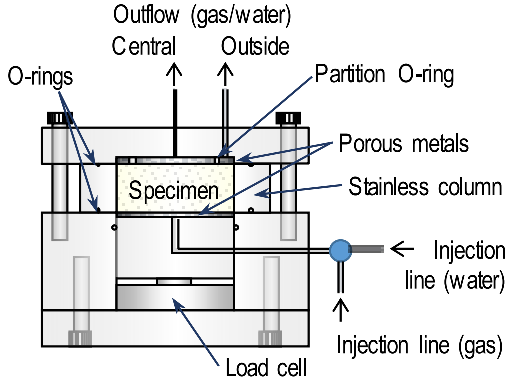
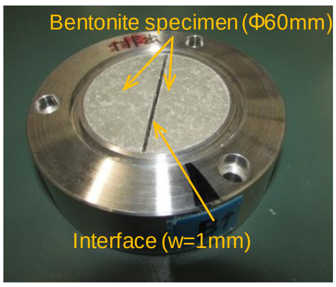


Fig. 3 column test equipment

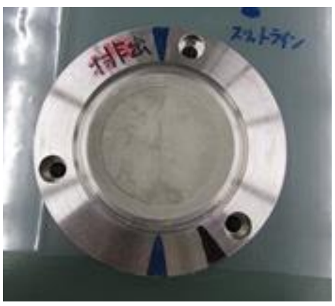
Tab. 1 Specification of specimen

	Case-1	Case-2
Interface	No interface	Interface (1mm)
Dimensions	Φ60mm × h25mm	
Material	Na type bentonite (Kuni-gel V1)	
Dry density	ρ_d=1.36 Mg/m^3	
Initial water content	wt=32.4% (Sr=90%)	

The (re-)saturation phase was initiated by water injection from the bottom of the column. At the beginning of water injection, no water pressure was applied to prevent the appearance of flow paths at the outer peripheral surface of the column specimen, and saturation proceeded only by capillary pressure of the bentonite material. After confirmation of stable water injection, the injection pressure was increased to 0.2



Before water injection



After saturation

Fig. 4 Bentonite column with interface

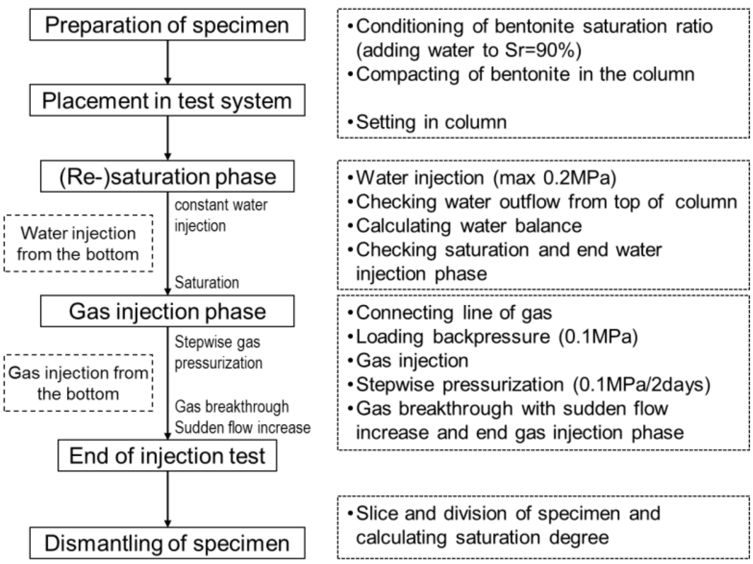


Fig. 5 Flow chart of test procedure

MPa which was lower than the expected swelling pressure (ca. 0.5 MPa) of the bentonite specimen. Full saturation of the specimen was comprehensively assessed by multiple factors such as water seepage from the top surface, total volume of injected water, flow balance of inflow and outflow, water permeability, and pH. Two fully saturation specimens were made for Case-2 so that the actual saturation ratio could be assessed by dismantling one specimen before proceeding to the gas injection phase. Fig. 4 shows the top view of the Case-2 specimen: before initiating water injection (top), and after saturation (bottom). The interface of the bentonite self-healed due to the swelling.

