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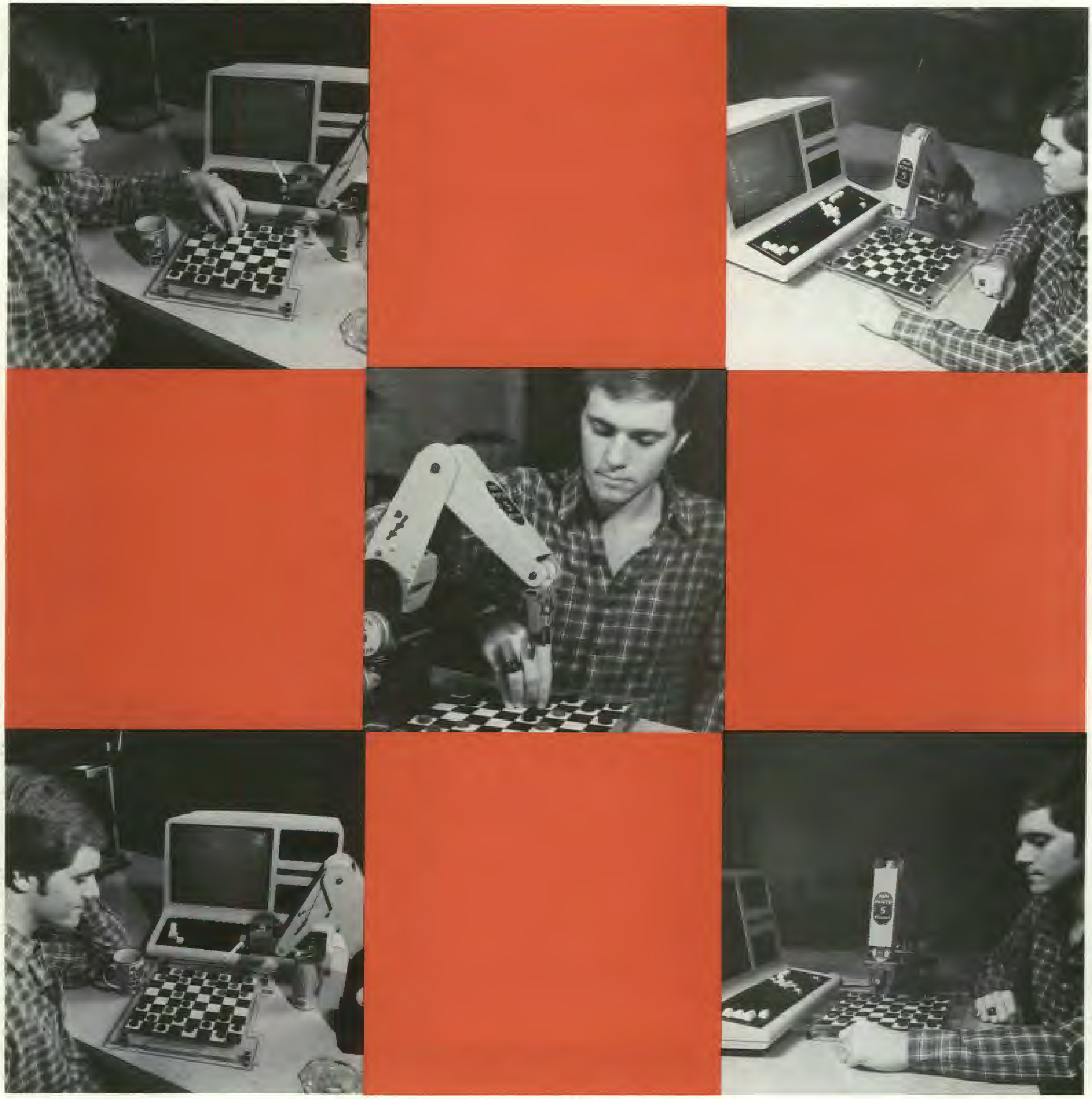
Four-Year Index



Artificial Intelligence and Robotics

THE COVER: Games robots play include checkers. ORNL mathematicians wrote a simple program to show that robots can perform an obstacle-avoidance task such as moving checkers on the board. This demonstration of artificial intelligence is discussed in the article on page 28.

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EDITORIAL: ORNL and the Environment

By HERMAN POSTMA

In its pursuit of knowledge to benefit the nation and in its activities to satisfy the needs of its sponsors, Oak Ridge National Laboratory must be a good citizen within all its jurisdictional boundaries. As an employer, it must look after the health and welfare of every one of its employees. Because ORNL conducts experiments, runs chemical processes, operates reactors, and produces radioisotopes, it has a potential for adversely affecting the environment. Therefore, we must exercise greater vigilance and improve our mutual efforts to minimize any possible adverse impacts.

What environmental impacts the Laboratory causes have been well documented over the years by ORNL staff members whose business it is to identify environmental problems and find solutions. These impacts are summarized in this issue by Roy Thoma, whose article puts ORNL's environmental risks and benefits in a proper perspective.

The Laboratory does not act independently; it has always operated under the regulating authority of a federal agency, currently the Department of Energy and previously its predecessor agencies. More recently, the Environmental Protection Agency and the State of Tennessee began the process of questioning DOE's legal right to regulate our discharges to the environment. This process has created increased public interest but has also left some misconceptions about our fulfillment of legal commitments.

Allegations by the Tennessee Department of Public Health and Environment and reports in Knoxville newspapers have exaggerated and distorted the impact of our operations by misusing the facts we have published over the years. We indeed do have environmental problems, and we have been the first to document them and seek solutions. But our operations do not threaten the health of people; in fact, as this issue of the *Review* reports, epidemiological studies indicate that ORNL employees have lower death rates from many diseases, including most cancers, than does the U.S. population as a whole. In addition, statistics show that one of the safest places one can be is not at home or on trips but working at ORNL.

We have always worked diligently to follow federal policy requiring that the Laboratory's operation comply with all environmental laws and regulations. In the past these regulations allowed more discharges into the air, water, and ground, because standards were less stringent, environmental-control technologies were much less advanced, and measurements were less sensitive than they are now. Our challenges are likely to extend into the future, but the bulk of our problems are the result of these long-past activities.

In an unfortunate spirit of sensationalism, the newspapers alleged that, because some mercury has gotten into the sediments of the Clinch River from ORNL operations and particularly from those of the Y-12 Plant, people who eat fish from the river run a high risk of mercury poisoning. In fact, the highest level of mercury measured in fish in 1982 was 0.56 part per million, well below the Food and Drug Administration limit of 1 part per million. A person could eat 1 kilogram (2 pounds) of fish per day forever without any harm whatsoever; in fact, a person would have to eat approximately 1800 kilograms (4000 pounds) of the most contaminated Clinch River fish in one sitting to ingest enough mercury to be fatal.

Fears that radioactive contaminants (which leach from ORNL's burial grounds into the groundwater) may reach public wells are unfounded, based on our knowledge of the way radionuclides are readily adsorbed by subsurface materials. Our calculations indicate that it would take 10,000 years for the contaminants to travel one kilometer (a little over half a mile) underground and that they would not reach the nearest well for 300,000 years, by which time most of their radioactivity would have decayed away.

Although White Oak Lake is alleged to be highly contaminated, actually people could pipe their daily drinking water forevermore from the lake without any more apparent health risk than they would receive from smoking two packs of cigarettes in one year. We still consider this environmental hazard to be too great, so we are taking steps to reduce leaching of radionuclides to our environment.

Above all, we hope that our staff will use the best practices technology permits to reduce our environmental effects and that our area residents have not been alarmed by the inaccurate statements and allegations reported in the media. We will continue to do whatever is technically and financially feasible to protect the environment and to ensure the good health of our staff and the citizens in our community.

How Does ORNL Affect the Environment?

By ROY THOMA

When the members of Congress decided to take steps to protect the environment in the United States, they created a classic piece of legislation called the National Environmental Policy Act of 1969 (NEPA). NEPA idealism is evident in such phrases as "attain the widest range of beneficial uses of the environment without degradation, risk to health or safety" and "achieve a balance between population and resource use which will permit high standards of living and a wide sharing of life's amenities." In a broad sense, these are principles to which Oak Ridge National Laboratory has been devoted since its inception.

Though the legislative prose of NEPA takes on the color of glowing political rhetoric, NEPA principles struck a responsive chord with the American people, who were becoming concerned about the health effects and environmental problems of air and water pollution, caused largely by discharges from motor vehicles, power plants, and industry. This landmark of environmental law expresses the desires of an increasing number of Americans who distrust big technology and

Roy Thoma was a member of the team that wrote *Environmental Analysis of the Operation of Oak Ridge National Laboratory (X-10 Site)*. This document (ORNL-5870), produced under the direction of John Boyle, was published in November 1982 and is the basis for the article that follows. Thoma's contribution to ORNL-5870 includes the analysis of ORNL's socioeconomic impacts. A member of the Environmental Assessment Section of ORNL's Energy Division, he has been involved in environmental assessment work since 1971, when the Calvert Cliffs case brought ORNL into an active role in determining the environmental impacts of power plants. In 1970 he was ORNL's representative at the first Westinghouse School for Environmental Management. Thoma came to ORNL in 1952 and worked in high-temperature chemistry related to molten salt reactors. Before that he taught chemistry at Texas Technological University and Sam Houston University. A native of San Antonio, he did his graduate work at the University of Texas in Austin. Here, he looks at the coal-yard runoff basin. In the background is ORNL's steam plant and coal pile.

ORNL's day-to-day operations for the most part have a beneficial effect on the human environment, although the impacts of its primary product—new scientific and technological information—are speculative. Slightly adverse impacts arise from releases of toxic materials from research activities; however, none of these releases is a threat to human health.

who have embraced the environmental movement to protect people, mammals, fish, birds, vegetation, water, air, and land from industrial pollution and misuses. Thus, NEPA has become one of the most powerful pieces of legislation ever enacted—one that has caused many changes in this country.

In response to NEPA, the American government in 1970 began to reexamine its previous actions that affected the environment and to incorporate environmental planning into proposed new major actions. A federal court decision in 1971 forced the Atomic Energy Commission (AEC) to consider the total environmental impact (not just the radiological impact) of nuclear power plants licensed by AEC after the law was passed. ORNL, as a national scientific resource, was asked by AEC to take a lead role in evaluating the total environmental impact of nuclear power plants. Thus, the Laboratory was thrust into addressing and solving national environmental problems.

This is the first in a series of articles on ORNL's impact on the environment. Most of the article is based on a 1982 environmental analysis; however, some of the statements in the article have been updated to reflect changes in the past two years. Helen Braunstein of the Department of Environmental Management in the Environmental and Occupational Safety Division has been responsible for the updating. The second article, to appear in the next issue, will deal with the compliance evaluation inspection of ORNL performed recently by the State of Tennessee and the Environmental Protection Agency.

But how about environmental problems at ORNL?

Because all aspects of the operations on the Oak Ridge Reservation, including the protection of the environment, rested with the agency responsible for administering programs on the site and because that agency possessed regulatory authority, the operations were exempt from the regulatory authority of any other agency. From the beginning, all federal agencies were subject to NEPA; however, they were expected to comply substantively, rather than procedurally, with the law. Operations were to be self-regulated; they were not subject to licensing, permitting, or inspection. Thus, activities at national laboratories were expected to adhere to the substantive meaning of NEPA, but they were exempt from regulation by either the U.S. Environmental Protection Agency (EPA) or the U.S. Nuclear Regulatory Commission (NRC).

Despite the fact that ORNL was not subject to formal assessment under NEPA, ORNL officials were mindful of the new attitudes sweeping the nation and began to examine Laboratory practices that were affecting the environment. While ORNL had always carried out its work with radioactive materials in ways that led to the development of advanced technologies for handling and disposing of these materials, ORNL managers found that a number of the Laboratory's earlier methods for disposing of radioactive and hazardous waste materials were inadequate. That is, these methods did not measure up fully to interpretations of some of the principles enunciated in NEPA. What was done before the enactment of NEPA was based on the prevailing concept at that time—that dilution would take care of environmental consequences. Little environmental technology existed to guide planners and decision makers, so their decisions were based on the effectiveness of what they thought were acceptable practices. Before NEPA, even the most concerned individuals did not know enough to understand that some practices might fail to meet our obligation to protect the environment for generations not yet born.

Varying Impacts

The operation of a major technical complex such as ORNL has widely varying environmental effects. In attempting to evaluate these effects by weighing the environmental "costs" against the environmental "benefits," standards must be established, contributing factors identified, and values assigned.

