Carlson

Battelle
Pacific Northwest Laboratories
P.O. Box 999
Richland, Washington U.S.A. 99352
Telephone (509) 376-0933
Telex 15-2874

June 17, 1988

DISTRIBUTION

Report on CEC MIRAGE Meeting, Brussels, Belgium, March 24-25, 1988

Attached is Donald E. Clark's informal report on the subject meeting for your information and use. The report does not contain a list of attendees (approximately 75) because it was not made available to the participants at the time of the meeting. A list, to be issued later, will be included in the formal record from the meeting organizers. Because of the large size of the attachments, they are not included here for most of the recipients; however, if you feel you want some or all of them, feel free to contact my office and they will be provided.

Don is a staff member of PNL's International Program Support Office (IPSO) who is located in Braunschweig, Federal Republic of Germany, and who assists all DOE offices in radioactive waste management liaison activities with European countries. If you have need for Don's services, please contact him at (49)531-8012-213, telefacsimile (49)531-8012-200 or contact me at FTS 444-0933.

Very truly yours,

K Schneider

for L.T. Lakey, Manager
International Program Support
Office

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R.C. Baker
Chicago Operations Office
U.S. Department of Energy
9800 S. Cass Avenue
Argonne, IL 60439

S.P. Cowan
Savannah River Operations
U.S. Department of Energy
P.O. Box A
Aiken, SC 29802

S.T. Hinschberger
Idaho Operations Office
U.S. Department of Energy
785 DOE Place
Idaho Falls, ID 83402

W.F. Holcomb *
U.S. Environmental
Protection Agency
401 M Street, SW
Washington, DC 20460

B.G. Kitchen
Savannah River Laboratory
E.I. duPont de Nemours
Aiken, SC 29808-0001

R.A. Robinson
Office of Waste Technology
Development
Battelle Project Management
Division
7000 S. Adams Street
Willowbrook, IL 60521

A.D. Rodgers
EG&G Idaho, Inc.
P.O. Box 1625
Idaho Falls, ID 83415

L.D. Tyler
Sandia National Laboratories
Albuquerque, NM 87185-5800

Albuquerque Operations Office (3)
U.S. Department of Energy
Albuquerque, NM 87115
Attn: W.R. Cooper (WIPP)
A. Hunt
M.H. McFadden

Nevada Operations Office (2)
U.S. Department of Energy
P.O. Box 98518
Las Vegas, NV 89193-8518
Attn: C.P. Gertz *
R. Levitch

T.H. Row, C-36
Oak Ridge National Laboratory
P.O. Box X
Oak Ridge, TN 37831

* *Attachments included*

Westinghouse Electric Corporation
P.O. Box 2078
Carlsbad, NM 88220
Attn: W.C. Moffitt

J.F. Strahl
R.F. Weston, Inc.
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8th Floor
Washington, DC 20024

U.S. Nuclear Regulatory
Commission (3)
Washington, DC 20555
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J.L. McElroy
Y. Onishi
S.C. Slate
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REPORT ON PLENARY MEETING OF THE CEC PROJECT MIRAGE - SECOND PHASE

HELD IN BRUSSELS, BELGIUM
MARCH 24-25, 1988

Reported by

Donald E. Clark, International Representative, Europe

International Program Support Office (IPSO)
Battelle Pacific Northwest Laboratories
Hoeben 38a
D-3301 Gross-Schwuelper
Federal Republic of Germany
Tel: (49)531-8012-213
Telefax: (49)531-8012-200

SUMMARY

D. E. Clark (Battelle Pacific Northwest Laboratories, IPSO) attended the Plenary Meeting of the CEC Project MIRAGE, Second Phase, in Brussels, Belgium, March 24-25, 1988. The attendance at this meeting of approximately 75 persons included CEC staff and representatives of the European Community countries, OECD/Nuclear Energy Agency, Sweden, Finland, Switzerland, Canada, and U.S.A. Summary reviews (ca. 30 minutes each) were given for the four areas below, followed by discussion periods of one hour or so. Highlights are presented in the main body of this report.

1. "Geochemistry of Actinides and Fission Product"
2. "In-Situ Migration Experiments"
3. "Natural Analogues"
4. "Calculational Tools"

The first phase of the MIRAGE Project was carried out during the 1981-85 time period; the second phase began in 1986. A 4-5 year follow-on phase (MIRAGE III) is planned starting in 1990 at about the same level of funding.

