Multiple roles of clays in radioactive waste confinement – introduction

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Abstract: Geological disposal provides the safe long-term management solution for higher-activity radioactive waste. The development of a repository (or geological disposal facility) requires a systematic and integrated approach, taking into account the characteristics of the waste to be emplaced, the enclosing engineered barriers, and the host rock and its geological setting.

Clays and clayey material are important in the development of many national geological disposal systems. Clays exhibit many interesting properties, and are proposed both as host rocks and as material for engineered barriers. Whatever their use, clays present various characteristics that make them high-quality barriers to the migration of radionuclides and chemical contaminants. As host rocks, clays are, in addition, hydrogeologically, geochemically and mechanically stable over geological timescales (i.e. millions of years).

As discussed in Norris (2017), many countries have chosen to dispose of all or part of their radioactive waste in facilities – typically referred to as a repository or a geological disposal facility – constructed at an appropriate depth in stable geological formations. The associated geological disposal system consists of the radioactive waste, the engineered barriers and the natural geological barrier; it is designed to inhibit the release of radioactivity.

In terms of long-term environmental safety, a suitable site for a repository will be one where the geology and appropriately designed engineered barriers contain the radioactivity, and any potential non-radiological hazards presented by the waste, for as long as required. The waste, the engineered barriers and the host-rock elements of the geological disposal system fulfill, separately and in a complementary fashion, multiple safety function roles (e.g. isolation, containment). The principle of 'defence-in-depth' is employed: multiple levels of protection are designed to enhance safety through their diversity and redundancy.

This Special Publication highlights the importance of clays and clayey material – also referred to as argillaceous media – in the development of almost all national geological disposal systems, either in the role of the repository host rock or as part of the repository engineered barrier system (EBS); the reader is referred to Norris (2017) and websites of national waste management organizations (e.g. UK Government 2018; Andra 2019; Bundesgesellschaft für Endlagerung mbH (BGE) 2019; COVRA 2019; Nagra 2019; NWMO 2019; ONDRAF/NIRAS 2019; Ontario Power Generation 2019; Posiva 2019; PURAM (RHK Kft.) 2019; SKB 2019; United States Department of Energy 2019) for further information. Note that such programmes publish extensively, and, as such, their related websites are a rich source of high-quality, externally peer-reviewed, citable literature covering clays and many other aspects of their activities.

Research and development studies performed internationally by such waste management organizations over several decades have highlighted the favourable properties of clays in relation to geological disposal (Norris 2017):

- Very little water movement: thanks to their low permeability, there is practically no water movement in clays. Radionuclide and chemical contaminant transport via this medium are thus strongly delayed.
- Diffusive transport: given the limited water movement, transport in clays is essentially diffusive, which means that species migrate primarily under the influence of their concentration gradient, and migration under the influence of the pore-water movement is minimal.
- Retention capacity: clays have a strong retention capacity for many radionuclides and chemical contaminants. Their migration through clays is thus considerably delayed.
- Buffer effect: clays display a significant buffer effect with regard to chemical perturbations. The thickness of the clay that is chemically perturbed by the disposal facility is therefore very limited.

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- Self-sealing capacity: clays show a high capacity for self-sealing. Any fractures and fissures that occur, in particular those created by excavation activities, close quite rapidly.
- Stability: the selected clay host rocks – and therefore their favourable properties – have remained unchanged over millions of years. The migration of natural chemical species through these clay host rocks has remained diffusive during at least the last million years.
- Vertical homogeneity: radionuclide and chemical contaminant transport properties are very homogeneous throughout almost the entire thickness of the selected clay host rocks.
- Lateral continuity: clays are present within simple geological structures, with a significant lateral continuity, which facilitates their large-scale characterization.

The national waste management programmes of France (Andra 2019) and Switzerland (Nagra 2019) have chosen to dispose of their high-level and long-lived intermediate-level radioactive waste in indurated clays in the Callovo-Oxfordian and Opalinus Clay formations, respectively; in Switzerland, Opalinus Clay is also the proposed host rock for low- and intermediate-level waste. In Belgium, the technical solution recommended for the long-term management of high-level and long-lived low- and intermediate-level waste is geological disposal in poorly indurated clay (Boom Clay or Ypresian clays) (ONDRAF/NIRAS 2019). In The Netherlands, clays are considered as a potential host rock for the disposal of all types of radioactive waste (COVRA 2019). Other countries also consider the use of clays as host rock (e.g. Radioactive Waste Management 2019; NWMO 2019).