EPA established the standards. It called for actions to protect and enhance the environment and to improve the "quality of life." These are clear objectives. Frequently, however, they call for value judgments, and value judgments are subjective. Even the selection of contributing factors (those elements of the action that influence the environment) entails bias. Because environmental considerations contain an almost frustrating mixture of science and nonscience, meeting the challenge of NEPA—which requires assessment of the significance of an action—ultimately rests almost as much on subjective as objective criteria.

What about ORNL's impacts?

ORNL's pursuit of nationally important scientific and technological information, its primary product, affects the human environment. Of the two principal impacts, one is socioeconomic, namely, the effect of research and development (R&D) results on society; the other is the effect on the environment of the dispersion of materials. A subsidiary local effect is the employment of about 4500 persons (1984 figures) and the consequent community well-being.

The ultimate impact on society of ORNL's R&D is unlikely to be evaluated in the short term because until R&D becomes incorporated into the body of science or becomes applied technology, its potential impact remains a matter of speculation. Moreover, once applied—for example, in new materials for nuclear and coal-burning power plants—ORNL's contribution will merge with that of other institutions, and recognition will be lost.

Nevertheless, the Laboratory's scientific strength is internationally acclaimed. Many members of the research staff have been recognized by their peers through awards, appointments, citations in the scientific literature, and invitations to participate in collaborative efforts both here and in other countries. ORNL staff members who have chosen to leave have often moved to positions of leadership in government, industry, and universities.

A more recent record of ORNL's contribution to technology, as measured by an external standard, is the 37 I•R 100 awards presented to the Laboratory. These awards are given annually by *Industrial Research & Development* for the year's most significant advances in technology. ORNL has received more I•R 100 awards than any other U.S. Department of Energy laboratory. It may be many years, however, before we know whether these prizewinning ORNL advances will become commercial products and processes that are useful to consumers or to society as a whole.

Although ORNL's impact on the world of science and technology is largely beneficial, one cost of operating this large research organization is the generation of wastes, some of which inevitably are released to the local environment. Thus, while ORNL became engaged in preparing NEPA assessments for the nation's nuclear and nonnuclear energy facilities in the 1970s, it also examined environmental problems in its own backyard. These problems, some of which originated as early as 1943, concerned mainly (1) radiological releases and the storage and disposal of radioactive wastes generated by the Laboratory; (2) sewage treatment of liquid wastes; and (3) disposal of nonradioactive wastes, including hazardous materials and coal-pile runoff. What are these problems in detail, and how do they affect the environment?

Radioactive Discharges: Gases

ORNL's Waste Management Operations Program is responsible for managing the Laboratory's radioactive wastes, both those currently being generated and those produced in programs dating back to 1943. A long-term objective of this program is to ensure that new technology and facilities will comply with future regulations. In fact, ORNL-developed technology frequently sets new standards to which regulations are adjusted. A more immediate objective is to reduce current radioactive discharges to the environment by upgrading facilities constructed in the 1940s and early 1950s. Since 1970 the Laboratory has spent about $55 million on waste management improvements.

Originally, nearly all waste gases, including those containing hazardous materials, were discharged through stacks. Because suitable filters were unavailable, particulate matter remained in gaseous effluent streams. Over the past 40 years, filters and techniques have been developed to remove most of the particulate matter and chemical vapors from gas streams. Today, most radioactive substances are removed by passing all waste gases through primary cleanup systems connected to the Laboratory's five research reactors, hot cells, and engineering development laboratories. A caustic scrubber removes reactive gases, and high-efficiency filter units remove particulate matter. The gaseous waste system, installed more than 20 years ago in the two Bethel Valley stacks from which most of the gases are discharged, is currently being upgraded at a cost of about $13 million. This system includes highly sophisticated monitoring equipment to ensure the efficacy of the removal systems and prevent accidental discharges.

But what about health impacts due to gaseous emissions?

To assess the health impacts of gaseous releases at the point of maximum exposure, ORNL health physicists used 1981 release data and made the assumption that the particulates emitted would be soluble in the lung upon inhalation. They found that the estimated annual total-body dose to the maximally exposed individual is 0.38 mrem, to which tritium contributes 95%. All doses are therefore well below 500 mrem/year, the maximum allowable standard (set by DOE) for exposure of the total body, gonads, and bone marrow, and far below 1500 mrem/year, the maximum standard for other organs.

The total-body dose ORNL contributes to the regional population is 11 person-rems/year, primarily from tritium releases. This amount is only about 0.01% of the dose people receive from natural background radiation. ORNL calculations show that radiation releases from routine ORNL operations can be expected to cause virtually no deaths from cancer: 0.0004 death per year to the world population, 0.0003 death per year to the U.S. population, and 0.0002 death per year to the regional population around ORNL.

Radioactive Discharges: Liquids

ORNL routinely handles large volumes of radioactive liquid wastes, nearly all of which is low-level waste (LLW). Currently, no high-level waste (HLW) is produced, but an HLW system that consists of two stainless-steel tanks in an underground concrete vault does exist in case it is needed.

When ORNL was built, an extensive underground piping system was installed to collect LLW. This waste was to be collected from the various ORNL facilities and transported to a central group of six "gunite" storage tanks. As ORNL grew, the storage tanks became inadequate to handle the volume of radioactive waste generated. The problem was solved temporarily by precipitating the wastes in the storage tanks with caustic and allowing the solids to settle; thus, the liquid above the radioactive sediment contained only a small fraction of the original activity. This liquid was drawn off to a large pond for further settling and then discharged into White Oak Creek, which flows eventually into the Clinch River.

This procedure was abandoned in 1949, and from June 1949 to June 1954 the LLW was concentrated in a pot-type evaporator. The radioactive concentrate was returned to one of the storage tanks, and the nonradioactive condensate was discharged to White Oak Creek. During this period the underground piping system was expanded by adding collection tanks near the various liquid waste sources.

From 1954 until 1962 the liquid wastes were discharged to three open pits, where they were concentrated by evaporation and fixed in place by ion exchange on the Conasauga shale. Three more waste pits were constructed as long, narrow trenches, from which the wastes seeped into the underlying shale beds, where they were fixed in place by ion exchange. These seepage trenches were used until 1965. In 1966 the practice of disposing of the concentrated wastes by hydrofracture was inaugurated, and this method is still in use today.

Because hydrofracture is a proven technique, it is the method of choice for disposal of radioactive liquid wastes. In the hydrofracturing process, hydraulic pressure is used to form cracks between layers of shale. An alkaline LLW solution is mixed with cement and other additives to form a grout. The grout is injected under pressure into the crack in the impermeable shale formation at depths between 210 and 300 m. As the injection continues, the grout fills the crack and extends it to form a thin horizontal sheet 200 to 300 m across. The grout sets in a few hours after injection, thereby permanently fixing the radioactive wastes in the shale formation. Monitoring over the years indicates that the sequestering of liquid wastes by hydrofracture is permanent.

Most liquid wastes are collected and pumped into underground shale formations in ORNL's Hydrofracture Facility. This new facility, which made its first injection in June 1982, is designed to inject up to 760,000 L (200,000 gal) each time. It consists of the injection well, a network of observation and monitoring wells, storage tanks, and associated equipment. Replacing the original facility used from 1966 to 1979, it provides storage for concentrated wastes until they can be mixed with grout and injected into the Conasauga shale formation.

The LLW stream of radioactive wastes is treated by evaporation to produce a concentrate containing virtually all of the radionuclides. The resulting condensate, which is virtually free of radionuclides, is treated along with other process waste by ion exchange before release to White Oak Creek. The creek, which meanders through the ORNL reservation and has been dammed since the mid-1940s, forms White Oak Lake, originally a settling basin that inhibited off-site dispersion of many of the radionuclides and chemical pollutants discharged into White Oak Creek from ORNL facilities. Over the years, approximately 130,000 m^3 (4.5 million ft^3) of sediment was collected containing an estimated 644 curies of activity. While White Oak Lake was constructed to act as a final settling basin for ORNL's wastes, it is no longer used for this purpose. It currently is used (1) to isolate contaminated sediments from potential pathways to humans and (2) to provide the capability, in the event of a major release, of stopping the discharge of water to the Clinch River until recovery steps can be taken.

Are the liquid discharges from White Oak Lake a danger to the environment and to the surrounding population?

The lake's deeper sediments have been contaminated by past operations; sediment will continue to collect in the lake naturally, burying old sediments and thus preventing their radioactivity from moving even during storm events. To further prevent potential radiological exposure, White Oak Lake and most of White Oak Creek's watershed are within a restricted access area.

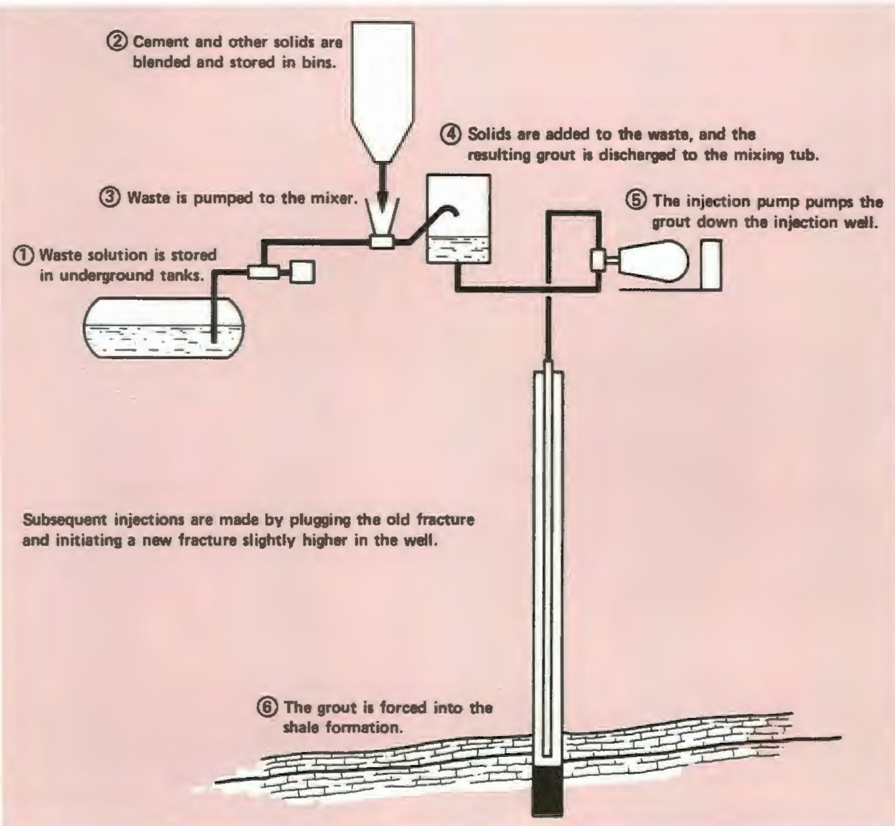
ORNL' new Hydrofracture Facility.

In 1982, the discharges leaving the lake at White Oak Dam met the concentration guides specified in the Department of Energy Order 5480.1, Chapter XI, for the general public. After the discharges reach the Clinch River and dilution takes place, the radioactivity concentrations meet the requirements for drinking water specified in EPA's primary drinking water standards (40CFR141).

The hazards to ORNL employees and the local population from releases of radionuclides to the creek, lake, and river are very small. The maximum individual total-body dose from all aquatic pathways is only 6 mrem/year, about 1.2% of the allowable standard of 500 mrem/year. The maximum organ dose to the bone is only 8.2 mrem/year, about 0.6% of the allowable standard of 1.5 rem/year. The total body dose from aquatic pathways (6 mrem/year) is not an actual dose but a theoretical one; probably nobody receives any amount close to this dose because it would require eating sizable quantities of "worst-case" fish.

Radioactive Solids

More than 170,000 m^3 (6 million ft^3) of solid radioactive waste has been buried in ORNL's six solid waste disposal areas. Solid radioactive wastes include the unwanted



Each injection into the Hydrofracture Facility is a large batch operation that requires about ten hours to complete.

by-products of ORNL's six operating reactors (used for research and isotope production) and a variety of contaminated items such as glassware, paper, rags, tools, valves, pipes, dirt, concrete, and building materials. How solid radioactive wastes are disposed of at ORNL depends on whether the wastes contain accountable quantities of uranium-235, transuranic (TRU) elements, or general radioactive wastes. Most of the wastes are handled routinely as solid LLW. ^{235}U-contaminated wastes and TRU wastes require special handling, but their total volume is quite low.

