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LAWRENCE LIVERMORE LABORATORY ENERGY AND TECHNOLOGY REVIEW

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August 1975

Briefs

The short items on this page announce recent developments of interest. Some of these items may be amplified in future issues; none of this material is reported elsewhere in this issue. Summaries of this month's articles appear on the opposite page.

NEW HARD STRUCTURE MUNITION SUCCESSFULLY TESTED

Early in August, we tested a new HE hard structure munition (HSM) whose warhead - an LLL design - is specifically intended to defeat such massive reinforced-concrete structures as bridge piers, hydroelectric dams, submarine pens, underground control centers, and hardened (specially protected) aircraft shelters. These structures have proved almost impregnable to conventional bombs and missiles, as demonstrated by the repeated, unsuccessful attempts during the Viet Nam conflict to destroy the Than Hou bridge.

The test, performed at Eglin AFB, Florida, was successful. The target was a huge reinforced-concrete monolith weighing 1000 tonnes (2.2 million pounds). The munition was mounted on a rocket sled propelled by three booster rockets. The detonation obliterated the target center and tore the remaining concrete into four major blocks, which were severely fractured and displaced.

The warhead, developed from an Air Force concept, is a dual-stage design containing a shaped charge to prepare the reinforced concrete for penetration of the main charge. On impact, the shaped charge fires immediately, but the main-charge firing signal is delayed. This allows the main charge case to penetrate deeply into the target.

Early tests have indicated that the munition is also effective against targets with soil or sand overburden. These tests are continuing.

It now appears that the HSM warhead may be the first of a new generation in which the damage potential is more than tenfold greater than conventional HE warheads. Other applications are being considered.

DEUTERIUM ENRICHMENT ACHIEVED WITH LASERS

For the past year, we have been investigating improved routes to heavy-water production. Heavy-water reactors offer advantages when compared to the light-water reactors now prevalent in the U.S., but the

high cost and insufficient supply of heavy water severely limit their use. Their advantages are twofold: 1) they can use natural rather than enriched uranium as fuel, and 2) they can be operated as efficient slow-breeder reactors, converting the abundant element thorium into fissionable uranium-233, a suitable alternate nuclear fuel. However, production costs for heavy water (D_2O) run about \$120/kg; a large reactor requires several hundred tonnes.

To improve heavy-water production, we are now experimenting with lasers to photodissociate formaldehyde into deuterium-enriched hydrogen gas. Early research suggested that if formaldehyde were irradiated at the right wavelength in the near-ultraviolet region, HD gas should be preferentially produced as a dissociation product; recent work has verified this concept. We have demonstrated highly selective isotope separation of deuterium in laser-irradiated mixtures of H_2CO and HD_2CO . Using an HD_2CO absorption line coincident with a helium-cadmium laser at 325.03 nm, we have achieved single-step, fifteenfold enrichment in the deuterium content of hydrogen gas. The initial deuterium concentration was 0.015 mole % (natural abundance).

Our next step is to verify that extremely high enrichments are possible by tuning the laser to a better wavelength. Deuterium is rare, and its initial enrichment is the most costly. For commercial feasibility, the ultraviolet photons in our process must be used more efficiently: enrichment by a factor of 300 or more is needed to ensure that absorption occurs only in molecules containing deuterium. We are now investigating wavelengths in the ultra violet absorption spectrum of HD_2CO that promise to provide this enrichment. An ultraviolet dye laser supplies the necessary tunable and monochromatic light.

If we can achieve the needed deuterium enrichment, the last step to commercial feasibility will be to develop lasers that are at least 1% efficient. (The efficiency of our helium-cadmium laser was only about 0.001%.) Several outside research groups are progressing on this problem, and two lasers - xenon-fluoride and krypton-fluoride - may prove suitable for our photodissociation process.

Contents

ADVANCED ENERGY SYSTEMS

Geothermal Energy Development Project: Status Report	1
--	---

At LLL, we are developing techniques and equipment for recovering geothermal energy from subterranean hot water deposits by the total flow process (using the total wellhead product). A turbine is being built. We have partially characterized a converging-diverging nozzle to expand the wellhead fluid; our data indicate nozzle velocity coefficients that exceed 0.9. Other laboratory and field studies are in progress.

ENVIRONMENT AND SAFETY

Livermore Regional Air Quality (LIRAQ) Models: Tracking Pollutant Concentrations	9
---	---

We have developed two regional air quality computer simulations for pollution control studies. Using local topography, meteorology, and pollutant-source inventory as input data, these simulations, called Livermore Regional Air Quality (LIRAQ) models, can calculate the time and space variation of pollutant concentrations in each of many fixed-grid cells. The Bay Area Air Pollution Control District will soon implement the models for various studies. For the EPA, we are adapting the models for use in and around St. Louis, Missouri.

ARAC: Real-Time Prediction of Hazards Due to Radioactive Releases	12
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The Atmospheric Release Advisory Capability (ARAC) is an ERDA-sponsored service designed to provide nuclear facilities with real-time predictions of the health hazards that might result from a release of radionuclides or other toxic materials. A two-laboratory feasibility study has proved the concept. LLL has built and is now operating a prototype ARAC component. ERDA plans to implement ARAC service at six of its nuclear facilities during the next three years.

NATIONAL SECURITY

X-Ray-Fluorescence Analysis for Nuclear Materials Accountability	17
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We are developing an x-ray-fluorescence analysis system for determining uranium and plutonium concentrations in the solution obtained when the fuel rods from light-water power reactors are dissolved in nitric acid. These concentrations are needed to inventory the uranium and plutonium handled in fuel reprocessing plants. Our analytical technique should make it possible to analyze dissolver solution with speed and a 0.5% accuracy.

NOTES AND REFERENCES	19
--------------------------------	----

Field work for the LLL geothermal energy development project is under way at Sinclair #4 well in the Salton Sea geothermal area, using facilities of a consortium comprised of Southern California Edison, Phillips Petroleum, and the Southern Pacific Land Company. The wellhead product, as shown above, is released into a large concentrating pond. Our studies include calorimetric measurements, brine analyses, scale morphology, materials testing, and nozzle characterization. Despite vast geothermal resources, the area presents the severest recovery problems to be found: flows of extreme salinity at very high temperatures and pressures. LLL is developing the technologies to harness these flows (see article beginning on the opposite page).