Clays may also contribute to the safety of geological disposal systems whose host rock is not clay by being present in their geological environment. For example, in Germany, the disposal facility for low- and intermediate-level waste is under construction in an old iron mine (limestone) located under a clay formation (BGE 2019); and in Canada, the host rock proposed for the disposal of low- and intermediate-level waste is limestone situated beneath a sequence of clay formations (Ontario Power Generation 2019).

In addition to natural clay units being considered for a repository host rock or present in the geological environment, properties of clays such as bentonite make them a material of choice for the repository EBS. (Bentonite is used to refer to smectite-rich material (regardless of origin) with favourable chemical, hydraulic and mechanical properties. Bentonite is the primary buffer and backfill material in several high-level waste and spent fuel disposal concepts. It has also been proposed for use as a buffer and/or backfill in some intermediate-level waste disposal concepts.) The multiple roles that clays are mainly planned for use here include:

- Buffer material: voidage between the disposal package and the host rock is filled with bentonite. As the clay saturates following emplacement, through contact with groundwater from the host rock, it swells and provides a low hydraulic conductivity barrier, enabling the container to be protected from deleterious processes, such as corrosion. The characteristic swelling behaviour of bentonite is due to the presence of significant quantities of sodium montmorillonite.
- Backfill material: clay (for instance, in the form of blocks or pellets) is used to fill excavated spaces (placement rooms, access ways), sometimes in combination with other materials.
- Sealing material: clay, sometimes in combination with other materials, is used to isolate parts of the disposal facility (e.g. vaults used for waste emplacement, tunnels connecting vaults, access ways). Seals and plugs are works of limited dimensions with a specific purpose placed at key locations in the disposal facility. For instance, in France, the seals aim to limit water flow within the underground facilities (e.g. see Posiva 2016 for additional information).

The geological disposal facilities in operation, under development or being planned in many countries thus contain, or are proposed to contain, clay, whatever the selected host rock. Table 1 (updated from Norris 2017) lists some of these facilities, sorted according to the progress of the national geological disposal programme. The use of clay as buffer, backfill or in sealing is indicated.

Although several decades of research and development studies have already been performed internationally in relation to clays in the context of geological disposal and radioactive waste confinement, waste management organizations responsible for implementing geological disposal – for example, Andra (France), COVRA (The Netherlands), KORAD (South Korea), Nagra (Switzerland), NUMO (Japan), NWMO (Canada), ONDRAF/NIRAS (Belgium), Posiva (Finland), PURAM (Hungary), RWM (UK), SKB (Sweden) and SÚRAO (Czech Republic) – are undertaking significant programmes of research, both nationally and internationally, to continue the advancement of the understanding of clays, their performance and behaviour in the context of an evolving repository recognizing coupled thermal, hydrogeological, mechanical, chemical and gas processes, and their contribution to ensuring safe long-term management of long-lived radioactive waste:

- Multidisciplinary approaches, including geology, mineralogy, geochemistry, rheology, physics

Table 1. *Use of clays as buffer, backfill or sealing materials in operational and planned national disposal facilities*

	Buffer	Backfill	Sealing
Operational facilities			
USA (United States Department of Energy 2019) Waste types: long-lived low- and intermediate-level military waste Host rock: salt			x
Hungary (PURAM (RHK Kft.) 2019) Waste types: short-lived low- and intermediate-level waste Host rock: crystalline rock (granite)			x
Applications submitted for disposal facility construction licence			
Canada (Ontario Power Generation 2019) Waste types: low- and intermediate-level waste Host rock: limestone overlain by clay			x
Sweden (SKB 2019) Waste type: irradiated fuel Host rock: crystalline rock (granite)	x	x	x
Finland (Posiva 2019) Waste type: irradiated fuel Host rock: crystalline rock (granite)	x	x	x
Host rock selected			
France (Andra 2019) Waste types: long-lived intermediate-level and high-level waste Host rock: clay		x	x
Switzerland (Nagra 2019) Waste types: irradiated fuel and long-lived low- and intermediate-level waste Host rock: clay	x	x	x
Host rock not yet selected			
Canada (NWMO 2019) Waste type: irradiated fuel	x	x	x
UK (UK Government 2018) Waste type: intermediate level, high level, irradiated fuel	x	x	x

and chemistry of clay minerals and assemblages, are required in order to provide a detailed characterization of the geological host formations considered for the disposal of radioactive waste and to assess the behaviour of engineered and natural barriers when submitted to various types of perturbations induced by such facilities.