The next Plenary Meeting of the CEC MIRAGE Project will probably be held in the Spring of 1989.

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CEC Project MIRAGE - Second Phase - Plenary Meeting, Brussels, 24-25th March 1988, Programme

Attachment 2
Geochemistry of Actinides and Fission Products in Natural Aquifer Systems, by J.I. Kim, Working Document Only, 3/88

Attachment 3
In-Situ Migration Experiments, by A. Avogadro, Working Document Only, 3/88

Attachment 4
Natural Analogues, by Neil A. Chapman, Working Document Only, 3/88

Attachment 5
Calculation Tools, by G. de Marsily, Working Document Only, 3/15/88

Attachment 6
The CHEMVAL Project: Status Report, by D. Read and T.W. Broyd, 3/88

INTRODUCTION

The Commission of the European Communities (CEC) serves as the executive body for the European Communities. The CEC provides part of the support for radioactive waste management R&D conducted by some research centers in the Member States. Thus, certain nuclear waste disposal programs of interest to the Member States may be supported under cost-sharing contracts with the CEC to which the respective parties make approximately equal contributions. The CEC Headquarters is in Brussels, Belgium. Additionally, the CEC operates two Joint Research Centers at which radioactive waste management studies are conducted, one located in Ispra, Italy, and the other in Karlsruhe, Federal Republic of Germany (FRG).

A Plenary Meeting of the CEC-coordinated project MIRAGE (MIgration of RAdionuclides in the GEosphere), Second Phase, was held in Brussels on March 24-25, 1988. In summary fashion, the recent work of about 40 laboratories of the Member States and of the Joint Research Center, Ispra, was reviewed. The Agenda for this meeting is given in Attachment 1. Approximately 75 persons attended the meeting, including CEC staff and representatives of the European Community countries, Nuclear Energy Agency (NEA) of the Organization for Economic Cooperation and Development (OECD), Sweden, Finland, Switzerland, Canada and U.S.A.

Serge Orlowski, Head of the CEC Nuclear Fuel Cycle Division, gave the opening address. He stated that Phase I of the MIRAGE Project (1981-85) was a success. Phase II, started in 1986, has the same overall objective as Phase I, namely, to provide a quantitative description and understanding of the processes whereby radionuclides may be transported in the geosphere (including deep subseabed sediments). At this stage, the emphasis is moving toward onsite testing and comparison of codes, databases, etc. A 4-5 year follow-on phase (MIRAGE III) is planned starting in 1990 at about the same level of funding.

A companion CEC project, PAGIS (Performance Assessment of Geological Isolation Systems), is now winding down. Funding for the PAGIS Project, which was started in 1982, has totaled approximately $10-12 million. Results obtained for the four formations (clay, granite, salt, and subseabed) studied in the PAGIS Project indicate that all are acceptable options if the sites are properly designed and constructed. However, it is important to maintain the geological barrier function over very long periods of time, and the MIRAGE Project is intended to provide quantitative assessments of radionuclide migration in the geosphere from wastes emplaced in geological repositories.

SUMMARY REVIEW DISCUSSIONS

Following the introductory remarks, summary reviews (ca. 30 minutes each) were given in the four research areas noted in Attachment 1, followed by discussion periods of one hour or so. The highlights are presented below.

1. "Geochemistry of Actinides and Fission Products"

   The work in this area was reviewed by Professor J. I. Kim of the Technical University, Munich. His presentation is contained in Attachment 2.

   Main research subjects in this area are:

   - complexation with organics (both natural and man-made)
   - colloid generation in groundwater
   - basic retention mechanisms in aquifer systems

   Initially, focus was on the first two research subjects, resulting in the workers referring to themselves as members of the "COCO Club". Groups from about 30 laboratories are now working in this area.

   Complexation of radionuclides with natural organics and colloid generation in natural aquifer systems are two subjects now receiving much attention in the safety assessment of nuclear waste repositories. Actinides and some fission products in the higher oxidation states have strong tendencies to complex with natural organics (e.g., humic acid) or to form colloids. Such behavior can have profound effect on their potential migration in the geosphere. Of the complexes studied, EDTA (man-made) is the most powerful complexing agent for the actinides; other important complexes in natural systems are carbonate, hydroxide, humate, citrate, sulfate, and chloride ions.