ADVANCED ENERGY SYSTEMS

GEOHERMAL ENERGY DEVELOPMENT PROJECT: STATUS REPORT

At LLL, we are developing techniques and equipment for recovering geothermal energy from subterranean hot water deposits by the total flow process. Utilization of hot water deposits is the most immediately feasible plan for exploiting U.S. geothermal resources. The total flow process involves expanding the entire wellhead product—brines plus steam—directly through a mixed-phase expander to drive an electric generator. Because this process uses the total energy content of the wellhead product, it is thermodynamically simple and inherently the most efficient.

Our program entails research and development in earth sciences, brine chemistry and handling, materials technology, and energy conversion. Laboratory and field studies are under way. The final goal is to combine these research results into a prototype power plant to be built in the early 1980's.

This article describes our development project and reports some recent engineering accomplishments: fabrication of a total flow turbine and partial characterization of a converging-diverging nozzle to expand the wellhead fluid. Our data indicate nozzle velocity coefficients that exceed 0.9.

Within the next decade, geothermal energy can make an important contribution toward reducing U.S. dependence on oil and gas for electric power generation.¹ ERDA is sponsoring several important research projects, including one at LLL to recover energy from hot water (hot brine) deposits by the total flow process (described below). Superheated dry steam deposits such as the Geysers, California, are easier to exploit but much less abundant than hot water deposits. The extent of exploitable hot dry rock resources (another form of geothermal energy) has not been determined nor has a recovery technology yet been demonstrated.

Thus U.S. geothermal energy development in the next decade will emphasize the exploitation of hot

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Fig. 1. Three methods for recovering energy from hot brine deposits. In the flashed steam method, the wellhead product is throttled to increase the vapor fraction; the liquid fraction is discarded. In the binary cycle method, part of the brine energy is used to vaporize a secondary working fluid; heat is lost in the exchange. The total flow method utilizes the energy of the entire wellhead product.

Fig. 2. Geothermal areas of the western U.S. (left) include the Salton Trough, a low-lying region that encompasses the Mexicali Valley (Cerro Prieto geothermal area) and the Imperial Valley (Salton Sea geothermal area). Both valleys have been extensively explored. Our field studies are concentrated at the Salton Sea.

water deposits. These are characteristically hot brines at reservoir temperatures of 150°C to more than 300°C, whose dissolved-solids contents range from about 0.1 to over 25 wt%. These brines pose several recovery problems, including precipitation, scale formation, corrosion, and erosion.

For recovering and converting the energy from hot water deposits, there are three candidate processes: flashed steam, binary cycle, and total flow. These are shown schematically in Fig. 1. The flashed steam method—a proved technology—requires separation of the vapor fraction from the liquid fraction, which is discarded; the vapor operates a conventional steam turbine. Overall plant thermal efficiencies are generally below 10%. The binary cycle method basically transfers part of the brine enthalpy to vaporize an organic secondary working fluid, which then drives a turbogenerator. Necessarily, only part of the brine's energy content can be transferred. The thermal efficiency of this system, too, is likely to be less than 10%. This method must be considered experimental because of its many unexplored variations.

The total flow process, while not yet demonstrated, is considered the most promising.² In one version of this process, the total wellhead product (brines plus steam) is expanded through a mixed-phase expander that converts its thermal energy to kinetic energy in the form of high-velocity fluid jets. The jets then drive a specially designed turbine wheel that operates an electric generator. Thermodynamically, the total flow process, utilizing two-phase flow (liquid and vapor), is the simplest of the three methods and has the potential for producing up to 60% more power. Total flow will always be more efficient in principle than any method requiring that part of the available energy be discarded. This advantage holds over the entire expected temperature range of hot water deposits.

We are now developing techniques and equipment for this process. Field work is under way at the Salton Sea geothermal area (see Fig. 2), where we are using a well and other facilities operated by a consortium comprised of Southern California Edison, Phillips Petroleum, and the Southern Pacific Land Company. To develop the conversion equipment needed for utilizing the two-phase flow, we have also installed a test facility at Livermore. Our overall project includes research and development in the following:

- Earth sciences, to characterize the reservoir and injection systems.
- Brine chemistry and handling, to resolve problems of precipitation, scale control, corrosion, and erosion.

- Materials technology, to develop corrosion- and erosion-resistant materials.

- Energy conversion, to develop machinery for converting thermal energy to electric energy.

Studies are under way concurrently in the field and at the test facility. Based on field characterizations, we have simulated the wellhead thermodynamic conditions in the test facility to develop and test conversion machinery and instrumentation. Brine chemistry studies are in progress to characterize the constituents that cause scaling. A typical example is shown in Fig. 3. Turbine development is proceeding (see Fig. 4) in preparation for laboratory and field testing.

The ultimate goal is to establish the feasibility of the total flow process in time to stimulate its commercial development in the 1980's. This will require developing and operating a full-scale prototype power plant of about 10-MW(e) capacity by 1980.

Brine Characterization Studies

We are emphasizing development of the hot brines of the Salton Trough, a geothermal region that extends from the Gulf of California northward into California's Coachella Valley (see Fig. 2). Estimates of the recoverable electrical energy from this trough range from about 60 000 to more than 10 million megawatt-years. (California presently consumes 20 000 megawatt-years of electrical energy annually.) The proximity of the trough to large population centers now largely dependent on oil and gas enhances its potential value.

Brines of more than 300°C are known to be located less than 1500 m beneath the surface of the Salton Sea geothermal area. At the wellhead of a 1500-m well, we expect to encounter temperatures of 215 to 220°C, pressures of 2.1 to 2.4 MPa (300 to 350 psia), and vapor fractions of about 20%. In some zones, the brines contain up to 25 wt% dissolved solids. The important point is that these are the worst recovery conditions to be encountered anywhere; the technology that solves these problems will harness any geothermal flows.