- The evaluation of the performances of the natural barrier, as well as of the impact of repository-induced disturbances upon the confinement properties of clay-rich geological formations, constitute major objectives for the experimental programmes being conducted or to be conducted in underground research laboratories, for interpreting the subsequent scientific results, for modelling the long-term behaviour of radioactive waste repositories and for carrying out safety assessment exercises.

The 20 papers published in this Special Publication provide insight for the reader into the range of clay-related work currently being undertaken internationally in relation to natural and engineered barriers for radioactive waste confinement. They have been classified according to different areas within the field of disposal of radioactive waste research. The assignment of a study to one of the topic areas was not always easy because many studies provide information of, and for, different aspects of disposal research. Topic areas covered by papers in this Special Publication consider research into clays and clayey material in the following topic areas.

Characterization of clay rocks

It is necessary to investigate clays that may be used either to host a repository or that could be part of the surrounding geological environment in order to understand, for example, their formation histories, their mineralogies, their physical and chemical properties, and how groundwater and gas could interact and potentially move through them.

Rabaute *et al.* (2018) consider logging data measured at the Benken borehole in Switzerland, which penetrated the Middle Jurassic Opalinus Clay Formation. The paper presents a statistical methodology to improve the description of the physical properties of the clay rock based on the well-log data. The methodology involves the classification of a set of local statistics, calculated from a reduced number of principal components computed from well-log properties. The use of a kernel-based method to calculate local statistics allows the analysis of spatial variability at different scales, and scale effects. The derived spatial variability of the log-units property is compared with earlier lithological descriptions and stratigraphic data.

Alcolea Rodríguez *et al.* (2018) present a workflow for the interpretation of well logs defined as an optimization problem. The workflow is applied to the characterization of metre- to decametre-scale stratigraphic units along 13 boreholes in northern Switzerland (1D resolution) and to millimetre-scale features over a wall at the Mont Terri underground rock laboratory (URL) in Switzerland (2D resolution). The results show that: (1) the workflow accurately maps lithological changes; (2) the interaction with the analyst is minimized, which reduces the subjectivity of the interpretation; and (3) outputs are available for on-site decisions.

A prediction–evaluation approach is developed and discussed in **Papafotiou *et al.* (2018*b*)** to assess the propagation of parameter, conceptual and scenario uncertainties in the estimated near-field temperatures of the full-scale emplacement experiment at the Mont Terri URL, Switzerland (penetrating Opalinus Clay). The uncertainty assessment is performed using a 3D thermohydraulic numerical model of the full-scale emplacement experiment that represents the emplaced materials and surrounding Opalinus Clay, and accounts for heat generation at the heaters. The propagation of parametric uncertainties is assessed using a first-order second-moment method supplemented by Monte Carlo simulations sampling the uncertain parameter space.

Boda Claystone is a very tight clayey rock with extreme low porosity and permeability, nano-size pores, and small amounts of swelling clays. Due to this character, it is ideal as a potential host rock for research into the possibilities of high-level waste deposition in geological formations and is considered in the Hungarian programme. Although the national programme research started more than 30 years ago, the genesis, the geotectonic history of the Boda Claystone Formation (BCF) and the geology of surrounding areas has only been sketched out recently. **Fedor *et al.* (2018)** report recent research, including advances in equipment and methodological developments, that have been progressed; these new results of BCF research help in the preparation of more sophisticated and directed experiments, in which there is a great interest internationally.