By law, the amount of fissionable ^{235}U in any material about to be delivered to a waste storage area must be accounted for. Once the amount of fissionable material is determined and precautions are taken to make sure the amount of ^{235}U is limited to avoid a criticality accident, the material is stored in unlined auger holes, which are filled and capped with concrete. A record is kept of the location and contents of each particular fissile package.

TRU wastes are handled according to the radiation level of the individual packages. About 75 m^3/year (2650 ft^3) is stored retrievably for eventual transportation to a federal repository. TRU wastes having radiation levels less than 200 mrem/h are normally packaged in stainless-steel drums, tagged, and transferred to the Retrievable Drum Storage Facility, which consists of concrete block structures that have the capacity to store up to 3500 drums [holding 110 or 210 L each (30 to 55 gal)]. Those TRU wastes having radiation levels greater than 200 mrem/h are normally packaged in reinforced concrete casks, which were formerly stored retrievably in trenches and, since January 1980, have been stored in a reinforced concrete building. TRU wastes with very high levels of beta-gamma activity are stored in stainless-steel-lined wells that are closed by concrete shield plugs.

Disposal of the much more abundant LLW is usually accomplished by depositing the wastes in either trenches or auger holes. Good engineering practices are used to develop the trenches, and the topological and hydrological features of the trench sites are given due consideration. The trenches are constructed and maintained to isolate the wastes from surface water and groundwater. If the trench is inadvertently excavated below the water table, it is backfilled with Conasauga shale to at least 0.6 m (2 ft) above the existing water table. A monitoring well is installed after trench closure.

Four of ORNL's six solid waste disposal areas have been closed; of the remaining active areas, one is reserved for retrievable storage of TRU wastes and the other is used for LLW. A seventh solid waste disposal area is planned.

In November 1983 DOE announced plans to build a Central Waste Disposal Facility to provide future capacity for solid LLW generated at Oak Ridge Gaseous Diffusion Plant (ORGDP), the Y-12 Plant, and ORNL. The location is on West Chestnut Ridge on federal property northwest of ORNL. According to DOE plans, the facility will be built and operating in 1985, and at current rates of disposal for all Oak Ridge plants, it will have the capacity for 40 years' worth of wastes.

(Continued on page 10.)

Shirley Fry (standing), acting director of the Center for Epidemiologic Research, confers with Dianne Gresham, assistant manager of the Death Certificate Retrieval Office (DCRO), who is entering data into computer files. The DCRO collects information on the vital status and the causes of any deaths among former DOE employees.

Health of ORNL Workers Gets High Marks

Preliminary results from a study of causes of death among Oak Ridge National Laboratory employees suggest there are no radiation-related health risks associated with employment at ORNL. Furthermore, results of epidemiological studies suggest that the death rates from many diseases, including most cancers, are lower for ORNL employees than for the U.S. population.

The ORNL employee study is part of the U.S. Department of Energy's Health and Mortality Study, a long-term epidemiologic research project that has been examining occupational health records and death certificates of current and former DOE (and predecessor agency) employees and employees of DOE contractors. Begun in 1964 by the Atomic Energy Commission (AEC), the project searches for any adverse health effects of long-term, low-level radiation. Because most groups with a history of radiation exposure are relatively small in number, the 500,000–600,000 DOE employees (going back to the Manhattan Engineer District of the 1940s) form a uniquely large population for the study of low-level effects.

The major portion of the DOE Health and Mortality Study is being carried out by Oak Ridge Associated Universities (ORAU), which recruited the School of Public Health at the University of North Carolina at Chapel Hill (UNC) to help with the analyses. The University of Alabama in Birmingham also plays an important role. Epidemiologists at Los Alamos National Laboratory have the responsibility for studies of DOE plutonium workers, while researchers at the Hanford Environmental Health Foundation and Battelle-Pacific Northwest Laboratory study all the workers at the Hanford facilities.

Shirley Fry, a physician and acting director of the ORAU Center for Epidemiologic Research, directs the ORAU-UNC studies of employee populations at all major Oak Ridge, Paducah, Portsmouth, and Savannah River plants. ORAU has prime responsibility for collecting and computerizing the data, determining vital status, searching for death certificates, and managing the project.

Data analyses are collaborative efforts, with an epidemiologist from ORAU or UNC having the lead on a specific study. UNC has had main responsibility for analyzing the data for the ORNL employee population. At the Health Physics Society's midyear symposium in January 1983, Harvey Checkoway, UNC lead epidemiologist for the collaborative study with ORAU, stated, "The most impressive finding from this study is the ORNL workers' apparently favorable mortality experience for most causes of death when comparisons are made with the U.S. population."

The current ORNL study population consists of 8681 white males who worked at ORNL for at least 30 days between January 1, 1943, and December 31, 1972. Although approximately 20,000 people worked at ORNL during this period, the study cohort was limited to persons not known to have worked at any other DOE (or AEC) facility in Oak Ridge. This limitation of the study to men who have worked at ORNL (but not at the Y-12 Plant or Oak Ridge Gaseous Diffusion Plant) helped to ensure that the evaluation of radiation exposures and related health effects would reflect the ORNL experience.

Vital status was determined for 90% of the cohort, and cause of death was obtained for 94% of the identified deaths. (All medical records are kept confidential under the Privacy Act of 1974.) Most of those for whom vital status was not determined were foreign consultants and workers with less than six months at ORNL.

Radiation exposure data were obtained from ORNL computer tapes containing personal radiation monitoring data. They showed that cumulative exposure levels of individual workers were generally low, with a median dose of 200 mrems per year, or 4% of the maximum permissible standard of 5 rems per year. Only 8% of the worker population studied received cumulative exposures of 5 rems or greater. Of those receiving internal radiation exposure, only 3% had whole-body counts of greater than 15% of the maximum permissible body burden or bioassays exceeding levels that require some action.

Mortality patterns of the ORNL cohort were analyzed by comparison with the mortality experience of the U.S. white male population during the period 1943–1977. Relative risk was determined as a ratio of observed to expected number of deaths—what epidemiologists refer to as Standardized Mortality Ratios (SMRs).

The ORAU-UNC epidemiologists report that the SMR for all causes of ORNL employee deaths is significantly low, 0.74. Based on general population patterns, 1374.8 deaths were expected, but only 1017 occurred. An important reason for the low mortality is the "healthy worker phenomenon"; working people are usually healthier than the general population, which includes the chronically ill and others unfit for employment. Also, ORNL employees generally have high levels of education and income—factors associated with good health—and have access to good medical care.

By examining selected causes of death, the ORAU-UNC group found that deaths in the ORNL population from all major causes were below the general population average, except for a nonspecific category (SMR = 1.74), defined in the *International Classification of Diseases* as "symptoms, senility, and ill-defined conditions." This category would include, for example, a death certificate notation of death caused by "old age." Low SMRs for malignant neoplasms (SMR = 0.75) and diseases of the blood-forming organs (SMR = 0.62) in ORNL employees are especially notable because these diseases have been associated with radiation in some studies of exposed populations.

A separate look at deaths caused by specific cancers showed that the ORNL population fared better than the general U.S. population for all but prostate cancer (SMR = 1.13; 14 observed versus 12.4 expected; 95% confidence interval, 0.62–1.89), leukemia (SMR = 1.16; 13 observed versus 11.2 expected; 95% confidence interval, 0.62–1.99), and Hodgkin's disease (SMR = 1.28; 6 observed versus 4.7 expected; 95% confidence interval, 0.47–2.78). None of these SMRs was found to be statistically significant, and prostate cancer and Hodgkin's disease have not been associated with radiation exposure in any populations previously studied.

The relationship between length of employment and health effects was also studied. A weak correlation with length of employment was found for leukemia and prostate cancer, but that result was equivocal because no deaths from these causes occurred in employees who had the longest service. This discrepancy could be explained by the lack of a sufficiently large population in the longest service categories for the purposes of this study.

In the next stage of the ORNL employee study, the researchers will be doing more refined analyses, looking at groups by job title to determine if exposure to other factors, such as chemicals in the work place, is affecting mortality. "A problem in doing a study like this," says Shirley Fry, "is that there are confounding factors that must be considered, such as industrial chemical exposure, smoking, alcohol consumption, and genetic and life-style factors, which all go into making up good or bad health. Occupational radiation exposure is just one factor."

Special studies are under way to look at brain and hematological malignancies among workers at all DOE facilities in the Oak Ridge area and to study the health effects of exposure to specific chemicals, such as mercury and nickel. A nationwide study of all DOE employees who exceeded the 5-rem exposure limit set by the U.S. Nuclear Regulatory Commission and DOE is also under way; that study includes some ORNL employees.

It will be several years yet before the entire Health and Mortality Study is complete. During this time, the focus of the efforts will be to determine whether health effects from long-term, low-level radiation can be detected and whether specific toxic chemicals present in the nuclear industry have confounding effects on the health of workers, including those employed at DOE research and development laboratories such as ORNL.—*Russ Manning, ORAU Office of Information Services.*

Transuranic (TRU) waste storage area in solid waste storage area No. 5. The TRU Waste Drum Assay Station, the aboveground metal building (upper right), is used to determine the TRU isotopes present in the waste packages (stainless-steel drums). Underground reinforced-concrete storage bins (lower center) are used to store wastes contaminated with TRU isotopes in concrete casks. These concrete casks provide the necessary shielding to protect workers from the beta-gamma radiation from the wastes. The array of wells (center, just above construction contractor's trailer) also provides storage of TRU wastes in capsules. The geometry of the wastes permits a more concentrated package, which is unshielded but which is brought to the storage site in lead-shielded casks. To the left of the road intersection are two structures for storing drums of wastes that can be contact-handled safely by workers. The reinforced-concrete cell-covers adjacent to the trailer were positioned on top of each cell in the structures. The building at the top is used for short-term temporary storage of TRU waste drums. Construction of the underground facility was completed in 1979.

Is there any need for concern about buried radioactive solid wastes?

After radioactively contaminated solid wastes are buried, they can release radionuclides to the creek and river as a result of runoff from rainstorms and melting snow as well as leaching by groundwater. Radionuclides that have reached aquatic pathways in this manner include strontium-90, cesium-137, iodine-131, ruthenium-106, cobalt-60, tritium, and some transuranics. In checking on the amount of releases, ORNL monitoring personnel have determined that the floodplains and solid waste storage areas at the Laboratory contribute about 65% of the total ^{90}Sr discharged to White Oak Creek, with as much as 30% coming just from solid waste disposal area No. 5.

The discharge of ^{90}Sr to White Oak Creek results from groundwater seepage into old trenches where LLW are buried. Remedial actions have been taken to prevent the seepage of water into these trenches. In 1976 bentonite seals were placed over 49 pits and trenches to prevent downward percolation of rain that falls directly on the backfilled areas. However, this action did not solve the whole problem. The water table still lies above the trench floors, and groundwater apparently enters the trenches by flowing laterally through their walls beneath the bentonite cap. Thus, other remedial actions are under way, such as a groundwater diversion system using drainage pipes that has been proposed to intercept the lateral groundwater flow at the perimeter of trench areas. The objective of all remedial actions in closed burial sites, as well as procedures in new areas, is to prevent any access of surface water and groundwater to buried waste.

Radioactive effluents are normally not discharged into ORNL's sanitary waste system. However, radioactivity has recently appeared in this system, largely because radioactively contaminated groundwater has flowed into the sewage effluent streams through broken and corroded pipelines that are more than ten years old. Some of the pipelines that originally carried radioactive effluents are no longer used; however, because they are in disrepair, they remain sources of radioactivity to groundwater. Other pipelines in disrepair are in-service sanitary lines, which may contribute to the flow of contaminated groundwater into the sanitary sys-

Bales of solid low-level radioactive wastes in solid waste storage area No. 6. These bales are produced at ORNL's low-level solid waste compactor. A temporary rain shield is kept in place until the trench is full of wastes, when it is backfilled.

tem. The matter is under serious study for implementing corrective actions as rapidly and effectively as possible.

What have we done to correct ORNL's environmental problems?

Since early 1982 ORNL has committed $21 million for upgrading the systems for handling and tracking its radioactive wastes. The exhaust system for radioactive gases at the Laboratory's main complex has been overhauled; weathered and corroded ducts, louvers, and filters are being replaced with stainless-steel components, and new motors and fans are being installed to improve the energy efficiency of the system, which filters, scrubs, and measures the radioactivity in the vented gas.

In addition, a Waste Operation Control Center is being built to collect data from the 250 sensors scattered around ORNL's grounds. The new system will record the data automatically; until now, these data had been manually recorded and logged. The new center will greatly increase the speed of detecting trends or sudden changes in conditions.