Our studies include calorimetric measurements (to determine the total brine enthalpy), brine analyses, and scale morphology. If we can fully characterize the flow, we may be able to devise techniques to prevent scale buildup, which is a crucial problem. To assist in these studies, we are presently developing a computer simulation of brine equilibria. These activities and

Fig. 3. Scale formation from brines flowing through a nozzle tested at Sinclair #4 well. The duration of the run was approximately 50 hours.

some preliminary results have been described elsewhere.³

Field work is now proceeding at Sinclair #4 well. Efforts are also under way to obtain an additional nearby site that can accommodate the ultimate construction of a 10-MW(e) experimental plant. Also, plans have been started to use a high-temperature, low-salinity well for carrying out the first field test of a total flow turbine. To support this work, we are designing a mobile test unit with instrumentation and apparatus to characterize the wellhead product. These activities are timed to permit field testing early next fiscal year if funds are made available.

The results from all these studies are being applied to our materials technology effort, which is supported also by field testing of candidate materials for the turbines, nozzles, and other system components. These materials studies are proceeding concurrently.

Geothermal Test Facility

We have developed this facility to evaluate the mechanical performance of turbine designs, obtain

Fig. 4. Blades for our prototype turbine were fashioned on the ILL numerically controlled, five-axis milling machine. This work required several hundred hours of machining. The design output at 5000 rpm, from a single nozzle, is about 50 kW. The turbine material — an aluminum alloy — is not erosion-resistant, but the blades will be nickel-plated to resist erosion for the duration of laboratory and field testing.

Fig. 5. Test stations at the geothermal test facility. All equipment for producing the working fluid (clear water) is located outside the building.

design information for turbine components, and investigate the mechanics of the two-phase flow. At issue here are two very important concerns: droplet size and accurate determination of the constituents and phase velocities in the two-phase flow. The droplet size is crucial to the turbine-wheel design.

The test facility³ is a closed-loop system that simulates the thermodynamic fluid properties: pressure, temperature, and vapor fraction of geothermal wells. It includes a hot-water generator, control valve, condenser, circulation pumps, and test stations through which the fluid circulates. Figure 5 shows the test stations. We use the facility for testing nozzles, turbine blades, diagnostic equipment and techniques, and conversion machinery.

The facility also has some materials-performance test capability but only for erosion by water droplets,

because it uses clean water. Consequently, the effects of corrosion, erosion by solid particles, and scaling will be studied in the field. Laboratory experiments will be used for preliminary screening of candidate materials, however, prior to the more difficult field testing.

Turbine Development

To extract the maximum available energy from the wellhead product, the total flow process must expand it, generally speaking, to as low a backpressure as possible. We have studied various turbines for this application: tangential- and axial-flow impulse machines, positive displacement machines, and impulse/reaction machines. Our conclusion is that the axial-flow, pure-impulse turbine is best suited to the total flow process—its construction is simpler,

Fig. 6. Simplified view of an axial-flow impulse turbine. Expansion of the two-phase wellhead product through a converging-diverging nozzle converts its thermal energy at high wellhead pressures (A) to kinetic energy in the form of high-velocity fluid jets at exhaust pressures (B).

thereby facilitating scale control, and its power output is higher per given size and width of turbine disk.

As shown in Fig. 4, we are now building our first experimental turbine. The wheel diameter is about 1 m, and it has a vertical axis of rotation with downward flow (see Fig. 6). It uses a single nozzle. The anticipated output is about 50 kW at 6000 rpm.

After laboratory testing, we plan first to field-test this unit at a power output approaching 1 MW if a source of relatively clean geothermal fluid is available. Subsequent work will emphasize development of a brine-tolerant system.

Nozzle Studies

The nozzle velocity coefficient is critical because its square is the nozzle efficiency for converting thermal energy to kinetic energy. The total flow process requires nozzle velocity coefficients greater than 0.9 to produce a turbine engine efficiency of 70%. Nozzle velocity coefficients can be maximized for only a small range of exhaust pressures for any given inlet condition. To develop the best nozzle design, we have started tests in our Livermore test facility.

From tests of simple axisymmetric, conical, converging-diverging nozzles like those shown in Fig. 6, we have obtained both static pressure profiles

and nozzle velocity coefficients. The static pressure profiles give us a picture of the pressure changes of the two-phase flow through the nozzle. We find the profiles to be determined by the inlet enthalpy.

Nozzle exit conditions vary with exhaust pressure. As shown in Fig. 7, there is an optimum exhaust pressure. When this is exceeded, the flow is overexpanded, and a shock wave forms just inside the

Fig. 7. Nozzle exit jet-flow patterns. Overexpansion (a) produces a shock wave just inside the nozzle exit. Near optimum flow (b), there is very little exit disturbance. Underexpansion (c) results in expansion at the nozzle exit.

nozzle exit. The result is jet turbulence and inefficient operation. When the exhaust pressure is less than optimum, the flow is underexpanded. Nozzle efficiency is again reduced because not all the available thermal energy is converted into kinetic energy.

We have obtained some very encouraging nozzle velocity coefficients. The nozzles evidently operate more efficiently with a higher inlet pressure (for example, 2.4 to 2.7 MPa as compared with 1.7 to 2.0 MPa), but in both cases our data indicate coefficients in the range 0.92 to 0.94. We expect that design refinements will produce even higher efficiencies.

We are now testing rectangular nozzles instead of conical nozzles because they are more compatible with the turbine geometry. The velocity coefficients are expected to be much the same.

Project Evaluation

The goal of the LLL geothermal project is to establish the technical feasibility of the total flow process in time to stimulate commercialization in the 1980's. To do so requires not only that the energy-conversion machinery be developed but also that the brines be characterized, precipitation and scale controlled, and means devised to manage and dispose of the brines. Corrosion-and erosion-resistant materials must be developed. These are all integral within the total systems approach we are pursuing.

The final, and perhaps most important, step will be to synthesize this knowledge and experience into manufacturing processes suitable for introduction into the commercial sector. Toward this end we are

maintaining liaison (and some joint studies) with manufacturers whose equipment might be appropriate for geothermal environments.³ We are also keeping in touch with other agencies interested in geothermal development.

Considerable progress has been made with promising conversion machinery. We are building an experimental turbine for testing and have proved, by experiment, that nozzle coefficients better than 0.9 can be realized. Brine-chemistry and materials-performance studies are in progress. This work will be reported in subsequent issues.