   Work of the COCO group in MIRAGE II has concentrated on the acquisition of basic data on humate complexation and colloid generation in natural aquifer systems. Interlaboratory comparison exercises have been conducted on the characterization of humic acids, complexation of radionuclides with humic acids, and colloid generation. These exercises have been very useful for determining uncertainties in the data from different laboratories and as a means to perfect existing experimental methods. The need to develop and validate new experimental methods is clearly evident from the work discussed by Kim.

   The ultimate objective of the geochemical study of actinides and fission products is to understand retention mechanisms of individual radionuclides in a given aquifer system such that retardation parameters for use in modeling can be validated. Basic retention mechanisms appear to be poorly understood at this stage. Kim discussed future work aimed at improving this understanding, including solubility, sorption and

diffusion studies; speciation studies; review of the role of organics and other complexing agents; and modeling exercises.

A very good job of coordinating all of the separate studies in this research area has been done, as is evident from Kim's presentation. The interlaboratory comparison exercises have been very useful in developing some confidence in the methodologies. However, it seems that there is need for the Project to adopt standardized test procedures to ensure that all of the experimental data can be intercompared. (The European groups generally have taken a less formalized approach to such matters than workers in the U.S.A. nuclear waste repository program).

There is a definite need to develop suitable new methods for characterizing colloids and determining their population and size distributions.

The existence and continuing development of various databases for this research area was discussed. It appears that these databases are somewhat loosely defined and not always controlled. This could cause some problems for U.S.A. workers wanting to make use of the the European results.

C. de Geuldre of the Forshaler Institute, Switzerland, discussed the Colloid Benchmark Exercise done at the Grimsel site. Granitic water was sampled at a depth of about 450 meters using conventional techniques. Concentrated samples maintained under anerobic conditions were sent to various COCO laboratories for characterization. Unfiltered water, ultrafiltered water, and colloids on membrane filters were also characterized by the COCO labs. For particles in the range of 50 to 1,000 nm, fairly uniform results of about 10($\pm$10) particles per liter were obtained. However, this exercise illustrated that large variations (orders of magnitude) are not uncommon when existing methodologies are applied to the characterization of colloids in natural waters.

2. "In-Situ Migration Experiments"

A. Avogadro of Ispra (Italy) discussed recent work in this area (see Attachment 3). The objective of this research area is to quantify radionuclide migration potential under realistic natural conditions. From the presentation and ensuing discussion, it appears that good experimental design is very difficult to achieve and many lessons are yet to be learned. Avogadro discussed three different approaches that have been adopted by the laboratories participating in this Project:

- in-situ migration experiments
- laboratory experiments with simulation of natural geochemical conditions
- column migration experiments in real natural environment making use of special probes placed inside a well

Two types of underground migration experiments are being conducted at the Underground Research Laboratory (URL) in Mol, Belgium:

- percolation experiments with labeled clay cores emplaced in boreholes (tracers used are Eu-152 and E-154; Sr-85; Cs-134; and U-233)

- direct injection of tracers in the clay formation (H-3 tracer)

Due to the extremely low permeability of the geologic medium (Boom clay), diffusion appears to be the dominant transport mechanism, even under very unrealistically high pressure heads. The results appear to verify laboratory measurements and predicted behavior in the rock formation.

More complex diffusion measurements at the Mol URL are planned by the Harwell Research Establishment in the United Kingdom (U.K.). Comparative laboratory tests will be performed using standard methods in an attempt to correlate predictions based on laboratory test results with those obtained in the field. A second Harwell experiment will be performed at a U.K. clay site. The tests will take into account the heterogeneity of the formation, comparing flows into silts, sand and fractures and into the clay. In-situ permeability measurements of the sand and silt layers are planned.

The British Geological Survey (BGS) plans to conduct an in-situ field tracer test in a shallow glacial sand aquifer confined between two clay layers. The test is intended to verify predictions of radionuclide mobility taking into account the potential effect of organics (natural or introduced) on radionuclide behavior. The final phase of the project will involve computer analysis of the breakthrough curves to model quantitatively the relationships between organics and migration rates.