After confirming saturation of the bentonite specimen, the gas injection phase was initiated by closing the water injection valve and opening the gas injection valve, both set at the bottom of the column specimen. Nitrogen gas was injected under a backpressure of 0.1 MPa after which the gas injection pressure was increased by stepwise pressurization of 0.1 MPa per two days until gas breakthrough (sudden flow increase) occurred. The temperature of laboratory was kept at 25 degree centigrade during all test sequences.

Fig. 5 shows the flow chart of the test procedure.

(2) Test results of Case-1: without interface

As test result of the (re-)saturation phase of Case-1, cumulative amount of water inflow, water injection pressure, and swelling pressure during the water injection are summarized in Fig. 6. In the first 3 days of the water injection, the saturation proceeded only by capillary pressure of the bentonite as there was no water pressurization. After that water injection commenced at 0.2 MPa injection pressure.

Water seeping from the top surface was observed after 36 days. The cumulative amount of injected water at that time was around 8.4mL which was reasonable considering the volume of the water needed for saturation of the specimen (3.49 ml) and void volume of the porous metal set onto the top surface of the specimen (3.86 ml). The water permeability determined was $6.6x10^{-15}$ m/s, which is a lower value compared to past research for the 1.36 Mg/m^3 dry density of bentonite (10^{-11}~10^{-13} m/s). The pH value of the outflow was weak alkaline at around 8 which suggested that the water came from the bentonite specimen. Based on a comprehensive assessment of these results, the water injection / saturation phase was finished. As Fig. 6 indicates, the swelling pressure measured during the test was 0.47 MPa.

Following the saturation phase, the nitrogen gas injection phase was initiated. A back pressure of 0.1 MPa was applied to the specimen in accordance with the test procedure, then the gas injection increased stepwise by 0.1 MPa per two days. Fig. 7 shows the change of effective gas pressure and gas inflow during gas injection. As result of the gas injection increase of 0.1 MPa per two days, the gas breakthrough with sudden flow increase occurred after 29 days, when gas injection pressure reached 1.6 MPa. Gas injection was automatically stopped after the gas flow rate reached 1,000 Nml/min due to the operation of a safety valve, at which time the whole test sequence of Case-1 was completed.

(3) Test results of Case-2: with interface

As test result of the (re-)saturation phase of Case-2, which included an interface at the center part of the column specimen, cumulative amount of water inflow, water injection pressure, and swelling pressure during the water injection are summarized in Fig. 8, as with the Case-1 results. In Fig. 8, the swelling pressure was calculated by subtracting water pressure from the total pressure measured at the bottom. After 42 days from the start of water injection, water seeping from the top surface was observed and it was confirmed that the total amount of inflow and outflow were consistent. After filling the outflow piping with water, the measured inflow rate and outflow rate were balanced to determine the permeability of the bentonite specimen. The water permeability estimated was on the order of 10^{-13} m/s which is consistent with past research results. Based on a comprehensive assessment of these results, the saturation phase was finished after 100days of water injection at which time the observed swelling pressure was 0.33 MPa. The swelling pressure was within the variation range of past research [8] although the pressure was slightly lower than for Case-1 (no interface).

The gas injection phase was initiated after confirming the saturation by the same procedure (nitrogen gas injection, and pressurization of 0.1 MPa per two days) as for Case-1. The change of effective gas pressure, water outflow from the top surface, and effective stress at the column bottom during gas injection phase are shown in Fig. 9. As the result of the stepwise increase of gas injection, effective stress at the bottom gradually decreased accompanying the stepwise pressurization. Gas breakthrough occurred after 25 days when gas injection pressure reached 1.3 MPa, which is slightly lower than observed for Case-1. The gas injection was stopped afterwards and the test specimen was dismantled to investigate the inside of bentonite material.