We have focused on the total flow process because it is inherently the most efficient conversion method. In principle, the flashed steam and binary cycle methods could be made to approach (but not exceed) the efficiency of the total flow process. However, we believe that the total flow method offers the greatest potential for achieving this efficiency with a reasonable capital investment. In addition, because of its inherent mechanical and thermodynamic simplicity, the total flow process provides greater opportunities for developing scale and corrosion control methods. This is especially important because we estimate that more than half of the energy stored in the hot water deposits of the Salton Trough region is contained in high-temperature, high-saline brines.⁵

Key Words: Salton Sea geothermal area; geothermal energy; geothermal energy - test facilities; geothermal brines; binary cycle process; flashed steam process; total flow process; conversion machinery; brine chemistry; materials technology; impulse turbines; converging-diverging nozzles.

Fig. 8. The Bay Area Air Pollution Control District was created by the California State Legislature in 1955 as the state's first regional agency to deal with air pollution. The District encompasses all of seven counties - Alameda, Contra Costa, Marin, Napa, San Francisco, San Mateo, Santa Clara - and parts of Solano and Sonoma counties. The total land area within its jurisdiction is 14 400 km² with over 4.5 million people and about 3 million motor vehicles. The region is characterized by complex meteorology and topography. For summer and early fall, the period of interest in air pollution studies, an elevated inversion exists over the area with a marine layer underneath. Hills and mountains surrounding the area define gaps and valleys that produce relatively high wind velocities (up to 15 m/s) during certain times of the day. Because of these winds, smog patterns can vary greatly over the Bay Area, increasing the difficulty of modeling pollutant transport.

Fig. 9. Operating scenario for LIRAQ codes. An initial set of file libraries is compiled from data supplied by the user. With the executive routine, the user then specifies the run parameters and file modifications if any. The output is pollutant concentrations for each grid square of the region under study.

ENVIRONMENT AND SAFETY

LIVERMORE REGIONAL AIR QUALITY (LIRAQ) MODELS: TRACKING POLLUTANT CONCENTRATIONS

We have developed two regional air quality computer simulations for pollution control studies. Using local topography, meteorology, and pollutant-source inventory as input data, these simulations, called Livermore Regional Air Quality (LIRAQ) models, can calculate the time and space variation of pollutant concentrations in each of many Eulerian (fixed grid) cells. Four different source classes are provided. The computer results — tables or plots showing the accumulation of pollutants within individual sectors of the region permit the comprehensive analyses needed for assessing the air-quality impact of new towns or shopping centers, regional population growth patterns, or short-term control strategies. There are real-time capabilities for determining, for example, when and where air pollution alerts should be issued.

The San Francisco Bay Area Air Pollution Control District now has access to two state-of-the-art LIRAQ models and a data base for local pollution control studies. This agency will soon implement the models for various studies. For the Environmental Protection Agency, we have agreed to adapt the models for use in and around St. Louis, Missouri.

For the past two and a half years, LLL has been working with the Bay Area Air Pollution Control District (BAAPCD) and the NASA Ames Research Center to develop regional photochemical computer simulations of the San Francisco Bay Area. Supported by the Research Applied to National Needs (RANN) program of the National Science Foundation, the project has resulted in two simulation models: LIRAQ-1, which provides a fine calculational grid (down to 1 by 1 km) without finely detailed photochemical kinetics, and LIRAQ-2, which provides detailed photochemical kinetics but with a coarser grid (5 by 5 km or larger). The difference in grid size is a function of the calculational complexity and the overall size of the region to be studied, as discussed below.

The LIRAQ project has also included the preparation of a data base for the BAAPCD area shown

in Fig. 8. Working with project team members at BAAPCD and NASA Ames, we have created data libraries characterizing Bay Area topography, meteorology, photochemistry, and pollutant-source inventory, as well as measured air quality data for model verification.

LIRAQ Codes

The LIRAQ computer codes are Eulerian multibox air-quality models incorporating both photochemical kinetics and mass-consistent advection and diffusion. Using the local data input, LIRAQ computes the time and spatial variations of ground-level pollutant concentrations and also the average vertical concentrations between ground level and the bottom of the inversion layer. The models are modular: that is, they contain separate modules (submodels) for each of the major calculational steps, such as pollutant transport, chemical kinetics, and the generation of mass-consistent windfields from meteorological and topographical data. Modularization greatly simplifies revisions or adaptation of the models to different regions. For example, the chemical kinetics submodel can be changed or expanded without significantly affecting the transport submodel, and the topography and meteorology of one region can easily be replaced with those of another.

The explicit calculational technique for physical transport in LIRAQ-1 is appropriate for studying nonreactive pollutants (such as carbon monoxide) or relatively simple reaction sets among reactive pollutants. LIRAQ-2, now in final development, is designed for the detailed treatment of complex chemical reaction sets. It uses a modified GEAR package⁶ to solve large sets of coupled ordinary differential equations with a high-order implicit method, and thus is capable of handling reaction sets that involve a wide range of time constants.

LIRAQ's operating scenario is shown in Fig. 9. Each submodel requires certain input data to be supplied by the user or transferred from another submodel or data file. We have designed an executive routine to handle the data inputs and most frequent scenario changes, such as changing the pollutant emission rate in a particular grid square (or set of squares) or

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Fig. 10. Regional modeling is concerned with regions from 100 to 200 km on a side. Depending upon the LIRAQ code used, we can study a region (or subregion) with grids 1, 2, or 5 km on a side. The grid fineness varies with the size of the area studied and the number of pollutant species tracked.

changing the size of the calculational grid. The executive routine is not intended for such major changes as a complete recalculation of the mobile source inventory or a restructuring of the chemical kinetics reaction set.

Based on user-supplied data, an initial set of file libraries for source emissions, meteorology topography, and initial pollutant concentrations is prepared. Each file specifies the appropriate values (e.g., emission rates for the desired pollutants) for each grid square of the region being studied. Typically, files can be provided for 1-, 2-, and 5-km-square grids (see Fig. 10). *Source files contain hourly values for both surface and elevated sources.* Files for meteorology and pollutant concentrations can be supplied for any of the specific days for which field data have been gathered and evaluated. These days are selected to represent typical meteorological occurrences, including well-ventilated (clean air) and extensive atmospheric inversion (stagnant air) conditions.

We have grouped the sources of air pollutants into four classes:

- Mobile sources, including road, highway, and freeway networks.

- Major point sources, such as refineries or industrial plants.
- Population-distributed sources (i.e., residential areas).
- Airports.