Laboratory simulation experiments are being conducted at the CEC Ispra Laboratory. Column tests using porous materials from the clay aquifer in Mol and quartz sand taken over the salt dome of Gorleben (FRG) are being made in gloveboxes with controlled synthetic groundwaters. Leach rates for Tc-99, Np-237, and Pu-238 from borosilicate glass and retardation factors in the porous media have been determined. This experimental approach appears to be quite versatile and can provide important data on the migration behavior of individual radionuclides.

Finally, the use of geochemical probes placed inside wells was described. This work is being done at Cadarache in France. The concept of using these probes for measurements in boreholes is rather interesting. However, the project sounded very complicated and it seems unlikely that these systems will perform as well as intended.

It is clear that development of appropriate experimental methods must be a high priority for future work in this research area. Since tests of this type are very expensive, they should be especially well planned and designed to optimize their end product(s). In-situ tests are very important (one can't always reproduce the field conditions in the

laboratory, for example), and most especially for the purpose of validating laboratory results.

3. "Natural Analogues"

Recent research work in this area was reviewed by Neil A. Chapman of the British Geological Survey (see Attachment 4). There have been lots of developments since the last MIRAGE Plenary Meeting in 1985 at which time there were four natural analogue projects. In response to increased interest in the subject, the Natural Analogue Working Group was formed in November 1985. This group is intended to provide an important link between the experimenters and the modelers. So far, it appears that the group has been effective in this regard. The growth of this area has resulted in a major CEC symposium in 1987 and the preparation of an IAEA Technical Report explaining the application of natural analogues to performance assessment methodology. Another CEC/DOE meeting on natural analogues is scheduled for June 1988 in the U.S.A.

There are now nine cost-sharing analogue studies in the MIRAGE Project, all of which are concerned with radionuclide transport and retardation processes, largely at low (ambient) temperatures. Five investigations concern diffusion and advective processes in sediments (mainly in clays), while the other four address matrix diffusion or elevated temperature elemental migration in granitic rocks. Several of the studies are just starting and consequently had little progress to report.

Chapman's presentation was decidedly upbeat. He feels that solid progress has been made over the past year or so, and as the area matures, it can be expected to make an important contribution both to our understanding of radionuclide migration in the geosphere and to the public acceptance of radioactive waste disposal. For these reasons, even though some of the results may be overstated and workers in the field are optimistic concerning the application of their results to actual licensing issues, it is worthwhile that continued effort be directed to natural analogue studies and that different countries will share in the benefits of such work through international exchanges.

Two contracts involve the investigation of large-scale fluid flow in clay formations in central Italy. In one study, data are being obtained on the abundance and distribution of radiogenic gases with crustal sources (helium and radon) in soils, groundwaters and boreholes to assess which zones of the thick clay sequences are most permeable, and to define 'leaky' areas where fluids are surfacing from great depths. This study aims to produce maps of the clay basins showing concentration contours for these radiogenic gases, related to structures and the locations of processes likely to be involved in their origin. Supporting information is being obtained through observations of permeability and fracturing. So far, this interesting study seems to show that certain parts of the fracture system are sealed and some are open. The question is how these measurements might be correlated with possible groundwater movements.

A second contract involves a study of the same clays related in particular to the way in which they affect bulk permeability and rock and pore-water chemistry. The work, which is just starting, will consist of a regional study of the structural and morphological development of the basin, along with a detailed geochemical study of the evolution of the clay basin. Data from this study should be useful for predicting the likely long-term movement of fluids from a repository in clay.

Another research area directed at smaller scale elemental migration in sediments is supported by three contracts. Two of the contracts are closely linked in a collaborative study of terrestrial examples of elemental diffusion/advection/retention in sediments in the U.K. Surface diffusion processes are being studied at two sites, Loch Lomond and Lundin Castle in Scotland, with attention to the distribution of iodine, bromine, and radon in sediments, marls, and clay layers. The British are also participating in collaborative modeling work with the French on radioelement distributions in sediments from the Needle's Eye site in Scotland (uranium series elements and carbon-14). The main value of this work seems to be the productive interaction that has been established between the modelers and the experimentalists, and illumination of the practical limitations of analogues when exact quantitative data on radionuclide migration rates are sought: the primary power of the approach is in revealing the critical processes at work in the natural system.