Fig. 10 shows the distribution of water content inside the Case-2 column specimen before gas injection (left) and after gas breakthrough (right). Both profiles were measured from sliced fragments by dismantling as two saturated specimens were prepared. The water content before gas injection decreased from the bottom toward the top of the column. Moreover, it was confirmed that the water content around the interface is slightly higher, compared to other areas. One of the reason for the variation of water content from the bottom to the top is thought to be due to the time lag of swelling and consolidation in terms of places and time with the water injection from the bottom to top. The time lag of swelling causes spatial differences of density and the water content will change accordingly. Similar phenomena are likely to occur around the gap 1mm wide. In the profile of the after gas breakthrough sample, a decrease of water content from the top toward the bottom is observed, but no significant differences are seen between areas in the vicinity of the gap and other areas. One of the reason of this phenomena is thought to be that the gas progressed gradually inside of the specimen from the bottom and pushed out the pore water at the same time. Continuation of gas flow after gas breakthrough

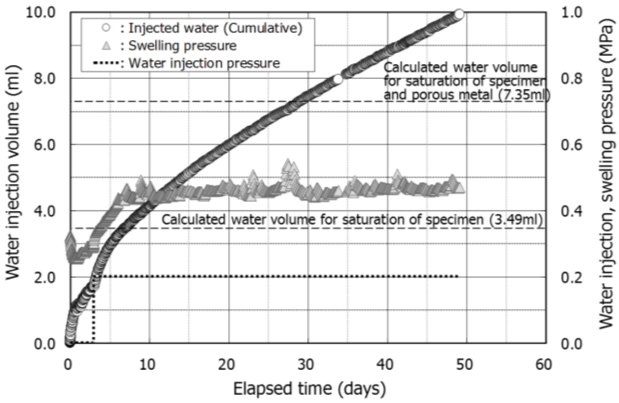


Fig. 6 Saturation phase (Case-1)

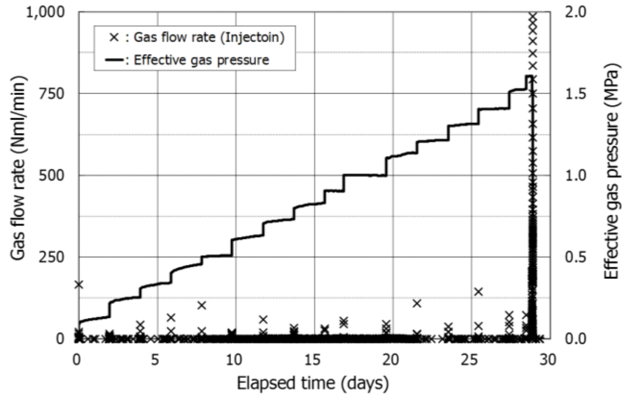


Fig.7 Gas injection phase (Case-1)

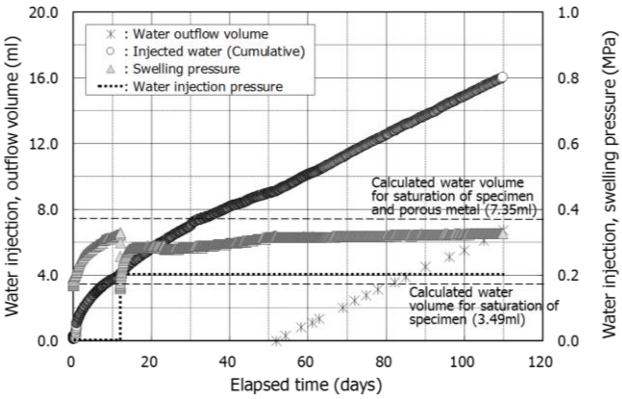


Fig. 8 Saturation phase (Case-2)

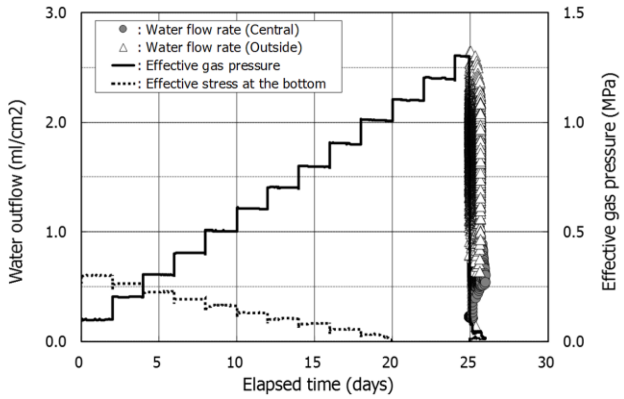


Fig.9 Gas injection phase (Case-2)