This grouping allows us to predict and assess the impact of specific source emissions, an important flexibility because the amount and kind of pollutants vary from class to class. Emission sources are also spatially and temporally variable.

Using the executive routine, the user specifies the run parameters (e.g., the version of LIRAQ to be used and the desired grid size), which library files are to be used, and how, if at all, the files are to be modified. (For an arbitrary subgrid or set of subgrids, the user can set the files to a specified value, multiply them by a coefficient, or add them to a given value.) The executive routine then causes the desired files to be gathered, modified, and put into the selected LIRAQ model. The resulting output is the pollutant concentrations for each grid square, both in printed (or microfiche) form and as plots of concentration isopleths (Fig. 11).

Ultimately, of course, computer limitations force a trade-off between the number of pollutant species that can be treated in a regional calculation and the fineness of the calculational grid—that is, the spatial resolution. The explicit calculational techniques of LIRAQ-1 require only temporary retention of the pollutant concentrations at each grid point for the previous time step, whereas the high-order implicit scheme of LIRAQ-2 requires the concentration history over as many as seven previous time steps. LIRAQ-1 is capable of modeling the San Francisco Bay Area for five pollutant species using a grid as small as 2 by 2 km, the fineness being computer-limited. (For studies of smaller subregions, a 1 by 1 km grid can be used to take advantage of the spatial resolution of the best available pollutant source inventory.) LIRAQ-2, because of its more rigorous chemical kinetics treatment, is limited to grids of 5 by 5 km or larger.

Present Applications

To facilitate access to the models by the Bay Area Pollution District, the LIRAQ codes and associated files have been transferred to CDC 7600 computers at Lawrence Berkeley Laboratory where BAAPCD personnel can run problems from their offices in San Francisco by telephone links. The agency now has access to state-of-the-art regional models, an initial data

base adequate for use with the models, and methodology (including some software) that will enable its personnel to expand the applications as further data on pollutant emissions and meteorology become available. The BAAPCD intends to implement the models this month for various studies. However, the results are not expected to influence its decisions

Fig. 11. Average concentration profiles for ozone (i.e., from ground level to the inversion layer) over the San Francisco Bay Area, expressed in ppm. These isopleths are for a partly smoggy day. The heavy increase in ozone from 6 a.m. to 10 a.m. is due in part to the sun's higher position but more to the Bay Area commute traffic.

for several months, during which time members of the BAAPCD Research and Planning Section will familiarize other agency personnel in the utility of the models. One important application will be land-use planning studies. The capability also exists for real-time (as events occur) applications: for example, to determine when and where smog alerts should be issued.

To date, the air quality models have not been used by other agencies although two Bay Area planning agencies—the Association of Bay Area Governments (ABAG) and the Metropolitan Transportation Commission (MTC)—the California Department of Transportation (CALTRANS), and the EPA have all expressed interest. We have also received inquiries about LIRAQ's availability from local air pollution control officials around the country.

We have received many requests from Bay Area governmental agencies for data developed under this project, particularly for the pollutant-emission source inventory. Information has been provided to CALTRANS and the two Bay Area planning agencies. One intended purpose is to assess the mobile-source emission data and the highway network loadings from which they were generated.

Our interagency agreement with the EPA, to adapt our models to the St. Louis area, is for that city's Regional Air Pollution Study. Under this agreement, St. Louis topographical, emission inventory, and meteorological data will be generated. LIRAQ will be accordingly validated; and proper arrangements, procedures, and user guides will be developed for EPA personnel to operate the model(s) at Lawrence Berkeley Laboratory.

Future Possibilities

Development of the LIRAQ codes and data libraries is part of LLL's atmospheric and stratospheric modeling research. The work also includes the Climatic Impact Assessment Program (CIAP) on aircraft operating in the stratosphere and the Atmospheric Release Advisory Capability (ARAC) project for ERDA nuclear facilities. Much of the underlying research is interrelated. For example, the LIRAQ submodel for generating mass-consistent windfields is also used in ARAC, whose purpose is the real-time prediction of health hazards that might result from an atmospheric release of radionuclides or other toxic materials (see following article). The LIRAQ photochemistry submodel benefitted significantly from work already done in the CIAP study.

In the future, we hope to make our LIRAQ models available to more regional pollution control districts and are already considering inquiries into this possibility. Also, depending on funding, we hope to extend the present LIRAQ models to include aerosols and sulfur chemistry. The latter is especially important for East Coast air pollution studies but may become

more generally applicable as more automobiles are equipped with catalytic converters.

Key Words: air pollution; air pollution analysis; air pollution computations; air pollution control; air pollution models; air pollution - San Francisco Bay Area; land-use planning; Livermore Regional Air Quality; LIRAQ.

ARAC: REAL-TIME PREDICTION OF HAZARDS DUE TO RADIOACTIVE RELEASES

The Atmospheric Release Advisory Capability (ARAC) is an ERDA-sponsored service designed to provide nuclear facilities with real-time predictions of the health hazards that might result from a release of radionuclides or other toxic materials. Since 1973, when the concept was born, a joint feasibility study by LLL and the Savannah River Laboratory has proved its workability. LLL has built and is now operating a prototype ARAC component. During the next three years, ERDA intends to implement ARAC service for six of its nuclear facilities. Implementation means installing a central facility at LLL for acquiring, processing, and communicating data; linking this facility with national and global weather services; and installing local data-acquisition, assessment, and communication facilities at the six sites.

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Facilities that handle radioactive materials have a paramount obligation to protect both their operating personnel and the public at large. Despite a remarkable overall safety record, the nuclear industry faces a great challenge to minimize damage that might result from a nuclear accident or incident, however unlikely.

Although the nuclear industry as a whole is working on this problem, the ERDA nuclear research and production facilities have a special responsibility. Typically, the sites face such questions as:

- What health hazards to operating personnel and the public would result from a nuclear accident or incident?
- More specifically, how fast and to what extent would radioactive materials diffuse under given circumstances and weather conditions?
- What kind of predictive information can be developed to help in emergency decision-making?