The third contract is for studies of uranium and decay chain radionuclides in core samples of seabed sediments from the Madeira Abyssal Plain. These organic-rich sediments are known to contain bands which are enriched in these radionuclides and have remained physically undisturbed for up to 750,000 years. The determination of radionuclide concentration profiles, correlated with geotechnical properties of the cores (e.g., porosity, etc.), will presumably facilitate realistic modeling of transport processes in these sediments.

The other anologue studies are concerned with transport processes in hard, fractured rocks. Two French studies are underway on elevated temperature migration in granitic rocks. Both are examining the geochemical transport processes which are responsible for the movement of elements in the late stages of granite formation when the rock is solidified and fractured, but large volumes of warm aqueous fluids are still moving through it. The work includes determining the distribution of radionuclides (natural decay chains) along with geochemical and mineralogical characteristics of the rocks. Matrix-diffusion processes in granitic rocks are being studied under two contracts by groups in the U.K. and Spain. One study is concerned with ionic diffusion (halologen anions and sulfate) from seawater into large quarried blocks of granite submerged beneath the sea for about 30 years. The other study concerns the nature of intragranular porosity adjacent to fractures in different types of granite, and concentrates on natural decay chain disequilibria resulting from preferential migration processes. The contribution of these studies will be primarily in contributing to a basic understanding

of the complex processes and natural structures affecting radionuclide migration, and in validating the experimental and modeling approaches.

A conclusion of these activities is that natural analogues can serve an important function for the validation of computer codes, but interpretation of the results of natural analogue studies need to be carefully considered. In particular, one must always deal with enormous uncertainties with regard to the history of natural analogue formations and, of course, with their considerable heterogeneities. The studies also tend to be rather costly.

4. "Calculational Tools"

Professor G. de Marsily from France reviewed the work being done in this research area. He stated that continuing progress is being made in the development and application of calculational tools for modeling radionuclide migration phenomena. Cooperation between countries has likewise increased, but additional efforts may still be needed. Codes are being exchanged by various groups and made available through computer networks. Furthermore, intercomparison and validation studies have been made that are of considerable benefit to modelers and also for improving the credibility of work in the MIRAGE Project. The current emphasis is on geochemical modeling, assessing the role of heterogeneities (e.g., fracture systems), and of irreversible thermodynamic processes (e.g., case of thermal gradients).

A detailed review of recent work done in this research area is given in Attachment 5. As in the U.S.A., there has been a considerable maturation of the modeling field as applied to nuclear waste disposal. Many of the codes used or developed in the U.S.A. program are likewise being used by the Europeans. Code validation is receiving much attention, as represented by the CHEMVAL Project (validation of chemical speciation/transport codes) which involves active participation of 14 research organizations within the CEC, Finland, Sweden, and Switzerland. As a final item on the program, the status of the CHEMVAL Project was presented by D. Read and T. W. Broyd of the U.K. (see Attachment 6). This has been an important undertaking, and there has been some very good work done here.

One concern that arises on hearing the modeling presentation is the question as to how all of the code work is integrated. Enough code development work has been done. The current emphasis should be (and is) on validation.

CONCLUDING COMMENTS

This was a good meeting for getting an overview of the MIRAGE Project activities. Most of the attendees were technical representatives of the various research areas, and the discussions were mostly of a scientific nature. There was really no critical assessment of the work in terms of its relative importance for sitingand licensing a nuclear waste repository, etc. Certainly, the participants were enthusiastic over the work in their

respective areas, and generally are proponents for continued efforts in this regard. The CEC serves as a source of matched funding for these projects. CEC staff do a very good job of bringing the projects together, assuring that coordination is maintained, facilitating information exchanges, and publishing reports, etc. However, much of the critical reviewing function, including an assessment of the relevance of an individual project to the nuclear waste disposal program of a given country, appears to be done in the home institutions and countries.

As per normal procedure, the CEC will publish a report on this meeting. The last meeting, which was held in 1985, was reported in CEC publication EUR 10023 EN.

According to an announcement made at this meeting, the next Plenary Meeting of the CEC MIRAGE Project will probably be held in the Spring of 1989.