Improvements in three stations to monitor stream flow at White Oak Dam, White Oak Creek, and Melton Branch have been made to control flow and to allow measurements of radioactivity at both normal and flood levels.

Other stream-monitoring stations have been or are being installed on Northwest Tributary, Raccoon Creek, and Ish Creek to provide data on radionuclide movement out of past and future waste disposal areas.

Upgrading of the environmental air-monitoring systems is also in progress. Three meteorological towers were erected to provide data on the direction and dispersion of ORNL emissions. The Perimeter Air Monitoring network, which provides environmental data along the boundary of the Oak Ridge Reservation, is currently being replaced with new, automated monitoring equipment. An environmental monitoring computer is also being procured to gather, store, and process the data from the new monitors on a real-time basis. This upgrading effort amounts to an additional expenditure of $1.5 million.

Are radioactive discharges our only environmental problem?

Nonradioactive Discharges: Gases

Until recently, a secondary level of importance was accorded the treatment of nonradioactive wastes because existing waste disposal facilities and practices had prevented serious pollution of the immediate environment. Even today, while many on-site facilities need upgrading, the concentrations of most chemical species that enter the environment from ORNL are at or below established limits.

ORNL's five-boiler steam plant provides heat to warm the buildings of the Laboratory complex. In 1979 in response to concerns about impending shortages and rising prices of oil and natural gas, four of the gas-fired boilers in the 35-year-old plant were converted back to coal (2–3% sulfur). The fifth boiler is fired by oil (containing about 1.5% sulfur) or natural gas.

The steam plant must be available at all times to ensure the operability of steam turbines used for backup emergency operation of gaseous exhaust systems for reactors and fume hoods. Generally during extremely cold weather, all units are operated simultaneously so that the fifth unit can provide a continued supply of steam if an outage occurs in the coal-fired units.

Each year the steam plant consumes up to 29,000 tonnes of coal per year and yields up to 2320 tonnes of fly ash. The ash is discharged to a storage silo, trucked to a waste disposal site near the steam plant, and deposited in a landfill in trenches located above the water table. The steam plant has electrostatic precipitators that remove about 99% of the particulates emitted by the burning coal, preventing their escape to the atmosphere.

When it operates at full capacity, the steam plant discharges sulfur dioxide and nitrogen oxides at rates of 44 g/s (250,000 lb/month) and 6.9 g/s (40,000 lb/month), respectively. These rates are in compliance with state standards.

Air-rotary and soil-auger drilling rigs ► set up on a drill site on the West Chestnut Ridge Site, where the Central Waste Disposal Facility will be built.

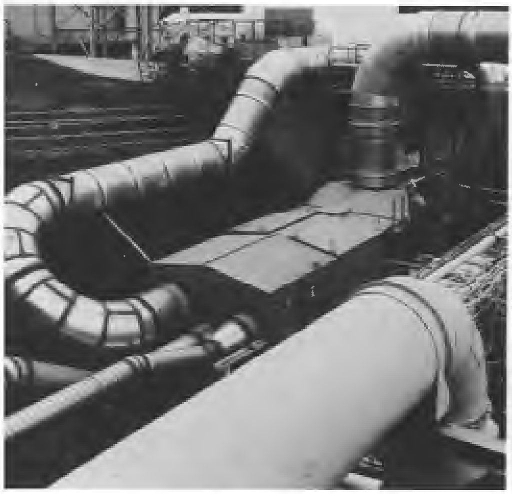
A geophysicist with Woodward-Clyde Consultants prepares to log a bedrock exploratory boring at the West Chestnut Ridge Site.

Nonradioactive Discharges: Nonsanitary Liquids

Besides these discharges of fly ash and noxious gases, ORNL's steam plant also can release pollutants to the environment when water from precipitation leaches toxic metals from the pile of coal used to fuel the plant. Coal-pile runoff is collected in an impoundment, treated to adjust the pH (degree of acidity) and remove solids, and then discharged into White Oak Creek at rates that may amount to as much as 18,000 m^3/year (4.78 million gal/year). ORNL has provided funds to modify the existing system by adding a clarifier in which solids will be removed from the neutralized coal-pile runoff, thus producing a sludge that will be dewatered and disposed of as a nonhazardous waste.

In addition to discharges from the steam plant, liquids from the sanitary sewage disposal system enter White Oak Creek from which they ultimately are discharged into the Clinch River. ORNL's other point sources of potential contaminants include discharges from numerous facilities and blowdown from cooling towers. Nonpoint sources include runoff from parking lots, streets, buildings, and the grounds, plus runoff and seepage from disposal areas for radioactive solid wastes. Although wastes discharged from nonpoint sources and potential contaminants from some ORNL facilities have not in the past been monitored, the water quality currently observed in White Oak Creek and the Clinch River can be used to gauge the environmental effects of ORNL operations.

The average flow of the Clinch River, 150 m^3/s (5300 ft^3/s), causes dilution of the materials discharged from White Oak Creek, by factors



Construction at the off-gas stack, Central Radioactive Gas Disposal Facility (Building 3039), to replace old ducts with new stainless-steel ducts.

This new weir structure with stilling well and low-flow and high-flow weirs at White Oak Dam (looking upstream) allows the discharge flow of radioactive effluents to the Clinch River to be measured during flood conditions as well as normal flows.

View from the air of the new monitoring station at White Oak Dam.

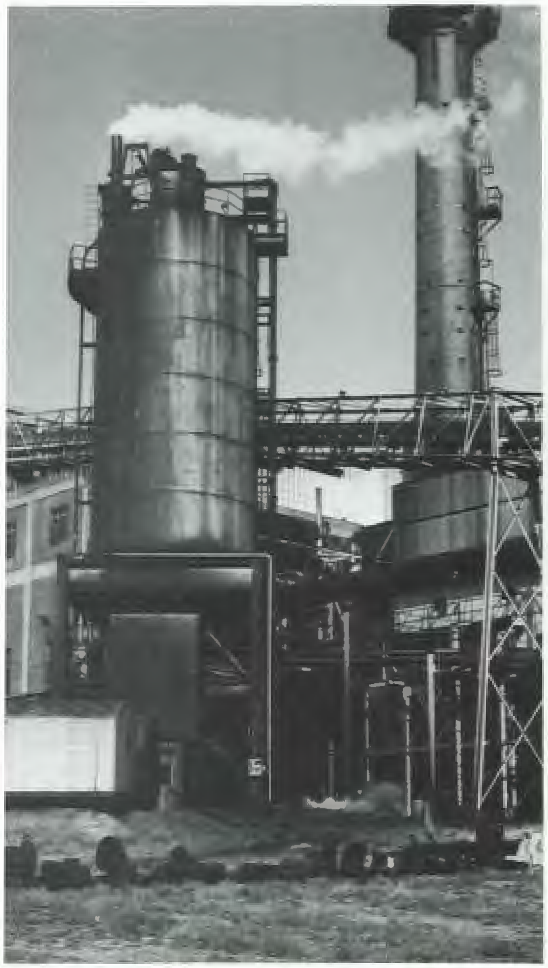
that can vary from 200 to 1000 depending on flows; thus, these discharges have a negligible effect on the water quality of the Clinch River. For example, the concentrations of nitrate and phosphorus in White Oak Creek are reduced to trivial levels as a result of mixing in the Clinch River. The principal impact of nitrate and phosphorus from White Oak Creek has been the growth of algae blooms in White Oak Lake and the embayment.

But how about heavy metals?

The average concentrations of cadmium, copper, and mercury in the Clinch River *upstream* from ORNL have been found to exceed EPA criteria and also to exceed the average concentrations released to the river from ORNL. Consider the case of cadmium in White Oak Creek. At a station in White Oak Creek *upstream* from ORNL, the average concentration of cadmium (0.12 μg/L) is approximately four times the EPA criterion for protection of aquatic life. However, aquatic life near this station is undisturbed by this background cadmium level, suggesting that the EPA criterion is more stringent than necessary for protection of indigenous freshwater life in the White Oak Creek basin. The average concentrations of cadmium, copper, and mercury at 16 stations in White Oak Creek downstream from ORNL have sometimes been found to exceed EPA's water quality criteria for protection of aquatic life, but these values are substantially *less* than the average ambient concentrations in the Clinch River *above* ORNL's White Oak Creek discharge.

Of the trace element contaminants in White Oak Creek embayment and White Oak Lake, mercury may represent the most serious problem. Ten bluegill sampled from the lake by ORNL aquatic ecologist Jim Loar in 1979 were analyzed for seven trace elements. The concentrations of all the elements except mercury and nickel were within the range found in fish from uncontaminated environments. The nickel level was only slightly elevated, but the average mercury concentration was 0.70 part per million (ppm), which is 70% of the level (1 ppm) at which the Food and Drug Administration recommends action such as periodic measurements of levels in the environment. One fish had a mercury concentration of 1.07 ppm, which is right at the action level. Because the lake lies within a restricted area, it is not accessible for public fishing. However, as an uncommon event, fish can enter the Clinch River if they are washed over White Oak Dam. This introduction of White Oak Lake fish to the Clinch River would only slightly increase the potential for mercury ingestion by sport anglers, because there is a large population of fish in the lake and because the mercury is not passed on to progeny. (In 1982 the highest level of mercury measured in fish in the Clinch River was 0.56 ppm, well below the 1 ppm limit.) Elevated concentrations of mercury, in both sediments and fish tissue in White Oak Lake and the embayment, indicate a significant degradation of water quality in these bodies of water.

Mercury and the other toxic metal contaminants can enter



Thoma surveys the environment of ORNL's steam plant.

White Oak Creek from both nonpoint sources and numerous point-source discharges. However, the source of present water quality degradation is most likely not current discharges but past discharges that have contaminated lake and stream sediments; contaminants released from the sediments contribute to the decline in water quality. White Oak Lake acts as a sink for many of the trace elements and nutrients that flow in from White Oak Creek and Melton Branch. At White Oak Dam water quality continues to improve, so at present, discharge from the dam appears to have an immeasurably small incremental impact on the overall (nonradiological) water quality of the Clinch River.

Thoma examines the runoff from the coal pile by ORNL's steam plant.

Are ORNL's sanitary discharges a problem?

Nonradioactive Discharges: Sanitary Liquids

The central treatment plant of ORNL's sanitary sewage system is ten years old. This two-stage, series-flow aeration lagoon system provides secondary treatment for organic wastes using aerobic bacteria. The two lagoons are lined with a membrane to prevent seepage into the ground. Air is blown through pipes to aerators located on the lagoon bottoms. The lagoons, which hold about 3800 m^3 (1 million gal) each, detain the liquid wastes as long as 11 days.

Effluent quality criteria are determined by a National Pollutant Discharge Elimination System (NPDES) permit. The NPDES permit requires that different types of materials in liquid effluents be sampled at a certain frequency to make sure that the maximum allowable concentration, or standard, is not exceeded. Ammonia, biological oxygen demand (BOD), suspended solids, fecal coliform bacteria, chlorine residual, and pH are sampled according to permit specifications.

Values for settleable solids, pH, and fecal coliform bacteria are almost always in compliance with NPDES permit criteria. However, keeping within limits for BOD and ammonia in the discharge has not been possible with the current aerated lagoon system. To ensure compliance with all state NPDES criteria, the current plant will be replaced by a new extended aeration plant, construction of which will be completed by October 1, 1985, at a cost of $1.2 million.

How about hazardous wastes at ORNL?

Nonradiological Solids

Nonradioactive solid wastes are categorized as "nonhazardous" or "hazardous." Each year about 19,000 tonnes of nonhazardous

Solid waste storage area No. 6, with White Oak Lake in the foreground. This area is currently being used for burial of low-level radioactive wastes.

waste and about 100 tonnes of hazardous wastes are generated at ORNL. The principal nonhazardous wastes are fossil fuel waste and construction material refuse. Other nonhazardous wastes, such as old tires, old batteries, paper products, and scrap metal, are sold. The remaining nonhazardous wastes—cafeteria and office waste, cooling tower sludge, fly ash, and coal-pile runoff—have been disposed of in landfills on-site.

The shallow water-table conditions at the Y-12 sanitary landfill in Bear Creek Valley have made it necessary to select a new landfill for disposal of nonradioactive and nonhazardous wastes. This landfill, now in operation, is located on Mt. Vernon Road near Y-12. It will accept wastes from Y-12, ORGDP, and ORNL. It will be given a permit by the State of Tennessee and will be operated in accordance with state requirements. Another landfill called the contractor's landfill, which is located about 1.6 km (1 mile) west of the ORNL central site, is an open pit into which nonsanitary, nonhazardous debris is placed and covered with fill.