Liu *et al.* (2019) report progress within the Chinese radioactive waste management forum. Deep geological disposal of high-level radioactive waste in a repository with a system of engineered and natural barriers has been recognized as an appropriate disposal concept by Chinese authorities since 2003, and both crystalline rocks and argillaceous rocks are considered as the candidate host rocks for a high-level waste (HLW) disposal repository. On the basis of detailed ground geological, hydrological and geophysical surveys, two test boreholes in the Tamusu area revealed that there are three lacustrine facies clay formations. The spatial extension of clay formations could meet the fundamental criteria to ensure the long-term safety of the repository. Initial mineralogical studies on core samples indicated that the mineral assemblage is dominated by analcite, kaolinite, illite and dolomite. The homogeneous argillaceous rocks rich in analcite in the Tamusu area could be a new type of host rocks for HLW disposal repository.

Water, solute and gas transport

Hydraulic testing has revealed dramatic underpressures in Paleozoic shales and carbonates at the Bruce nuclear site in Ontario, Canada. Although evidence from both laboratory and field studies suggests that a small amount of gas-phase methane could be present in the shale, previous studies examining causal linkages between the gas phase and the underpressure have been inconclusive. To better elucidate processes in such a system, **Plampin & Neuzil (2018)** use a 1D representation of the site to test the effects of various factors on the evolution of gas-phase methane and pressures within the system, concluding that the presence of multiple fluid phases is unlikely to explain the underpressure at the site. This is consistent with prior conceptualizations of the underpressured section as a thick aquiclude, in which solute transport occurs extremely slowly, bounded by aquifers of significantly higher permeability.

Gas migration

Waste emplaced in a repository may, for example, on corrosion or degradation, generate gases. It is necessary, as part of the consideration of repository evolution in the long term, to understand how gas could interact with components of the disposal system, including clays in the EBS and clays in the natural barrier.

In the Canadian programme, bentonite-based materials are currently the preferred choice of seal materials for use in the repository. Understanding the long-term performance of these seals as barriers against gas migration is an important component in the design and long-term safety assessment of a Canadian deep geological repository. The study by **Dagher *et al.* (2018)** proposes a hydromechanical linear poroelastic viscocapillary mathematical model for advective–diffusive controlled two-phase flow through a low-permeability expansive medium (bentonite). The results were verified against experimental results found in the current literature. Parametric studies were performed to determine the influence on the flow behaviour. Based on the results, the mathematical model looks promising and will be improved to model flow through preferential pathways.

A key component of the site comparison planned for the deep geological disposal of spent fuel and high-level waste (SF/HLW) in Switzerland is the assessment of the evolution of repository-induced perturbations in the repository near field associated with thermal effects from heat production due to the radioactive decay of radionuclides, as well as gas pressures developing in the backfilled underground structures from the anaerobic corrosion of the steel waste canisters and tunnel support materials. **Papafotiou *et al.* (2018*a*)** presents the development of a methodology for an indicator-based assessment of heat- and gas-induced effects in a SF/HLW repository in Opalinus Clay integrating a probabilistic treatment of parametric uncertainty. The workflow is demonstrated using preliminary data, repository configurations and indicators.

Geology and hydrochemistry

If there is high water inflow into a repository local to a bentonite barrier, the water pressure may act on the emplaced bentonite and piping, and erosion may occur; this can lead to the formation of channels in the bentonite. Bentonite might also be eroded in dilute waters as a result of chemical processes that lead to bentonite swelling, and the formation and dispersion of colloids.

In considering how a repository for higher-activity wastes could evolve, it is necessary to undertake demonstration experiments – often complemented by modelling – at various scales to inform understanding. To understand piping and erosion, in the study of **Jo *et al.* (2018)**, the outflow behaviour and the condition of buffer materials are investigated using a test pit drilled into host rock at the Horonobe URL, Japan, to consider countermeasures to contain the outflow of the buffer material.

Colloid concentration is an important parameter in models of colloid-facilitated transport. The purpose of the study reported in **Sasamoto & Onda (2018)** is to characterize colloid concentrations and colloid stability in natural groundwater from the Horonobe URL in Hokkaido, Japan. Colloids in Horonobe groundwaters appear to be less stable, with a moderate potential for transport, than colloids investigated in similar international studies. This reduced stability may be due to relatively higher ionic strengths and moderate dissolved organic concentrations in Horonobe groundwaters compared to their international counterparts.

Hydraulic and diffusion processes

Bentonite-based buffer materials play an important safety role in engineered barriers in geological disposal repositories for higher-activity radioactive wastes. The effectiveness of buffer materials is dependent on the status of groundwater saturation during resaturation of the repository. Accordingly, it is important to determine the behaviour of buffer materials during saturation and to predict post-saturation conditions such as the distribution of residual dry density and chemical alteration.