ERDA is now establishing the means to help site managers respond to such questions. Under a program

THE SAVANNAH RIVER RELEASE: TEST OF ARAC CAPABILITIES

On May 2, 1974, an unexpected low-level atmospheric release of tritium from the Savannah River Plant, Aiken, South Carolina, provided a realistic check of ARAC capabilities. A joint ARAC feasibility study by LLL and the Savannah River Laboratory was then in progress but not completed. Nine levels (heights) of meteorological instrumentation were functioning on a TV tower at the plant. Meteorological-data communication facilities were operational between the plant and Livermore. LLL was capable of receiving, selectively averaging, printing, storing, and graphically displaying the input data in real time.

It was quickly determined that the release because of the small quantity involved and the release site (well within the plant confines) presented no danger to the public. Thereafter, with calculations based on simple models, LLL provided supplemental information to Savannah River personnel, exercising ARAC capabilities and communications equipment. Among the transmitted data (shown in Fig. 12) were the simplified Gaussian normalized concentration plots and trajectory calculations described in the accompanying article.

Savannah River's only real-time air-concentration measurement, taken approximately 40 km from the release site, showed the LLL estimated concentration at that point to be within 20% of measurement. Both laboratories consider this outstanding. LLL successfully predicted the cloud direction and, in general, the surface concentrations. Although a few anomalies were observed in later vegetation measurements at the Savannah River site, both laboratories believe they were probably caused by small-scale variations in atmospheric motions not modeled by the larger-scale computer codes.

Fig. 12. Typical data plots transmitted to the Savannah River Laboratory by the LLL ARAC group: (a) Gaussian solution to the diffusion equation for "E" and "F" stabilities (degrees of atmospheric turbulence), with an estimate of the likely tritium concentration in water extracted from vegetation in the cloud path; (b) Gaussian solution to the diffusion equation for "D" stability (a slightly more turbulent atmosphere expected as the day progressed); (c) cloud-center, sector-average, and surface normalized concentrations for "D" stability; (d) wind profile, in which the solid curve represents measured windspeed values over a 5-min averaging period and the dashed curve represents the least-squares profile for the wind speed based on the calculated value of p (derived from a fit of the measurements to a power-law profile in the least-squares sense). The sector-average concentration is that expected over a 22° sector.

of the Division of Biomedical and Environmental Research, LLL has developed a centralized service to provide real-time (as events occur) predictions of the consequences of an atmospheric release of radioactive materials. This service is called ARAC.⁷

Purpose and Advantages

ARAC's chief purpose is to provide solid information, as quickly as possible, on the probable effects of an atmospheric release. The plan is for ARAC to send a series of advisories (processed

information and recommendations) to the site to guide its planning. Central to this concept are numerical models that provide real-time assessments - up to distances of 100 km - based on input data from the site. These models, when coupled with state-of-the-art technology for handling and communicating data and for converting radionuclide concentrations into dose-to-man, permit accurate predictions of the effects of any toxic materials released.

Although ARAC's primary function is emergency assistance, some routine uses are also planned. These

include calculating and maintaining an inventory of radioactivity at the site, calculating doses from routine operations, and performing sensitivity studies to assess the likely effects of projected facilities or changes in site operations.

The ARAC concept is quite sweeping. The predictive capability at each site is based on a local, automated system appropriate for close-in distances (to about 5 km). Links to the central facility permit countermeasure planning based on comprehensive data not immediately available at the local site. Provision is made for emergency backup links: that is, in the event of a release, countermeasure planning can proceed away from the local site. Graphic data display at several remote locations permit coordination and alternative operations. Centralization allows economies that are not practical for individual sites.

Among the benefits will be the inclusion, in ARAC predictions, of transient as well as persistent regional transport processes. This will improve the quality of the advisories. Iterative improvement of off-site countermeasures and postemergency cleanup operations will be possible, based on actual data from the release area. ARAC also will serve as the focal point for future improvements. All these will facilitate ERDA's role of assisting operating nuclear sites.

ARAC Components

The large-scale computers needed for calculating the regional advisories will be located at the Livermore central facility. Communicating with this facility through data and/or voice telecommunication will be the nuclear facilities served by the system. Any number of U.S. sites can be served. National and global meteorological services will also communicate with the central facility. If desired, ERDA or local officials will be able to receive the advisories at the same time as the individual sites.

Site Facilities. Each ARAC-served site will have a minicomputer for local data acquisition, assessment, and communication. Specific functions of the site facility will be to:

- Gather data from local environmental sensors at regular intervals.
- Provide local data-quality control.
- Continuously calculate and display concentration estimates (based on a Gaussian diffusion equation) for close-in distances, using the latest local meteorological data.
- Transmit local environmental measurements to the central facility.

- Receive and display regional calculations from the central facility.

Because the site facility can perform certain functions without a direct data link to the central facility, this was the first part of the ARAC system to be designed and tested. In FY 1975, LLL bought the hardware and started the software development for an operating site facility at Livermore. The site equipment consists mainly of a minicomputer with core memory, a graphic display console, a printer/plotter, and interfacing hardware. Software written for the Livermore site can be customized to satisfy local requirements at other sites.

The interactive operating system at Livermore allows users to display several types of information: for example, local meteorological data, concentration estimates, calculated trajectories of the released material, and advisories from the central system. Most of this information can be superimposed on maps of various scales. For example, Fig. 13 shows a normalized Gaussian diffusion calculation based on the latest available local meteorological data, superimposed on a 5-km-radius map of the LLL site.

Specifications for interfacing each site facility to the local environmental sensors have been written. These are available from LLL.

Fig. 13. Gaussian diffusion calculation overlaid on a 5-km-radius map of the LLL site for the weather conditions prevailing at the time. When this picture was generated, the contour labels were not included; in the final version of the system they will be added.

Meteorological Services. LLL now receives meteorological data from the National Weather Service (NWS). We expect to link our meteorological data-acquisition facility to the Air Force Global Weather Central (AFGWC) to get grid-point-forecast meteorological data from its fine-mesh and boundary-layer models. AFGWC would also send us the latest global observational data and general forecast information for specific areas. NWS plans to automate its service with the Automation of Field Operations and Services (AFOS) system within the next several years. When this system becomes operational, we intend to include it, too, as part of our meteorological data-acquisition facility.

Central Facility. Once the central facility begins to function, it will receive every four hours, during normal operations, site environmental data plus any site messages or instructions. The central facility will manipulate these data for storage and routine site-environment assessments.