Hazardous materials consist of four major groups of materials: asbestos, compressed-gas cylinders, chemicals, and waste oils. Such wastes may be placed in retrievable storage on the site or disposed of in other ways. Asbestos and animal carcasses (from biological research facilities) are placed in dedicated trenches in solid waste disposal area No. 6 and covered the same day. Hazardous wastes that also contain radioactivity are placed in aboveground retrievable storage on the site until a suitable treatment method or disposal site is available. Gas cylinders, nonradioactive chemicals, and oils contaminated with polychlorinated biphenyls (PCBs) are transported off the site to a licensed commercial hazardous waste facility for disposal. PCBs, which are very widespread in all environments containing large electrical equipment, have been detected in ORNL sediments but not in groundwater. This finding suggests that earlier gradual releases of PCBs occurred, probably from ground surface runoff with accumulation in the sediments.

How does ORNL benefit the human environment?

Socioeconomic Benefits

The primary operational product of ORNL is new scientific and technological information of national importance. The information produced by ORNL includes formal reports, publications in the scientific literature, assessments for governmental bodies (including congressional testimony), research results to be disseminated to national and international technical groups, consultations with industrial representatives to stimulate technology transfer, and patent disclosures pertaining to new technology.

Although ORNL produces and sells a few commodities (i.e., radioactive isotopes and special materials), its central function is to develop high-risk, high-payoff technology and to increase scientific knowledge. It is charged with identifying and providing solutions to generic problems of energy-based technologies; providing assistance to various governmental agencies; making special equipment available to various user groups (from universities, industries, and other research laboratories throughout the world); and giving universities access to major research facilities and programs. In addition, as a result of recently liberalized consulting and patent policies, ORNL has more freedom to stimulate the economy by lending its technical

How Safe Can It Be To Work at ORNL?

Any research facility that studies toxic chemicals, cancer-causing agents, and radioactive materials must be pretty dangerous for workers, right?

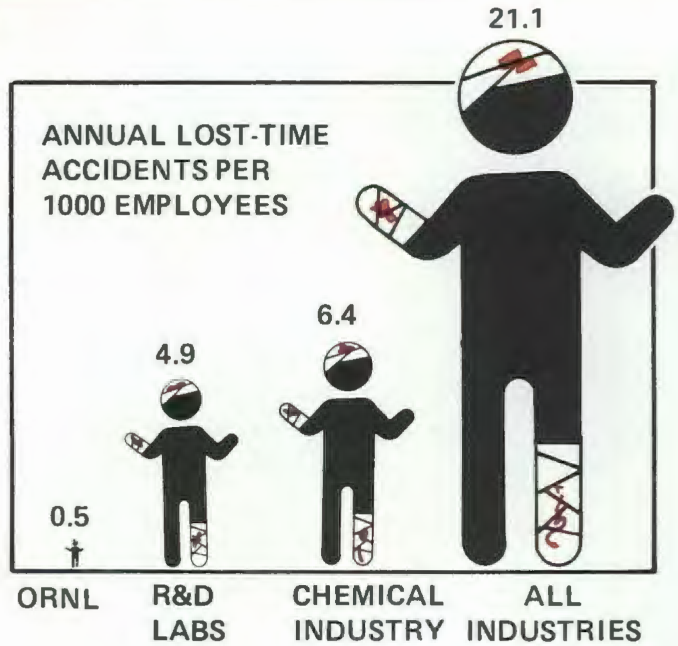
Wrong. ORNL is one of the safest places anyone could work. One of the safest chemical-related facilities. One of the safest research and development (R&D) centers. One of the safest workplaces of any kind. In fact, ORNL employees are about ten times as safe from disabling injuries at work as they are at home.

ORNL's safety performance has set many records and earned many awards:

- National Safety Council's highest annual award, the Award of Honor, which is won by only about the top 6% of the nation's safest employers. ORNL has won this award every year since 1975.
- Most employee-hours ever worked without a lost-workday accident—17 million—by any R&D facility reporting to the National Safety Council.
- Gold Safety Award from Union Carbide Corporation (the Laboratory's operating contractor until this spring), for operating 16 million employee-hours without a lost-workday accident. ORNL is the only single Carbide-run facility ever to win this award.
- DOE's Award of Excellence for holding the yearly number of lost workdays and medically restricted workdays to less than 1.1 per 100 full-time employees for five years in a row.
- Best 1983 safety ranking of any DOE national laboratory with more than 2500 employees.

According to Mike Knazovich, head of ORNL's Safety Department, safety has always been a high priority at the Laboratory. "From its beginnings, the Atomic Energy Commission [DOE's forerunner] put a strong emphasis on safety," says Knazovich, "much stronger than outside industry did. Outside industry's general philosophy in the past was to get the product out or the job done; accidents were often regarded as inevitable." That philosophy received a jolt in 1970, says Knazovich, with the creation of the Occupational Safety and Health Administration (OSHA), which set new standards aimed at improving workplace safety. "AEC already had a safety program as stringent as OSHA's," says Knazovich, "so it asked for—and received—an exemption from OSHA's jurisdiction. Then AEC simply adopted the OSHA standards as supplemental safety regulations." Despite more than a decade of safety improvements in industry since OSHA was formed, says Knazovich, "in the area of safety performance we still stand head and shoulders above our industrial counterparts."

Discussion of industrial safety often revolves around this question: Which is more important—safe working conditions or safe practices? "Regulations such as



OSHA's concentrate on safe work conditions," says Knazovich. "By now, though, most serious unsafe work conditions at ORNL have been taken care of." According to Knazovich, most injuries at ORNL these days happen when people fail to follow the right procedures or don't plan their work carefully.

Lapses in safety practices also contribute to injury-causing accidents away from work, which are ten times as likely as on-the-job injuries for ORNL employees. "Off-the-job injuries don't affect the Laboratory's occupational safety record," says Knazovich, "but they cause just as much pain and just as great a loss in productivity as on-the-job injuries do. Off-the-job injuries cost ORNL $500,000 last year for sick pay, insurance claims, replacement personnel, and other expenses." According to Knazovich, ORNL's Safety Department—like other industrial safety organizations—is now placing more emphasis on improving off-the-job safety.

What can safety specialists do to protect employees who aren't at work? "We can't control off-the-job conditions," says Knazovich, "so all we can do is focus on practices: We give advice and encouragement. While there may always be more off-the-job than occupational injuries, we hope to reduce the difference in rates."

The task could be made easier by a better understanding of why people are more accident-prone at home, according to Knazovich. A proposal has been developed to investigate the root causes of off-the-job injuries; the study would involve the Energy Division and Environmental and Occupational Safety Division staff. Says Knazovich, "If ORNL could develop an effective approach to reducing off-the-job injuries, it would be useful not just here, but nationwide."—*Jon Jefferson.*

Captain J. A. Fields of the ORNL Fire Department conducts a drill involving equipment and personnel from all three Oak Ridge facilities. A fire is simulated at the warehouse storage building in the 7000 area. Because ORNL has its own fire department, it does not depend on the community for fire-protection services.

expertise to industry. The Laboratory's researchers can more easily transfer the technologies they have developed to the private sector through consulting, and industries are now more willing to use federal facilities to develop new technology to the commercial stage because DOE is now waiving its patent rights and allowing contractors working at federal facilities to retain rights to their inventions. These incentives should allow ORNL to help stimulate development of high-technology industries in the Technology Corridor of East Tennessee and elsewhere.

The existence of ORNL has a strong impact on the Oak Ridge community and East Tennessee area. Each year ORNL's facilities and research projects attract hundreds of foreign scientists, who give the community an academic and cosmopolitan flavor. Because of their strong academic background, ORNL employees promote high quality in public education, as seen in the schools of Oak Ridge and West Knoxville. ORNL employees tend to become more involved than nonemployees in community affairs and cultural organizations, such as the Civic Music Association, Civic Ballet Association, Community Playhouse, and Community Art Center of Oak Ridge.

ORNL's impact on the local economy is very positive. The direct effect of ORNL's payroll and procurement expenditures during 1981 was the support of 5600 local jobs and the creation of $139 million of local income. In the local region, the ORNL payroll accounts for most of the direct effect (4900 jobs and $128 million in income in 1981). The secondary effects of payroll and procurement are estimated to be an additional 4800 jobs and $74 million in income for the local region, giving a total local impact of 10,400 jobs and $212.7 million in income. Nearly three-quarters of ORNL equipment and supplies is procured outside of the local region. This procurement and other effects outside the local region bring the total economic impact of ORNL in the United States to an estimated 16,000 jobs and $301 million in personal income (based on 1981 activity). In sum, each ORNL employee supports an additional 0.75 unit of employment—that is, for every four ORNL employees, three other people locally or elsewhere have jobs also.

ORNL and its employees are assets to the local community in yet another way: They impose little burden on local services. Although difficult to quantify, the public service burden of ORNL families on community services such as correctional institutions, indigent care, and public mental health services is below national and regional averages because of the relative affluence of ORNL employees and because of their third-party insurance coverage. ORNL itself presents little impact on community services because it provides its own fire and safety protection and uses water provided to the city by DOE.

What is the bottom line?

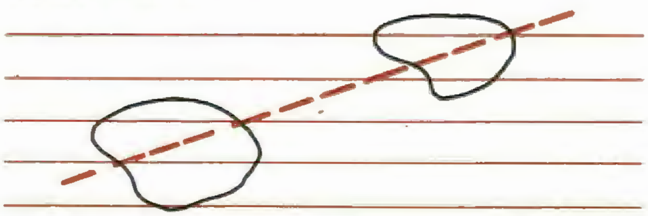
In order of significance, then, ORNL's environmental impacts may be summarized as (1) primary and secondary R&D effects of long-range significance, (2) continuing major socioeconomic effects on the regional community, and (3) small but identifiable effects associated with the dispersion of small amounts of materials, some of which are hazardous, to the environment. Thus, ORNL is a national resource that has a major impact—positive in terms of scientific and technology contributions, negative in terms of the potential for disruption of the natural environment. Over the decades of its operation, the negative impacts of ORNL are far outweighed by recognized present and future benefits. ornl

take a number

BY V. R. R. UPPULURI

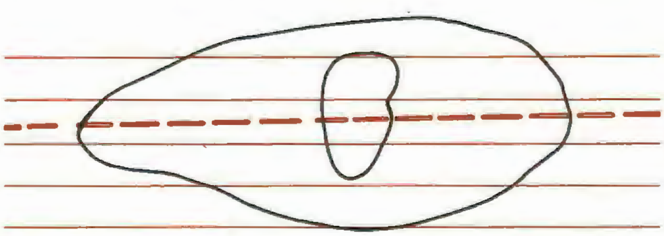
The Pancake Problem

A theorem dealing with two pancakes states that the area of two plane pancakes of arbitrary shape can be simultaneously bisected by a straight-line cut of the knife, as shown below.



This theorem is general because the two pancakes can have any arbitrary shape. The price one pays for this generality is that the theorem has only an existential nature: It tells that there exists a linear knife-cut that bisects the pancakes; it does not explain how to find the exact bisecting cut. The theorem is therefore a part of elementary real-variable theory, or elementary topology.

This theorem has a number of special cases and generalizations. For example, the two pancakes may overlap one another, and a bisecting knife-cut can still divide them into two equal parts, as seen below.



If this theorem is extended to three-dimensional shapes, it is referred to as the "ham sandwich" theorem. It can be applied to the case of two persons who wish to split a sandwich formed from a piece of white bread, a piece of dark bread, and a slice of ham. This theorem states that a planar slice of a knife can bisect the volume of the sandwich so that the two persons can eat equal shares.

Unfortunately, this theorem cannot be extended to three pancakes arbitrarily arranged on a platter—that is, one cannot divide three pancakes into two equal shares with a knife-cut. More details on this problem may be found in the book *The Mathematical Experience* by P. J. Davis and R. Hersh (Birkhauser 1981).

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Fun with Factorials

The product of the first n natural numbers is called the factorial of n and is denoted by $n!$ For example, $4! = 1 \times 2 \times 3 \times 4 = 24$, and $5! = 120$. Zero factorial is defined to be equal to 1.

The number 145 has the interesting property that $145 = 1! + 4! + 5!$ (i.e., $1 + 24 + 120$, respectively). Are there any other natural numbers equal to the sum of the factorials of their digits? It can be verified that 1, 2, and 40,585 are the only other natural numbers with this property.

