This paper appears out of order-It was presented in the HLW-II-Poster Session

STATUS OF THE INTERNATIONAL ALLIGATOR RIVERS ANALOGUE PROJECT

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ABSTRACT

The International Alligator Rivers Analogue Project (ARAP) commenced on September 1, 1987 under the sponsorship of the OECD Nuclear Energy Agency. Six organizations from five NEA Member countries are participating in a three-year program with a budget of A$2.6 million (US $2 million) in 1987 dollars. The main objective is to contribute to the production of reliable and realistic models of radionuclide migration within geological environments relevant to the assessment of the safety of radioactive waste repositories. A detailed technical program was approved by a Joint Technical Committee and this comprises six main sub-projects: modelling of radionuclide migration, hydrogeology at the Koongarra uranium deposit, uranium/thorium series disequilibria studies, colloid and groundwater studies, fission product studies and transuranic nuclide studies. Collaborative research, service work and consulting contracts are being carried out by organizations in Australia, Japan, the UK and the USA. Modelling workshops are held regularly and experimental data are being provided as a test case within the INTRAVAL international model validation study organized by the Swedish Nuclear Power Inspectorate.

INTRODUCTION

The nuclear industry worldwide is planning to dispose of high level and long-lived radioactive waste by burial underground in stable geological formations. The assessment of the long-term safety of a repository with respect to the release of radionuclides to the biosphere and uptake by the public will depend heavily on mathematical models. These models must include all of the important processes which can occur during the migration of the radionuclides through the geosphere. A substantial data base will be needed as input to the models and there are limitations on the timescale over which experimental data can be obtained from laboratory or field studies. Natural analogues are an important source of long-term data to provide assurance that the most important processes have been included in the models and that they behave as predicted.

Long-lived fission products and transuranium nuclides in radioactive waste are of particular concern in the assessment of the long-term safety of repositories. Some of these radionuclides exist naturally at very low concentrations in and around uranium ore bodies due to the interaction of neutrons with uranium. Methods are available to measure these radionuclides in samples of rocks and natural waters. Uranium deposits can be considered as analogues of radioactive waste repositories and a study of the migration of radionuclides around them can provide valuable information for the assessment of the performance of repositories.

The Australian Nuclear Science and Technology Organization (ANSTO) has extensively evaluated uranium ore bodies in the Alligator Rivers Uranium Province in Australia as analogues of radioactive waste repositories. This work was supported by contracts from the U.S. Nuclear Regulatory Commission from 1981 to 1987 and from the UK Department of the Environment from 1986 to 1987. The results of the work were reviewed at a Symposium on Natural Analogues in Radioactive Waste Disposal in 1987 (1).

The work was extended by setting up an international project entitled the International Alligator Rivers Analogue Project (ARAP) under the sponsorship of the OECD Nuclear Energy Agency (NEA). This three year research program commenced on September 1, 1987 with six participating organizations from five NEA member countries (Australia, Japan, Sweden, UK, USA). The present paper reviews the status of the project as it reaches its halfway stage.

ESTABLISHMENT OF THE INTERNATIONAL PROJECT

The OECD NEA Steering Committee agreed to sponsor ARAP for a three-year program commencing on September 1, 1987. The Agreement was prepared by the NEA and was signed by the following organizations: the Australian Nuclear Science and Technology Organization (ANSTO); the Japan Atomic Energy Research Institute (JAERI); the Power Reactor and Nuclear Fuel Development Corporation of Japan (PNC); the Swedish Nuclear Fuel Inspectorate (SKI); the UK Department of the Environment (UKDOE); and the US Nuclear Regulatory Commission (USNRC). ANSTO is the managing participant and the budget agreed initially for the three-year project was A$2.6 million (US $2 million) in 1987 dollars.

A detailed technical program was approved by a Joint Technical Committee and this comprises six main sub-projects:

I. Modelling of radionuclide migration

II. Hydrogeology of the Koongarra uranium deposit.

III. Uranium/thorium series disequilibria studies.

IV. Groundwater and colloid studies.

V. Fission product studies.

VI. Transuranic nuclide studies.

The overall objectives of the project are to:

i. Contribute to the production of reliable and realistic models for radionuclide migration within geological environments relevant to the assessment of the safety of radioactive waste repositories.

ii. Develop methods of validation of models using a combination of laboratory and field data associated with the Koongarra uranium deposit.

iii. Encourage maximum interaction between modellers and experimentalists in achieving these objectives.