In case of a real or potential emergency, a data and voice communication link will be established at once between the affected site and the central facility. Simultaneously, data will be requested from the meteorological data-acquisition facility, and the regional model computer codes will be made available on the large computers. The meteorological data, stored in a computer-compatible format, will be retrieved, analyzed, and used to compute a trajectory. Then these data will be fed into the large computers and used for the regional model calculations (up to 100 km in radius). This process can be repeated as often as needed, using updated measurements from the local site, until the danger no longer exists.

During the postemergency period, numerical-model calculations can be made in greater detail to assess the total environmental consequences of the release.

System Operation

In Fig. 13 we have shown a typical Gaussian diffusion estimate for a local site based on the most recent locally measured meteorology. This "quick fix," intended to give local personnel an immediate estimate of the expected normalized concentrations of released material within short distances, is the first information available locally.

Within five minutes of notification, the central facility can send a trajectory prediction to the local site. The purpose is an early estimate of the downwind movement of the release.

The regional-scale prediction is available about 35 minutes after notification. Thereafter, the central facility can send a constant stream of advisories, according to need, based on information flow and measurements from the local site. Thus operations can be adjusted to changing conditions or new field data.

Numerical Modeling and Environmental Assessment

For the detailed regional assessments we now have three-dimensional numerical transport and diffusion models to estimate the regional air concentrations and ground deposition from continuous or instantaneous point sources. We used these models - MATHEW and ADPIC - during a successful joint feasibility study with the Savannah River Laboratory, South Carolina.⁸

MATHEW is a meteorological data-adjustment model developed to provide transport and diffusion data with input windfields that are mass-consistent, three-dimensional, and representative of the available meteorological measurements.⁹ This code does not forecast the windfields but uses available measurements over the region of interest to produce windfields that are assumed to persist until new meteorological data are available and the fields are recalculated. Research is now under way at LLL and other laboratories to develop fine-mesh meteorological predictive models. As these are validated and become available, we will include them in the ARAC central facility. Meanwhile, we plan to use forecast products available from NWS or AFGWC to help define meteorological changes that might occur.

ADPIC is a three-dimensional particle-diffusion code capable of calculating the time-dependent dispersion of inert radioactive air pollutants under many conditions, including stratified shear flow, calms, varying topography, and wet and dry deposition.^{10,11} This code also has been adapted to simulate the fallout patterns of particulates with given particle-size distributions and the depletion of particulates from a plume of released material over various terrains.¹² ADPIC has been validated using measured air-concentration data taken during a tracer release at the Idaho National Engineering Laboratory (Idaho Falls)¹¹ and measured argon-41 concentration data from the Savannah River Plant.⁸ We plan further validation and sensitivity studies during FY 1976.

The ADPIC-calculated regional concentrations and surface depositions may be fed into a dose-conversion computer code, DOSCON, for deriving the individual

and population whole-body or organ doses via inhalation, external, and ingestion pathways. By using the model developed by the Task Group on Lung Dynamics of the International Commission on Radiological Protection,¹³ one can compute the dose to various organs of the respiratory tract if the aerodynamic particle size and the chemical and physical properties of the specific radionuclides are known. External exposures due to direct gamma radiation from gaseous plumes can be evaluated by integrating the activity over the volume of the plume. External exposures from surface depositions of gamma-emitting radionuclides can be derived from the calculated photon flux per unit source strength and flux-to-exposure-rate conversion factors.¹⁴ The calculation of dose from food ingestion, however, requires the use of reconcentration factors¹⁵ of radionuclides by biological processes to determine the activity of specific radionuclides in each food. Several models are available for calculating the ingestion dose through specific food-chain pathways.¹⁶ Although these dose models are not now available as ARAC models, we plan to include them during FY 1976.

Another future prospect is extension of the modeling techniques to aquatic systems. Research in this area is planned to begin in FY 1977 to permit subsequent inclusion in the ARAC capabilities.

ARAC Implementation Plan

The initial three-phase study started at LLL in 1973 provides for concept development, construction and operation of a prototype facility, and implementation of the system. The prototype phase has been completed and a three-year implementation phase begun to establish service at ERDA operating sites. This plan contemplates the incremental building of ARAC-system components until a fully operating network is completed (FY 1978). The service might then be offered to other sites.

This fiscal year, we hope to achieve several major goals of the three-year implementation plan:

- The prototype site facility already established at LLL will be made fully capable of stand-alone operation; i.e., it will not be communicating with the central facility although the necessary communication software will be completed.

- Site-facility hardware will be installed and the software customized for an ERDA research site (Savannah River Plant) and an ERDA operations site (Rocky Flats Plant, Golden, Colorado). These facilities will also be operating in the stand-alone mode. During

this interim period, advisories can be received from the central facility by telecopier or other means.

- Hardware for the basic central-facility configuration will be purchased and software development initiated. The operating system for the central facility requires extensive software effort, so the communications link between this facility and the other sites will probably not be established until early in the following fiscal year.

- A real-time operating and data-management code will be developed to run and monitor the principal regional transport and diffusion calculation (ADPIC). Also, tailored output for LLL, Savannah River Laboratory, and Rocky Flats will be designed to assist in interpreting the results of these calculations.

- A method for producing probabilistic concentration calculations based on uncertainties in the input data will be investigated. We will also investigate the use of forecast meteorological data such as the output from the Air Force boundary-layer model. These data will be factored into the site observational data such as to provide forecast capabilities for the ARAC advisories.

- Studies to validate the ARAC models and check their sensitivity to data volume will continue.

According to the implementation plan, two more sites would be added to the system each year during FY 1977-78. Communication links between the LLL central facility and the first three ERDA sites are scheduled for early FY 1977.

Because ARAC is in the initial stages of implementation, changes can be expected in its final form. However, the concept is fully developed and has been successfully demonstrated. During the LLL-Savannah River Laboratory joint feasibility study, LLL received real-time meteorological data from SRL and performed detailed regional assessments, the results of which were transmitted back to SRL. The concept was also tested realistically in May 1974 when the Savannah River Plant experienced a low-level atmospheric release of tritium (see the description at the beginning of this article).¹⁷

Although our immediate goal is to apply the ARAC service to a limited number of ERDA sites, the system is designed with enough flexibility for expansion to other sites. Success in applying ARAC should significantly help the nuclear facilities in responding to the urgent questions about radioactivity hazards.