David Church, a native of Gorham, New Hampshire, is an associate professor of physics at Texas A&M University. After receiving a Ph.D. degree in physics from the University of Washington, he worked as a postdoctoral research associate at the universities of Bonn and Mainz in West Germany and at the University of Arizona. He then served as a staff physicist at Lawrence Berkeley Laboratory. His current research interests include collisions and spectroscopy of ions stored in ion traps and coherence and polarization spectroscopy on fast ions. He has collaborated in research at ORNL since 1980. Ivan Sellin, a native of Everett, Washington, was recently appointed Distinguished Service Professor at the University of Tennessee. A Harvard College alumnus who earned his M.S. and Ph.D. degrees in physics from the University of Chicago, Sellin served two years as assistant professor at New York University before coming to ORNL in 1967 as a full-time staff member for three years. Since he joined UT in 1970, he has retained a part-time position at ORNL. In 1977 he received a Senior Fulbright Award and the Alexander von Humboldt Senior U.S. Scientist Award from the Federal Republic of Germany. In late 1983 he received the Jesse W. Beams Medal from the Southeastern Section of the American Physical Society (APS); this award is given annually for "significant and meritorious original research in physics" performed by an APS member residing in one of the ten southeastern states. In addition to his research with Professor Church in charged-particle trapping, Sellin has worked in the areas of beam foil spectroscopy, ion-atom and ion-molecule collisions, and electron spectroscopy. Here Church explains how ions are confined between the horns of a saddle-shaped electromagnetic trap; his listeners are Sellin and a UT graduate student, Teresa Underwood.

BUILDING A BETTER ION TRAP:

Atomic Physicists Study Recoil Ions

By DAVID CHURCH AND IVAN SELLIN

A new technique developed at ORNL under the leadership of two university professors traps ions of very low energies. This "recoil ion storage" technique permits studies of the transfer of electrons from atoms during collisions with multicharged, low-energy ions and opens the way to future precision spectroscopy experiments on such ions.

Multicharged ions are a familiar subject of study for both nuclear physicists and atomic physicists at Oak Ridge National Laboratory. Such ions, which start out as neutral atoms, are usually stripped of several electrons before emerging with millions of electron volts of energy from heavy-particle accelerators such as the Oak Ridge Isochronous Cyclotron (ORIC), the EN Tandem Van de Graaff accelerator, and the Holifield Heavy Ion Research Facility. Nuclear physicists use these positively charged ions to study phenomena affecting or caused by particle interactions with the nuclei of atoms (see Joe McGrory's "Heavy-Ion Physics," ORNL *Review*, Summer 1979). In nuclear

Texas A&M professor Robert Kenefick looks on as Sellin adjusts the thin target that strips many electrons off projectile ions just prior to their entry into the Penning ion trap structure.

physics, heavy multicharged ions are often used as atomic artillery to attack the nuclear fortress and expose the inner workings of the nucleus.

As atomic physicists, on the other hand, we focus on the electrons orbiting around the nucleus rather than on the nucleus itself. Together with our colleagues, we specialize in the dynamics of electrons involved in ion-atom collisions, measuring the cross sections (effective target areas) associated with atomic collisions, or studying the energy spectra of photons or electrons that are emitted following such collisions. Many of these atomic physics studies center on high-energy ions that, after emerging from an accelerator, have been stripped to higher charge states; in other words, the ions have had additional electrons removed by subsequent passage through a thin foil.

However, in many natural phenomena low-energy, highly charged ions play a dominant role. Examples include many astrophysical processes such as those occurring in the solar corona and heavy-particle interactions occurring in the upper atmosphere. Low-energy ions also are found as contaminants on test probes in the plasma fuel of experimental tokamak fusion reactors, such as the devices operated by or planned for ORNL. Thus, much incentive exists to study these processes. Recently at ORNL's Physics Division, thanks to the efforts of David Crandall (now with the Office of Fusion Energy of the U.S. Department of Energy) and Ron Phaneuf, multicharged ion beams have been made available at significantly lower energies than those produced by the large accelerators.

But is it possible to produce ions at *really* low energies—electron volts or below—instead of the thousands or millions of electron volts that ions in beams usually have? Ion beams slowed to low energies lose their definition, and

the "orbiting" collisions of these ions with atoms make efficient ion detection difficult. Therefore, new techniques are needed for producing and detecting ions at very low energies. Now, a new technique called recoil ion storage has been developed to trap low-energy ions produced during fast-ion collisions with atoms. The story of this development is a tale of a collaboration that includes scientists from Oak Ridge, the University of Tennessee, and Texas A&M University.

A Key Observation

While studying the X rays emitted in fast ion–atom collisions, Ivan Sellin of the Physics Division and the University of Tennessee noted that the spectral lines (or characteristic energies) of some of the X rays from multicharged ions did not exhibit appreciable Doppler broadening. The Doppler effect is basically a shift in detected X-ray energy caused by the motion of the source of X rays relative to the detectors. In sound waves, we experience the Doppler effect as a change in pitch, for example, in the sound of a race car as it passes us at high speed.

Sellin's observation indicated that many of the X rays came from *slow* multicharged ions, which were produced when fast ions collided with atoms. For example, fast ions of chlorine in collisions with neon gas atoms would produce some very

University of Tennessee research assistant professor Chun-Sing O adjusts tuned circuit elements coupled to a new radiofrequency trap housed in the large vertical pipe beneath his right hand. The ion trap and vacuum components shown were custom fabricated at UT and are now installed at ORNL.

slow neon ions. Sellin immediately realized that high-precision spectroscopy would be possible with these recoil ions if they could be observed for relatively long times. His calculations showed that the recoil ion energies might be as low as a few electron volts. But a problem remained. A method was needed to use the ions' low energies, coupled with high positive charges, to achieve high precision in either spectroscopy or collision measurements that could test the basic theories of physics. Such a method was not apparent. Thus, while it had been possible to determine the probability of electron transfer between an atom and an ion with a single positive charge, such measurements had not been possible for collisions of highly charged ions at electron volt energies.

Ion Confinement

At Texas A&M University, David Church had initiated a program to study low-energy ions in ultrahigh vacuum by confining them in a device called an ion trap. Church and his postdoctoral associate Mike Holszcheiter were using a Penning trap, named after a scientist who did early work with related devices. This trap was later refined in Scotland, Germany, and the University of Washington. Penning confinement uses a strong uniform magnetic field, which causes ions to spiral in "cyclotron orbits." The ions can still move freely parallel to the direction of the field, so an electric potential is used to contain this component of the motion. The electric potential is called a saddle potential, because the "horns" of the saddle hold in the ions in one direction but permit them to "fall off the sides," like an equestrian saddle. However, the cyclotron orbits about the magnetic field inhibit this sideways instability, allowing the ions to be held in space near the top of the saddle. In fact, ions with essentially no energy can balance at the center of the saddle.

The metal electrodes used to produce the saddle potential ideally follow the shape of geometric figures called hyperboloids of revolution. Our applications require deviations from this ideal shape but still permit stable confinement of ions that have energies of electron volts or below.

Church had planned to study the collisions of low-energy, multicharged ions with atoms because of the intense theoretical interest in these collisions. Extensive worldwide efforts over the last several years have led to theoretical predictions of the probability of transfer of an electron from atomic hydrogen to a variety of multicharged ions at electron-volt energies. This cross-section problem has been worked on, for example, by Alexander Dalgarno, a previous member of the Physics Division's Advisory Committee, at the Harvard Smithsonian Astrophysical Observatory.

Unfortunately, at the lowest energies, the collisions seem to exhibit no simple dependence on the ion charge—that is, no simple relationship has been found between the charge state of the ions and the probability of electron transfer from the hydrogen atoms to the ions. However, Chris Bottcher of the Physics Division and his collaborators—often associates of Dalgarno—have been able to extend some of these calculations of electron transfer probability to higher energies of collision between ions and atomic hydrogen. So far no target more complex than atomic hydrogen has been treated theoretically at an appropriate level of exactitude.

Church's goal was to conduct experiments to test the low-energy theories. He and Holzscheiter had planned to produce multicharged ions in the Penning trap by knocking electrons off target atoms with a beam of higher energy electrons. However, the cross sections for these collisions decreased rapidly as more electrons were removed from the atom; in other words, the probability that the electrons would highly ionize atoms declined as the target ions achieved higher charge

states. Church and Holzscheiter could easily produce useful numbers of ions in their Penning trap, but these ions had positive charges of only 2 or 3. Church wondered if more ions of a higher positive charge could be produced. He had heard of Sellin's recoil ions but had the impression that their energies would be too high to permit effective confinement in an ion trap. He began to reconcile himself to the study of ions that have low charge states.

Conferring and Collaborating

As is often the case, further progress came about at a physics conference. In 1979 at the Gordon Conference on Atomic Physics at Wolfeboro, New Hampshire, Church happened to mention his ion trapping work to Sellin. A few weeks later Sellin called to tell Church of his energy calculations for the recoil ions and to suggest a collaboration in trying to trap them. Just then, Lew Cocke of Kan-

Church and Chun-Sing O tune trap electronics on a radiofrequency trap fabricated at UT and installed at ORNL. The ion trap is housed in the large, cylindrical, vertical pipe below Church's right hand.

sas State University published some measurements that set upper limits on recoil ion energies. Following more calculations, Sellin and Stuart Elston from Oak Ridge met Robert Kenefick (also of Texas A&M) and Church at the annual meeting of the American Physics Society's Division of Electron and Atomic Physics in Houston. Everyone agreed to the details of a collaboration.

At Texas A&M Church and Kenefick would design and build a special Penning trap and vacuum system to bring to Oak Ridge for this work. Sellin, Elston, and Sven Huldt, a Swedish postdoctoral associate, would assemble an ultrahigh-vacuum beam line on the EN Tandem Van de Graaff accelerator. Excellent vacuum is necessary to maintain the high charge state of the ions; otherwise, the highly charged ions are likely to grab electrons in collisions with nearby gas atoms before definitive measurements are possible. The goal of the first experiments would be to study collisions of highly charged ions with atoms, because no measurements of electron transfer from atoms to ions lacking more than three electrons had been performed at electron volt energies.

Of course, not much physics can be done without research funding. The first move was to secure it. The work at Texas A&M was given support by the Texas A&M Center for Energy and Mineral Resources, and travel to Oak Ridge was funded by Oak Ridge Associated Universities. Sellin and Elston secured a seed money grant from ORNL.

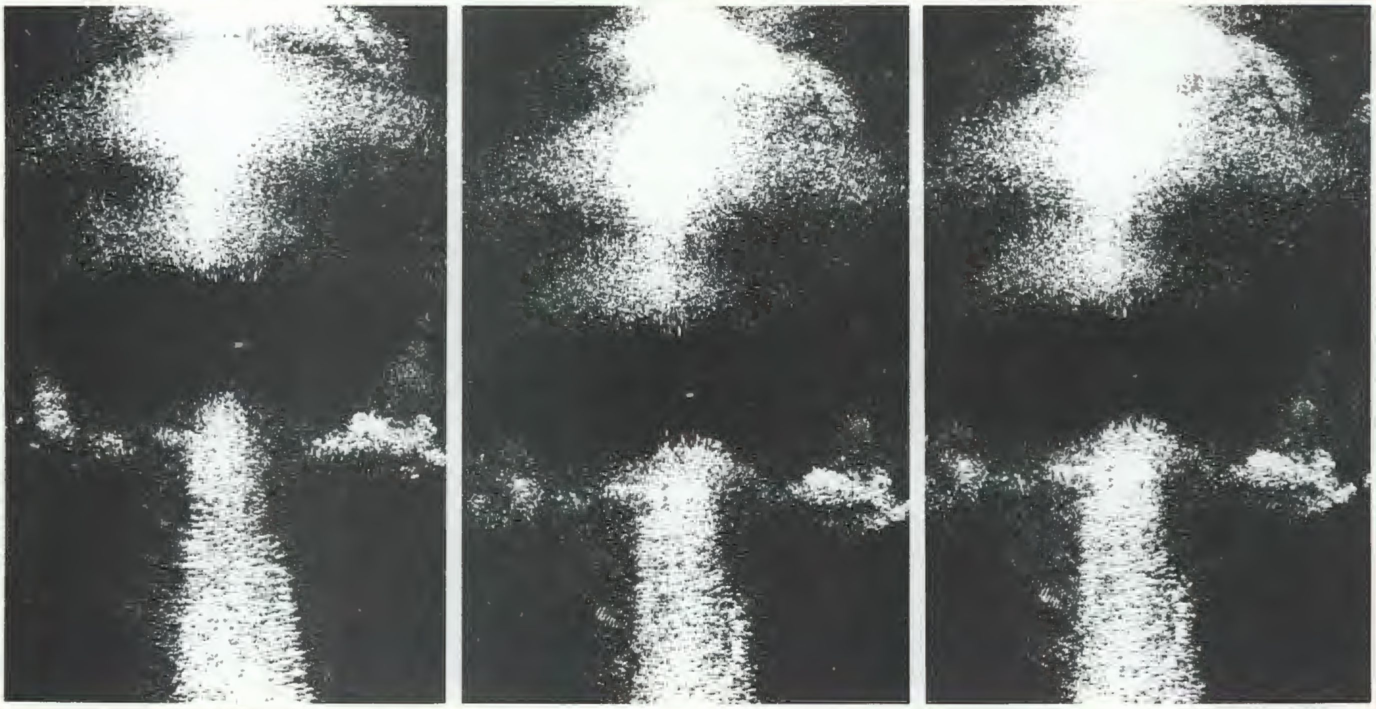
Ions on Radio

For electron transfer measurements, ion confinement alone is not enough; the ions have to be identified. Of course, if a particular target gas is struck by a fast beam of ions, multicharged recoil ions of the target atoms will certainly be produced. If neon, for example, is struck by chlorine ions, multicharged neon recoil ions result. To study a particular ion type, however, one must know its charge. The number of ions having this charge must be measured, and only those ions should be trapped for studies of charge-transfer collisions in which the ions later change their charge through collisions with target atoms. Also, to make the experiment work, stray ions of other elements must be expelled.