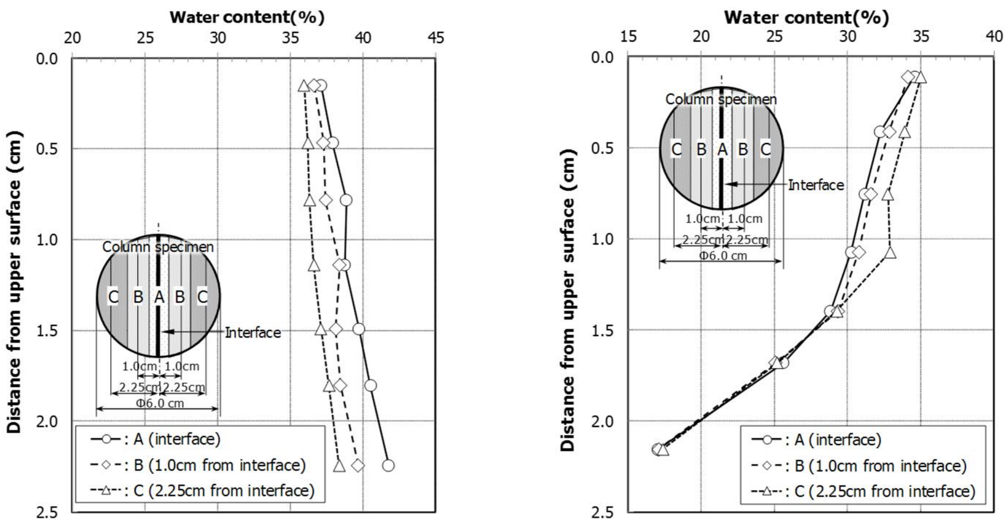


Fig.10 Measurement of water content inside the bentonite Case-2, after saturation (left), and after breakthrough (right))

could also be a factor reducing the water content. Meanwhile, the water content at the top surface of the specimen before gas injection and after breakthrough show almost identical values. This could be because the gas breakthrough occurred before the gas front reached the top surface of the specimen, although it must be noted that both specimens were not the same.

(4) Lessons Learned from laboratory tests

The gas breakthrough pressure obtained through gas injection test was 1.6 MPa in Case-1 (no interface) and 1.3 MPa in Case-2 (with interface). Both pressures were slightly different but reasonable well within the range of variation observed in past studies. Therefore, it is thought that the existence of the interface has no effect on the gas breakthrough pressure although this is based on the limited results with a 1mm wide interface. The profile of water content after gas breakthrough does not show obvious differences between interface vicinity and other areas and this means that the interface does not function as a preferential gas pathway. In addition, it is recognized that the interface may affect the distribution of water content and dry density in an extremely small range.

As 1mm width of the interface was chosen for this study, the effect of different widths would be an issue to be confirmed in further studies.

4. Conclusions

As the latest findings through the RWMC's study on gas migration behavior which proceeded over ten years, the laboratory gas injection test to evaluate the effect of interfaces expected in bentonite material was discussed in this paper.

The major findings of this study are:

- The existence of interface has no significant effect on the gas breakthrough pressure.
- The interface does not play a role of a preferential gas pathway.
- The interface may affect the distribution of the water content and dry density in an extremely small range.
- The effect of the width of interfaces would be an issue to be confirmed in future studies.

Further challenging test program focusing on the width of interface and the interface between different materials are proceeding under the direction of RWMC.

It is expected that all findings acquired through this decade of study are summarized and integrated within this fiscal year (2017) taking into consideration the parallel studies on gas scenario development and integration of coupled modeling, and contribute to the reliable advanced repository design and performance assessment.

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