Key Words: Atmospheric Release Advisory Capability, ARAC; nuclear safety; radioactivity measurements; radioactivity releases; MATHEW; ADPIC; DOSCON; computer models.

NATIONAL SECURITY

X-RAY-FLUORESCENCE ANALYSIS FOR NUCLEAR MATERIALS ACCOUNTABILITY

Highly accurate measurements of samples of uranium and plutonium in the light-water power reactor fuel-reprocessing cycle are needed to establish the physical inventory of these materials at each reprocessing facility. At present these measurements are being made by mass-spectrometric isotopic-dilution analysis. Although accurate, this method has several disadvantages, so we proposed an x-ray fluorescence method that would overcome them. Under a contract from ERDA's Division of Safeguards and Security, we have been developing this method for application to inventory control. The work is not yet completed, but it has progressed to the point that three U.S. companies are now marketing preliminary versions of our system. Successful completion of work now in progress should lead to an x-ray-fluorescence analysis system with the desired accuracy, speed, flexibility, and cost.

The first step in the fuel rod reprocessing cycle for light-water reactors is to dissolve cut-up sections of spent fuel rods in nitric acid. The concentration of uranium and plutonium in this dissolver solution is particularly important to the Division of Safeguards and Security because it is the first point in the reactor fuel cycle that the integrity of the fuel rods has been breached. It is also the first opportunity to measure the amount of plutonium produced by fuel burnup in the reactor. Consequently, the amounts of uranium and plutonium in the dissolver solution become the input accountability numbers for these elements for the entire reprocessing plant.

The uranium and plutonium concentrations in the dissolver solution need to be known to a one-standard-deviation accuracy of at least 0.5%. (This is state-of-the-art accuracy.) Instrumentation for making these measurements needs to be dependable and difficult to contravene. Furthermore, it must be capable of good statistical reliability despite the difficulty of obtaining representative samples of the large batches of solution. Rapid analysis of large numbers of samples is one approach; in-stream measurement of the flowing solution is another.

At present, the uranium and plutonium concentrations in dissolver solution are usually determined by mass-spectrometric isotopic-dilution analysis. This method is highly accurate, but it is expensive, slow (because it involves wet chemical steps), and not adaptable to in-stream measurement. Another method, wavelength-dispersive x-ray-fluorescence analysis, is used successfully in West Germany to measure uranium and plutonium concentrations in dissolver solutions to 1% accuracy. However, this method is also expensive and requires the addition of an internal standard to the dissolver solution before analysis—a step vulnerable to subversion.

The method we have developed uses nondispersive x-ray-fluorescence techniques, by which the energy of all the stimulated fluorescent x rays from the sample is recorded simultaneously by a lithium-drifted silicon detector. This method is much less expensive than dispersive x-ray analysis and allows the flexibility of measuring many different elemental concentrations without changing the analysis instrument. It is difficult to contravene because there are few operator-performed steps.

Our first prototype instrument, designed and built at LLL in 1973, was a relatively small, portable x-ray-fluorescence instrument operating on 115-V, 10-A power.¹⁸ It used the $L_{\alpha 1}$ and $L_{\alpha 2}$ x rays from uranium and plutonium for analysis; these x rays were stimulated by a special silver-transmission-anode x-ray tube designed at LLL. A commercial Si(Li) detector collected the signal from solution samples or samples prepared by evaporating a small amount of dissolver solution on filter paper. A computer analyzer, also built at LLL, recorded the data and calculated the uranium and plutonium concentrations. We sent the completed instrument to the Savannah River Plant (E. I. du Pont de Nemours & Co.) for a one-year evaluation in the plant's analysis laboratory.¹⁹

The in-plant evaluation uncovered several problems mostly relating to hardware rather than the instrument's basic design. In particular, the system provided an accuracy of only 1.5% (compared with the desired 0.5%) for filter-paper and solution samples bearing uranium or plutonium separately. Also, it

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could not satisfactorily measure the two elements simultaneously in the solution samples (filter-paper samples were not tested for simultaneous measurement). Based on these results, we redesigned the system and built a second prototype, which is now undergoing accuracy and reliability tests at LLL.

Among the improvements incorporated in this second prototype are a new miniature x-ray tube that produces about 10^{10} photons/cm²·s at 4 cm and dissipates only 100 W; a new scattering chamber and detector configuration; a new computer system capable of performing on-line data analysis, and a system for producing freeze-dried samples. Freeze-dried samples are preferred over evaporated samples (as in the first prototype) because analyses based on evaporated samples are subject to inconsistencies due to particle-size effects.

Preliminary data indicate that the second prototype's accuracy (for each element separately) will be about 0.5%. We also plan to modify the new system to provide simultaneous measurements of uranium and plutonium.

The evaluation period for the first prototype instrument also showed that the most desirable sampling procedure for dissolver-solution analyses would use in-stream measurement of the flowing solution itself, rather than samples removed from it. We are presently investigating ways to accomplish this. We are also investigating the possibility of using a polarized silver x-ray beam as an exciter source for measuring the high uranium-to-plutonium ratios (50:400) in the dissolver solution.²⁰

We have learned from experiments at LLL that the normal method of preparing standard samples of uranium and plutonium by pipetting is accurate only to 1% at best. We have therefore developed a gravimetric standard-making procedure that has an accuracy of 0.2 to 0.3%.²¹ These gravimetric standards are being used in tests of the second prototype x-ray analysis system.

In addition to testing the new equipment and standards-making procedure, we are also testing a new method of determining highly accurate calibration curves from the standards data. The method is a nonlinear fitting technique that takes into account the errors of the standards simultaneously with the errors generated by the x-ray analysis technique. Work has begun on a very accurate lineshape characterization for the L x rays we use for analysis. This promises to enable us to determine the ultimate precision for measuring uranium and plutonium ratios in an x-ray technique using a Si(Li) detector.

Three U.S. companies are now offering for sale preliminary versions of the x-ray-fluorescence analysis system we have developed. Several systems have been ordered for use by the nuclear fuel system industry.

Key Words: x-ray fluorescence analysis; x-ray spectral analysis; elemental concentrations; uranium; plutonium; reactor fuels - reprocessing; nuclear fission fuel cycle analysis; freeze-dried samples; particle-size effects; silicon detector; minicomputer.

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