The Penning trap is well suited to these requirements. The motion of the ions in the saddle potential and uniform magnetic field is harmonic, i.e., periodic like the motion of a pendulum. In addition, just as the oscillation frequency of a pendulum is determined by the length of the supporting rod and the local acceleration of gravity, the frequencies at which the ions oscillate in the trap are determined by their charge, mass, and the strengths of the electric potential and magnetic field in which they move. An ion of a specific charge and mass actually has three characteristic frequencies: the axial oscillation between the horns of the saddle potential, the cyclotron oscillation about the magnetic field, and a rotation at lower frequency called the magnetron oscillation. For typical conditions, all of these frequencies lie in the radiofrequency range. In other words, it is possible to listen to confined ions with a radio.

An AM radio station transmits energy as a wave that has a fairly well-defined frequency. The voices and music that we hear are the result of controlled changes in the transmitted signal strength, called modulations. When an antenna is placed near the ion trap and a sensitive marine radio is tuned to one of the ion oscillation frequencies, the ions announce their presence in the received radio noise. This noise modulation is produced by random fluctuations of the amplitude of motion of the confined ions. The noise grows louder if more ions are trapped or if the average ion energy is increased.

We can also force coherent modulations in which the ions oscillate back and forth together in phase. A weak radiofrequency electric potential is established by placing an external oscillator across the Penning trap electrodes. If the depth of the saddle potential is reduced at a constant rate with time, each ion axial oscillation frequency successively passes through "resonance" with the applied frequency. At resonance, the ions absorb energy from the radiofrequency potential, which produces

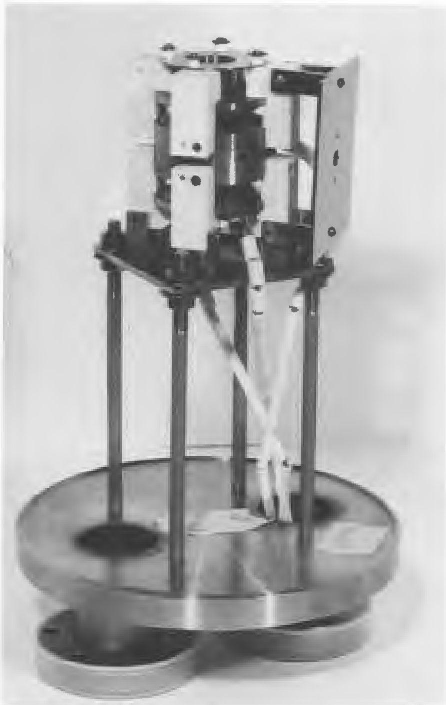


Above are sequential photographs of three ions, two ions, and even one ion of barium stored in a small Penning trap. The photographs were taken by Peter E. Toschek and colleagues at the universities of Heidelberg and Hamburg in the Federal Republic of Germany. The luminous, pointlike ion clouds of diminishing radius near the center of each picture were photographed by exposing film to fluorescent light. The light was emitted by the ions following repetitive resonant excitation of the barium by microscope-magnified laser light. The clouds diminish in size as this three-two-one sequence is realized because a single ion is not compelled to avoid nearby orbiting neighbors through the repulsion of like charges (Coulomb's law). It therefore can occupy a nearly stationary position very close to the trap center. The ~2-μm diameter of the smallest cloud is limited by diffraction effects in the microscope used to magnify the image.

Trapping and Photographing a Single Ion

Research at ORNL has been concerned with many multiply-charged ions. Elsewhere in the United States and Europe, however, researchers have succeeded in trapping and photographing a *single, charge one ion*. Peter E. Toschek of the University of Hamburg, Federal Republic of Germany, recently led experiments at the University of Heidelberg that confined a single barium ion within an electrodynamic quadrupole trap. Scientists at the National Bureau of Standards in Boulder, Colorado, have recently confined single magnesium ions in an electromagnetic trap.

The scientific value of trapping ions was recently discussed in an unpublished paper by Toschek entitled "Localization and Observation of Single Atomic Particles." In his conclusion, he wrote: "Sensitive techniques for single-particle detection are well known from standard methods of nuclear and high-energy physics, but the trace or count they observe is something like the particle's transient shadow—the belated hint to its existence. Unlike those particles, a trapped ion is not lost in the process of detection. It remains fully manipulatable, and what we see when viewing it is light scattered off this entity [just as we see] light scattered by macroscopic bodies." Toschek suggests that this technique offers the most irrefutable proof of the existence of atoms, which serious critics of atomism insisted was missing from Aristotle's time up to that of the natural philosopher Ernst Mach.



View of an assembled radiofrequency ion trap. White insulating blocks are shown positioning the central ring and end cap electrodes that constitute the heart of the trap structure. Holes in the ring electrode such as the one seen near the gaps between the insulating blocks serve to admit projectile ions, which create slow, highly charged, recoil ions near the center of the trap, and beams of laser light for subsequent recoil ion excitation. Laser light represents one of several means of changing the internal energies of stored recoil ions near the center of the structure.

a strong, narrow modulation. Each modulation signal is characteristic of a definite number of ions that possess a particular mass and charge. By monitoring these resonance signals after ions have been produced in the trap and confined for a selected time interval, we can determine how the number of highly charged ions in different charge states depends on storage time. The length of time that a certain percentage of ions resist changing their charge state is related to the probability of electron transfer from gas atoms to the ions.

At Texas A&M, preparations of the ion trap, ion detection electronics, and vacuum system were finally complete. In January 1981 a one-week run on the EN Tandem accelerator at ORNL was scheduled. Church and graduate student Bill Burns drove the 1000 miles to Oak Ridge in a station wagon loaded with apparatus. There they were joined by Bob Kenefick and undergraduate Russ Doerner. The major components of the beam line had been assembled at Oak Ridge by Sven Huldt, Elston, and Sellin but were not yet installed. The scientists then put in several 16-hour days and, with the assistance of riggers, pipe fitters, and machinists, installed the beam line and ion trap.

Confining Recoils

We decided to use a 35-MeV beam of chlorine ions, stripped of ten electrons, as the "pump" ions to produce the low-energy recoils. Neon atoms were used as the target gas. Slow multicharged ions of neon were produced from the collisions of the chlorine ions with the neon atoms. Would the numbers of neon recoil ions and their energies agree with the results of the calculations? Would the recoil ions be confined and detected as expected? Everyone was excited (and relieved) when the answers from the apparatus came back "yes."

Of course, everything did not work perfectly the first time, but subsequent efforts resolved the outstanding problems. Apparatus sensitivity improvements permitted the detection of trapped neon recoil ions that had been stripped of eight electrons, the maximum expected with the chosen "pump" beam.

The initial elation at storing the recoil ions helped sustain all concerned in the long hours of studying the collisions by which the ion charge is changed. Joined by other scientists, the team found that highly charged neon ions could be held in the trap for times up to a second. As expected, collisions of the neon ions with neon target gas atoms eventually led to a loss of ions. When an electron is transferred from an atom to a multicharged ion, both the original atom and the ion have new positive charges. Because the two are close together during electron transfer and because charges of like sign repel (Coulomb's Law), the ion pair flies apart at a high velocity, escaping the trapping fields. The same collision process occurs when the recoil ions are formed, but the fast pump ions move away so quickly that the recoil ions do not gain much velocity and remain confined. On such small differences are successful experiments designed.

From the start, both of us had been interested in the possibilities of doing precision spectroscopy with stored recoil ions—that is, making precise measurements of the electron energy levels of highly charged ions. This area is now occupying much of our attention. In such measurements, the energy levels of the electrons of an ion are measured by using oscillating electric and magnetic fields. As explained by the Time-Energy version of Heisenberg's Uncertainty Principle, the inherent precision of a frequency measurement is directly related to the time available to perform it; thus, the longer the ions are available for study, the more precise the measurements of the ions' electron energy levels. If ion storage times as long as 1 second are used, a characteristic ion frequency of 10 billion Hertz (cycles per second) may be measured to a precision of 1 Hertz. Such precise measurements have already been achieved on certain trapped singly charged ions but not on highly charged ions. Now, we foresee a

Sellin arranges the precise positions of low-noise signal cables at their point of entry into an ultrahigh vacuum chamber located on an EN Tandem accelerator beam line at ORNL.

practical opportunity to make precision measurements in a trap to determine the electron energy levels of highly charged ions to an unprecedented degree.

Radiofrequency Trap

We have already mentioned that the relatively low energy of the stored recoil ions reduces Doppler broadening effects, which tend to decrease spectroscopic precision. However, it is also desirable to store the ions without a magnetic field, because the field changes the internal ion energies. Control of the field strength in space and time to very high precision is possible but difficult. Encouraged by our results with the Penning trap, we are about to attempt recoil ion confinement in a "radiofrequency" (rf) trap, which requires no magnetic field. This use of radio fields to trap ions is a dynamic containment method, again based on the saddle potential. By inverting the saddle potential at a high frequency, ions falling off the saddle at one instant will be pushed toward the center of the saddle at a later instant. Imagine attempting to balance a baseball bat vertically on your finger. If you hold your hand still, the bat will fall over. But if you move your hand rapidly to correct for the tendency of the bat to start to fall in a given direction, balance can be sustained (indefinitely if you practice). The rf ion trap maintains the stability of the ions at low energy by an analogous but well-defined change in the sign of the saddle potential with time. Although this trapping method is elegant, it is relatively difficult, and that is one reason we did not attempt it first.

The success achieved with the simpler though more spatially confined Penning trap made the design, construction, testing, and use of a more accessible rf trap feasible. At this point, we thought it essential to attract a scientist who had expertise and interest in the use of rf traps and was able to make a full-time commitment to developing these more sophisticated devices in our laboratory. Chun-Sing O, who had acquired his graduate training in atomic physics at New York University and had had postdoctoral training and experience with rf traps at Texas A&M University, joined our collaborative effort at ORNL about one-and-a-half years ago. He now plays a leadership role in this next phase of our experimental ion-trapping effort.

Many forces exist that tend to pull apart established research collaborations. Perhaps one of the chief of these is the possibility that some members of the group will become vitally interested in some related area of research. We expected that the challenge presented by our planned precision spectroscopy efforts, as well as the diversity of the experimental skills and techniques required, will tend to strengthen the collaboration as well as broaden everyone's experience. Great opportunities are presented by confining highly charged ions of low energy. From the interest expressed in this work by physicists the world over, we expect that the future of this area will be an active one.

awards and appointments

On December 13, 1983, Martin Marietta Energy Systems, Inc., was named as the new contractor to replace the Nuclear Division of Union Carbide Corporation on April 1. The following promotions and management changes went into effect April 1: **Herman Postma**, ORNL director, has been named vice-president of Martin Marietta Energy Systems; **Raymond Wiltshire** of Martin Marietta in Denver, Colorado, has been named executive director for Support and Services, replacing Kenneth W. Sommerfeld, who has been appointed vice-president for Enrichment; **Fred R. Mynatt**, director of ORNL's Instrumentation and Controls Division, has been promoted to associate director for Nuclear and Engineering Technologies, replacing **Donald B. Trauger**, who has been named senior staff assistant to Postma. Two former ORNL employees have also been promoted: Clyde Hopkins has been named senior vice-president; and Gordon Fee, Y-12 Plant manager, has been named vice-president of Martin Marietta Energy Systems.