The method of operation is to undertake visits to the Koongarra site to obtain samples of uranium ore, rocks and water from drillholes. The samples are then analyzed by a variety of chemical and physical methods in laboratories of the participants or of contractors with special expertise and equipment. At the start of the project samples of cores from 71 drillholes through the orebody were available. Samples of water were obtained from several of the 40 larger diameter drillholes. All of these drillholes were established during the early exploration phase by Denison Australia Pty Ltd. Initial hydrology studies were carried out by consultants to this company. More recent hydrology studies by contractors to ARAP recommended additional holes should be drilled and these were completed in December 1988.

Modelling workshops are held at regular intervals to enable maximum interaction to take place between modelers and experimentalists and to guide the direction of the experimental program.

The organizations now taking part as contractors in the project in addition to the six primary participants are: University of Arizona, Johns Hopkins University, University of Sydney, Los Alamos National Laboratory, UKAEA Harwell Laboratory and the Department of Mines and Energy, NT, Australia.

DESCRIPTION OF THE KOONGARRA URANIUM DEPOSIT

The Alligator Rivers Uranium Province in the Northern Territory of Australia is located about 200 km east of Darwin. The Koongarra uranium deposit of Denison Australia Pty Ltd which was chosen for the international ARAP project study lies about 25 km south of the small town of Jabiru. The area has a monsoonal climate with almost the entire rainfall occurring in the wet season between November and March.

Uranium mineralization occurs at Koongarra in two distinct but related orebodies separated by about 100 m of barren schists (2). Both orebodies strike and dip broadly parallel to the Koongarra Reverse Fault, which is the footwall to the ore zone. Primary mineralization is largely confined to quartz-chlorite schists immediately above the fault zone. A graphitic quartz-chlorite schist forms a distinctive hanging wall unit. The more south-westerly of the two orebodies (No. 1 orebody) has a strike length of 450 m and persists to a depth of about 100 m. Secondary uranium mineralization is present from the surface down to the base of weathering at about 25 m depth and forms a tongue-like body of ore dispersing downslope for about 80 m to the east.

An extensive study of the distribution of uranium, thorium and radium isotopes in and around the No. 1 orebody has identified well-defined areas of leaching and accumulation within the weathered zone (3). Geochemical data are also available for groundwater samples from several of the drillholes in the vicinity of the orebody. Hydrological data have also been obtained in selected parts of the area. The data accumulated so far has been used as input to models established to calculate the extent and timescale of migration of uranium through the secondary dispersion zone (4). Preliminary analyses of selected long-lived fission products and plutonium have also been obtained with a view to modelling their migration behavior in the orebody.

PROGRESS IN TECHNICAL SUB-PROJECTS

Modelling of Radionuclide Migration

ARAP is giving priority to the modelling of radionuclide migration within the secondary dispersion zone and has presented the available data as one of the natural analogue test cases (case 8) to the international INTRAVAL project initiated by SKI in October 1987. The preliminary modelling work was described in 1986 (4). Modelling workshops were held at Sydney from February 8-12, 1988 and at Tucson from November 21-22, 1988, the latter being held immediately after an INTRAVAL meeting at Tucson.

Five specific modelling tasks are being addressed within ARAP:

a. Determination of the sequence and timing of weathering at Koongarra.

b. Migration of radionuclides in the weathered zone and from the zone where there was originally primary ore.

c. Open system modelling using radiochemical data from crystals of the uranium mineral saleeite to evaluate the timescale of advance of the secondary dispersion zone.

d. Geochemical modelling to identify the controlling reactions for formation of the uranium phosphate zone and uranium silicates from primary pitchblende ore.

e. Hydrogeological modelling in association with sub-project II.

Hydrogeology of the Koongarra Uranium Deposit

The initial groundwater hydrology data were obtained over the exploratory phase of the deposit by consultants to Denison Australia Pty. Ltd. and the data were interpreted by Snelling (3). A series of new aquifer tests were carried out for ARAP by the University of Arizona in May-June 1988 and considerable heterogeneity and anisotropy of the near-surface aquifers was demonstrated. Additional drillholes for more aquifer tests and water analyses were recommended as essential and therefore 22 new holes were drilled during October-December 1988.

Several of these holes were fitted with special water sampling ports at selected depths. A new series of aquifer

tests are planned for June-July 1989 as well as geochemical sampling in April 1989.

Uranium/Thorium Disequilibria

The large amount of early data on uranium/thorium disequilibria in solid rock and water samples was reviewed in 1987 (1). A number of new studies on core samples from initial and new drillholes are underway at ANSTO, Sydney University and JAERI. The main tasks being undertaken at JAERI are to study the alteration mechanism of chlorite, uranium adsorption/desorption with pure chlorite and other clay minerals, and sequential extraction of samples with measurement of uranium, thorium and actinium series nuclide disequilibria in each mineral species. The objectives of the work at ANSTO are to provide a two - dimensional uranium series database in two sections through the ore deposit, and to study disequilibria and total elemental analysis of mineral phases and saleeite crystals, all to assist in the modelling of migration. The Sydney University work will study the relationships between uranium contents in separated mineral phases (particularly clay minerals).