In the study of **Ishii *et al.* (2019)**, the rate of groundwater uptake into a buffer material was determined to clarify the behaviour of the material during the saturation process. As mechanical changes and chemical alteration of buffer materials are generated by groundwater permeation, knowledge of the water uptake rate is necessary for the prediction of post-permeation conditions. In the experiment reported here, 1D permeation by distilled water and a NaCl water solution at a constant rate was monitored over a period of more than 7 years. The results indicated that the seepage and saturation front moved in proportion to the square root of the seepage time. The coefficient of the relationships between the seepage and the saturation fronts with time of the reference bentonite used in Japan was determined.

Hydromechanical processes

Giot *et al.* (2018) present work to help understand how cracks in a clay material can evolve on exposure to water, which is potentially relevant to the evolution of the host rock immediately adjacent to the repository that may have been perturbed during construction and operational periods.

Self-sealing tests were carried out on cylindrical samples artificially cracked on one-third of the diameter with a perfectly controlled aperture. Water was then injected into the crack. An innovative cell was developed and used, the body of which is transparent to X-rays. The sample could fully rotate in the nanotomograph, allowing a 3D reconstruction of images before, during and after tests, a visualization of the evolution of the cracked zone, and a quantification of the variations in crack volume during self-sealing. Permeability measurements were made to quantify the influence of self-sealing on flows. Two facies of claystone with different calcium carbonate contents were tested.

Sorption

It is important to understand how radionuclides, as could be present in disposed waste and potentially subject to migration from a repository and through the host rock, could interact with rock itself, possibly leading to retardation.

The speciation of selenium (Se) in clay-rich host rocks is important within the framework of geological disposal of radioactive waste since it affects its migration. Removal of selenite from solution can occur by reduction and adsorption. Adsorption could potentially inhibit or delay reduction. In **Hoving *et al.* (2019)**, the interplay of adsorption and reduction of selenite is investigated in batch experiments with Boom Clay (BC) and its separated size fractions. In all experiments, dissolved Se concentrations (Se_{aq}) showed a fast initial decrease that was followed by a slower decline until removal was almost complete. The progress of Se_{aq} removal and Se^{IV} reduction to Se^{0} could be described by a kinetic model involving reversible adsorption on clay minerals and reduction by pyrite.

Process at interfaces

Radioactive waste disposal relies on a multi-barrier concept that includes engineered components – which, in many cases, include a bentonite buffer surrounding waste packages – and the host rock. Contrasts in materials, together with gradients across the interface between the engineered and natural barriers, lead to complex interactions between these two subsystems. Numerical modelling, combined with monitoring and testing data, can be used to improve our overall understanding of rock–bentonite interactions and to predict the performance of this coupled system.

The Prototype Repository (PR) tunnel is located at the Äspö Hard Rock Laboratory near Oskarshamn in the SE of Sweden. In the PR tunnel, six full-sized deposition holes (8.37 m deep and 1.75 m in diameter) have been constructed. Each deposition hole is designed to mimic the Swedish reference system for the disposal of nuclear fuel, KBS-3V. The PR experiment is designed to provide a full-scale simulation of the emplacement of heat-generating waste. **Tsitsopoulos *et al.* (2018)** report a coupled integrated thermal–hydrological model for predicting the wetting and the temperature of bentonite emplaced in fractured rock, accounting for the heterogeneity of the fractured rock. The adopted approach provides an evaluation of the predictive capability of models; it gives an insight into the uncertainties in the data and demonstrates that a simplified equivalent homogeneous description of the fractured host rock is insufficient to represent the bentonite resaturation.

Finsterle *et al.* (2018) report a study in which different conceptualizations and modelling tools were used to analyse the Bentonite Rock Interaction Experiment (BRIE) conducted at the Äspö Hard Rock Laboratory in Sweden. The range of predicted bentonite wetting times encompassed by the ensemble results were considerably larger than the ranges derived from individual models. This is a consequence of conceptual uncertainties, demonstrating the relevance of using a multi-model approach involving alternative conceptualizations.

In their paper, **Baxter *et al.* (2018)** also consider observations from the BRIE, which are explained using an integrated model that is able to describe the saturation of bentonite emplaced in a heterogeneous fractured rock. The approach taken in this study provides a framework to understand the key processes in both the rock and bentonite. The predictive capability of these models is investigated within the context of uncertainties in the data and the consequence of predictions of the wetting of emplaced bentonite. A consequence of these findings is that the characterization of the fractured rock local to the bentonite is critical to understanding the subsequent wetting profiles. In particular, prediction of the time taken to achieve full saturation of bentonite using a simplified equivalent homogeneous description of the fractured host rock will tend to be too short.

Uyama *et al.* (2019) present work from the Japanese programme; geological disposal is the most realistic option for high-level radioactive waste in this country. In considering long-term stability for geological disposal, several types of materials were studied as engineered barriers with a host rock. This study focused on metal and bentonite as engineered barrier materials, and investigated the long-term corrosion tendency of the metal exposed to bentonite. An electrochemical method for inducing accelerated corrosion was studied in a laboratory, and some field samples from an FEBEX dismantling project (FEBEX-DP) in Switzerland were analysed for comparison.

General strategy for safety and design

As part of a national programme to develop a repository, it is necessary to undertake safety assessments covering, for example, the construction of a repository, its operation and related waste emplacement, and its evolution following closure in the very long-term future (typically to 1 myr; the timescale may vary according to national regulations).

A safety concept and a safety demonstration concept for the disposal of high-level radioactive waste in German clay formations have been developed and are discussed in **Lommerzheim *et al.* (2018)**. The main safety objective is to retain the radionuclides inside a 'Containment-Providing Rock Zone'. Thus, the radionuclide transport should be restrained by adequate safety functions of the geological and geotechnical barriers. The compliance with legal dose constraints has to be demonstrated for probable evolutions and less probable evolutions. Two catalogues of FEPs (features, events and processes) have been developed as a basis for system analysis, generic geological reference models, disposal concepts and repository designs for northern and southern Germany, relevant for future system evolution.

Monitoring

After components of a repository EBS have been emplaced, but before the facility is closed and sealed, it may be desirable to investigate the evolution of the components (e.g. the rate of resaturation, or swelling pressure evolution).

Geophysical electrical resistivity tomography (ERT) is a promising measurement technique for non-intrusive monitoring of an EBS during the operational phase of the geological disposal of high-level radioactive waste. Electrical resistivity is sensitive to water content and temperature, which are the key variables characterizing the response of the EBS. In order to assess the technology readiness level of the ERT technique for EBS operational monitoring, a field demonstrator has been developed at the URL in Tournemire, France, within the European Commission project 'Modern 2020' (http://www.modern2020.eu/). The paper by **Lopes *et al.* (2018)** reports preliminary ERT surveys that were carried out in January and November 2017 to establish the background resistivity of the experimental area and assess the quality of electrode installation and survey protocols

Sealing systems

Once waste has been emplaced in a repository, it will be backfilled, closed and sealed. Clay materials could be used as part of the sealing processes.

Dixon *et al.* (2018) report work from the Enhanced Sealing Project (ESP), monitoring the thermal–hydraulic–mechanical (THM) responses of a full-scale shaft seal at Canadian Nuclear Laboratories (CNL) former URL site at Whiteshell. The evolution of such a full-scale construction has direct relevance to closure design of repository shafts (and tunnels) in a range of host rock types.

Intended to demonstrate an ability to install a sealing structure that can limit the movement of water between the two hydrological regimes, the main shaft seal consists of a 40% bentonite clay:60% fine aggregate by dry mass component (*c.* 6 m thickness) sandwiched between two 3 m-thick concrete segments in the approximately 5 m-diameter main shaft. The clay and concrete components provide the primary hydraulic sealing and mechanical constraint to the sealing material, respectively. This paper presents the monitoring results of the ESP from its installation in 2009 through to mid-2017.

As national programmes for radioactive waste management progress towards their respective licence applications for repository implementation, associated national and international research programmes are continuing their investigations into clays and clayey materials. This will allow uncertainties relating to, for example, the behaviour of an evolving repository system to be reduced, will increase safety margins and will help with disposal system optimization. As such, geological repository research is a challenging and engaging task for current scientists, and hopefully will provide an interesting and long-term career path that is attractive to young scientists.

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