Groundwater and Colloid Studies

This sub-project interacts with sub-project II and will provide extensive geochemical data from water samples obtained at various depths from selected drillholes. Double packer systems are now being used to obtain samples at defined depths. Samples were obtained in special field visits in May and October 1988 (end of wet and end of dry seasons respectively) and are being analyzed for an extensive range of anions and cations, and also for tritium, stable isotopes, chlorine-36, carbon-14 and iodine-129. Further special field sampling programs will be undertaken in May and October 1989, while a limited number of analyses will be made on monthly samples from selected wells to study short term variations.

A number of measurements of the colloid content of groundwater at Koongarra have been made in recent years and results of collaborative work by ANSTO, the University of Sydney and Harwell were reported in 1988 (5). This work, and more recent studies, shows that colloidal transport has been occurring in Koongarra groundwater. Colloids identified include clay particles, and particles containing uranium, lead, titanium and magnesium silicate. Most of the samples have been obtained from water from the weathered zone and further studies are planned to obtain water samples from below this zone.

Fission Product and Transuranium Nuclide Studies

The occurrence of selected long-lived fission products (technetium-99 and iodine-129) and plutonium-239 has been studied in solid and water samples from the Koongarra deposit over the last few years. The work has been carried out in collaboration with the University of Arizona and the Los Alamos National Laboratory. A review of the progress made was presented at an IAEA Research Coordination Meeting at Keyworth, UK, in November, 1988 (6). Methods of extraction of these isotopes from large volumes of groundwater have been developed, as well as methods for their measurement at the very low natural levels. Only a limited number of samples have been analyzed to date, but there is agreement within a factor of 3-4 between measured and predicted ratios of Tc-99/U and Pu-239/U, whereas the predicted I-129/U ratio is about one order of magnitude higher than the measured value. Seven ore samples from two parallel transects through the secondary ore zone are now being studied and additional water samples will be obtained in 1989 from better defined levels in selected drillholes using packer systems.

CONCLUSIONS

The international ARAP has reached the halfway stage in its planned three-year program. Considerable progress has been made in the sub-project of modelling which was given priority, and in obtaining a high level of interaction between modellers and experimentalists. A series of hydrological measurements in the first year revealed the gaps in the previously available hydrogeological data base and a series of new drillholes were completed in late-1988 to enable more detailed hydrogeological data to be obtained. Chemical and geochemical data measurements and associated modelling are proceeding on water samples obtained from well-defined levels using packer systems. Initial data for modelling the migration of radionuclides in the weathered zone have ben provided as a test case for the international INTRAVAL project and additional data will be provided as it becomes available.

The field trips and workshops planned in the second half of the project are expected to contribute significant new data on hydrogeology, geochemistry, radioisotope disequilibria, and fission product and plutonium distributions in this system. These data will be available as input to a series of models to enable a better understanding to be obtained of radionuclide migration mechanisms in natural systems relevant to the performance assessment of geological repositories for radioactive waste.

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ACKNOWLEDGEMENTS

The work in the Alligator Rivers Region was supported by the USNRC from 1981-1987 and by the UKDOE in 1986-87. The international ARAP is supported by ANSTO, JAERI, PNC, SKI, UKDOE and USNRC. The work reviewed in this paper was carried out by the following persons whose contributions are gratefully acknowledged:

Airey, P.L., Calf, G.E., Golian, C., Lowerson, R., Lowson, R.T., Nightingale, T., Payne, T., Roman, D., Short, S. (ANSTO); Snelling, A.A. (Consultant); Dickson, B.L. (CSIRO); Sorentino, C., Watt, G. (Denison); Giacomel, A. (AGE); Pidsley, D., Scott, P., Treloar, R. (NT Govt.); Davey, B., Edghill, R., (Univ Sydney); Davis, S., Norris, J., (Univ. Arizona); Curtis, D., Fabryka-Martin, J., Perrin, R.E., Rokop, D., Baker, M. (Los Alamos N.L.); Sverjenski, D., Garven, G. (Johns Hopkins); Lever, D., Ivanovich, M., Longworth, G. (UKAEA, Harwell); Yanase, N., Murakami, T., Sekine, K., Kasai. A. (JAERI); Ochiai, Y., Nakatsuka, N., Higashi, K. (PNC).