Marvin L. Poutsma has been named director of the Chemistry Division, replacing **O. Lewin Keller**, who has been appointed director of the Transuranium Research Laboratory.

Ivan Sellin has received the Jesse W. Beams Award for 1983 from the Southeast Section of the American Physical Society. The award was given for "significant and meritorious original research in physics."

John B. Storer, M.D., and **Richard F. Wood** have been designated Corporate Research Fellows of Union Carbide Corporation.

Robert W. Roussin has been appointed director of the Radiation Shielding Information Center.

Robert M. Nicklow has been named head of the Crystal Physics Section of the Solid State Division.

Bennie J. McNabb has received the 1983 Engineering Associate Achievement Award of the American Society for Metals.

Stanley K. Borowski has received the Mark Mills Award from the American Nuclear Society for his paper "RF-Assisted Current Startup in the Fusion Engineering Device." The $500 award is given annually for the best original scientific paper by a graduate student in the area of nuclear science and engineering.

Leon N. Klatt has been elected secretary of the Analytical Chemistry Division of the American Chemical Society.

Helga Gerstner has been named technical assistant to Chester R. Richmond, associate director for Biomedical and Environmental Sciences. **Tim Ensminger** has replaced her as director of the Information Center Complex in ORNL's Information Division. **Helen Pfuderer** has been named acting director of the Hazardous Materials Information Center.

Harold L. Adair has been elected vice-president of the International Nuclear Target Development Society.

Leroy Stratton has been named manager of ORNL's Low-Level Radioactive Waste Research and Development Program.

Jim Bentley has been elected to the board of review of *Metallurgical Transactions*.

Hugh R. Brashear has been named head of the Research Instrument Section of the Instrumentation and Controls Division.

L. Dean Eyman has been appointed manager of the lead office (located at ORNL) of DOE's new Hazardous Chemical Defense Waste Program. **Norman Cutshall** has been named head of the Earth Sciences Section of the Environmental Sciences Division. **Carl Gehrs** has been appointed head of the division's Aquatic Ecology Section, and **Steven G. Hildebrand** has been named head of the division's new Environmental Analyses Section.

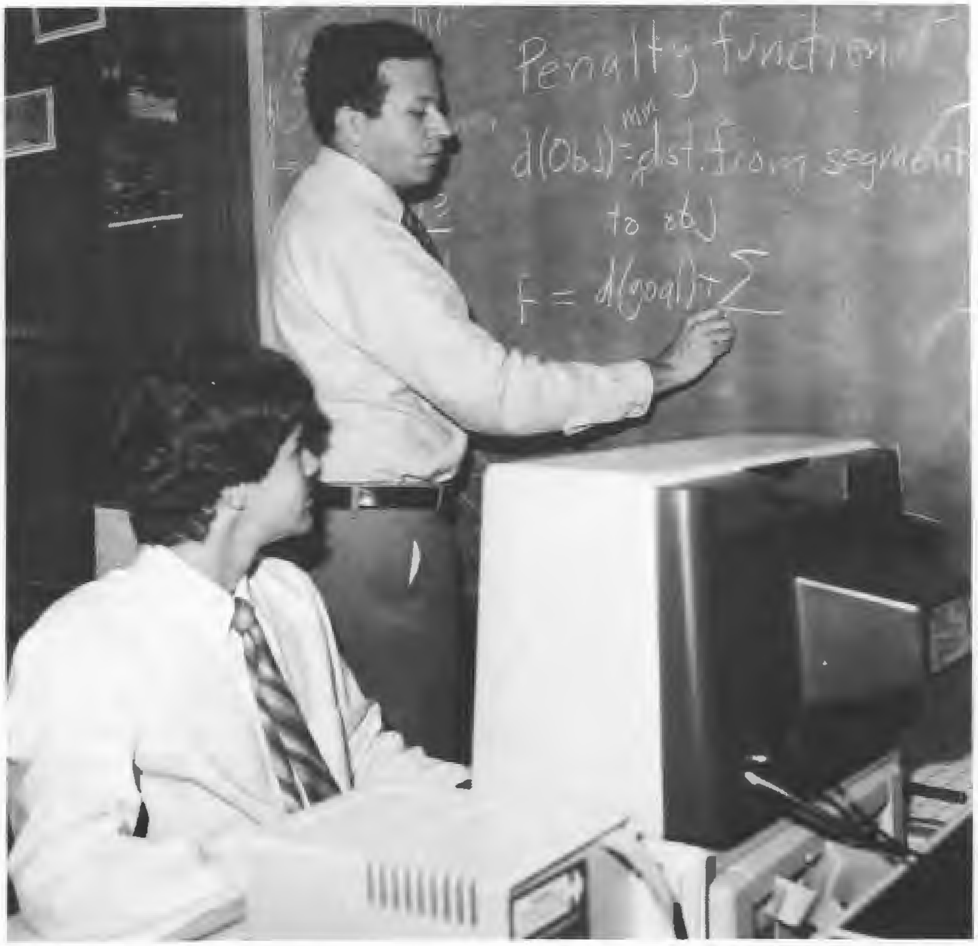
Everett E. Bloom has been named a Fellow of the American Society for Metals.

Joseph A. Setaro has been named head of the Technical Section of the Operations Division.

R. G. Donnelly, **J. I. Federer**, **K. H. Galloway**, **R. L. Heestand**, **Henry Inouye**, **C. T. Liu**, and **A. C. Schaffhauser** each received a citation and a photograph for their work on the *Voyager* mission.

J. H. Smith has been appointed to serve as the Board of Directors representative to the Section Management and Membership Committee of the American Society for Nondestructive Testing.

D. W. Yarbrough has been selected as chairman of the 19th International Thermal Conductivity Conference, and **David L. McElroy** has been elected to serve a second term as chairman of the Governing Board of this conference, which will be held in 1985 at Tennessee Technological University.



Alan Solomon is currently head of the Mathematics Group of the Mathematics and Statistics Research Section of ORNL's Engineering Physics and Mathematics Division. Prior to February 1, 1984, he was head of the Mathematics Section in the Mathematics and Statistics Research Department of the Computer Sciences Division of Union Carbide Corporation's Nuclear Division. He received his Ph.D. degree from New York University in 1963. After serving for four years on the faculty of New York University, he held positions at Tel Aviv University and at Ben Gurion University of the Negev before coming to Oak Ridge in 1976. His principal areas of research include moving-boundary problems of partial differential equations, melting and solidification modeling, and latent heat thermal energy storage. His interests in artificial intelligence and robotics are concerned with the identification and understanding of basic problems dealing with recognition and focusing. Here, Solomon plans a robotic obstacle-avoidance program as Gary Hall, a Millsaps College student, looks on.

The Mathematics of Artificial Intelligence

By ALAN D. SOLOMON

The term *artificial intelligence* (AI) is used increasingly by newspapers, magazines, and television. Like most novel terms such as *laser, modern art,* or *hard rock,* AI's full meaning is not well understood by most of us, although its mention in parties or get-togethers is met with wise nods and followed by interesting conversation. I will try to describe what AI is from the point of view of a mathematician who finds it to be an interesting topic and who believes that it will have a profound effect on all of us.

My remarks will address three questions: What is AI? What are its problems? How does mathematics enter into it? I will also describe a specific mathematical problem from this field and some of our efforts at Oak Ridge National Laboratory in working with it. Finally, I will explain how our AI work relates to ORNL's new efforts in developing robots for energy-related applications.

What is AI?

AI is a research field in which we seek ways to have machines perform tasks "intelligently." The term *machine* is defined by the context of the tasks being performed. If these are "cognitive" or "thinking" tasks, *machine* refers to a computer. If they are physical tasks, it means a machine that moves other objects or material as a robot or a carburetor can. If the task involves thought and action, then it consists of a computer and physical device, linked together in some way.

Intelligence is usually understood in the human context. In this setting, it is synonymous with such terms as *flexible, adept, wise,* and *foreseeing.* Intelligent tasks include playing chess, proving mathematical theorems, repairing cars, get-

ting along with people, remembering names and faces, and speaking English or French.

An outstanding example of intelligence is furnished by an old "I Love Lucy" television episode. In it, Lucy is placing chocolate bonbons in paper boxes, 12 per box, as they arrive on a moving belt. Her boss observes that she is doing well and speeds up the belt. Because Lucy cannot box the bonbons as fast as they are now arriving, she begins to eat 1 of every 13. Naturally, the line speeds up, and she copes with her problem by eating 2 of every 14—and so on.

Mathematicians at ORNL are applying the principles of artificial intelligence to energy-related problems. Their goals include designing an economical, energy-efficient solar house and programming a robot to avoid obstacles so that it can operate in a hazardous environment, such as a nuclear reprocessing plant.

Lucy learned a skill (boxing bonbons) and developed a strategy for dealing with a new situation (speedup of the belt)—namely, eating the extra candy. This combination of learning and developing strategies for dealing with novel situations is a hallmark of AI efforts.

Intelligent Machines

The goal of creating intelligent machines has occupied mankind since the dawn of civilization. The Egyptians are believed to have built water clocks and robotic figures 5000 years ago. Extremely elaborate mechanical figures, capable of ringing bells or drawing pictures, captivated Europeans as early as 1500. The 18th century Jacquard loom was the first of the "program-controlled" devices, in which the machine performed a task according to directions that could be altered without physically changing the machine itself. Similarly, during the Dark Ages in Europe, "feedback control" devices (i.e., devices like the float in a toilet water tank, which shuts off the water inlet when the float reaches a desired level) were developed to respond to changes in their environment. The 20th century development of electronic digital computers has sparked great interest and has resulted in some real achievements, making it possible for machines to play board games such as chess and checkers, prove mathematical theorems, and diagnose diseases.

Energy-related problems that AI can address fall into roughly five categories: (1) control of large complex systems, (2) data management and information, (3) systems for control of large computer codes, (4) systems that control intelligent autonomous devices, and (5) systems to control self-reproducing devices.

An example of the first is a mechanism for control and emergency handling of systems such as a power plant or the traffic flow in a large urban street network. The second, data management and information, involves the development of a system for answering questions about the state of knowledge in a field or the availability and pricing of resources.

An example of a system for control of a computer code is another code, whose purposes would include selecting what output to give at a specific time, drawing qualitative inferences from computed results, and reducing computer time. Immediate applications of such codes could be made to heat and mass transfer codes, nuclear reactor simulations, and resource planning programs.

An ideal autonomous machine is a robot with an onboard computer, capable of carrying out tasks in a hazardous environment in which unexpected things might occur (as in the "Lucy" example). One self-reproducing machine could be a factory capable of producing a "baby" factory, which would then build itself up to full, operational size and possibly become even more sophisticated than its parent.

Each of these categories involves intelligence, manifested by such capabilities as forming qualitative opinions, making decisions, formulating a plan of action, focusing on what is important, and manipulating what is known.

Enter Sam

For example, consider a computer code inference system called SAM (Simulation Analysis Module) that we are currently developing to simulate the heating and cooling of a solar house and the costs involved. This simulation, in support of the Thermal Energy Storage Program of ORNL's Engineering Technology Division, will provide information such as the amount of money that can be saved over several months by altering window sizes, wall size and construction, and placement and choice of special materials. The program also can give the temperature in the walls, at their surfaces, and in the room at all times. However, if we wish to print this information on paper, a single week's simulation will result in a stack of paper 0.3 m (1 ft) high. Though after a while most of this information is fairly predictable, occasionally a difference in behavior of, say, the surface temperature of a wall will be noted. Hence, a key aim of SAM is to restrict the output to novel, unexpected, or significant information.

What else can SAM do? It can operate like pollsters who sample opinions and predict the outcomes of elections. They rely on experts to advise them how to select people to sample, what to look for, and how to analyze the data that they have collected. SAM can perform similar tasks because it can reach qualitative conclusions about the system by selectively monitoring and analyzing data as they are generated.

In the case of the solar house, a typical conclusion is that "window thickness is unimportant as long as the window has at least one air gap." When a conclusion has been reached, we might well say that SAM is "learning." If this has happened, then SAM's grasp of the heat storage and transfer process of the house has reached the point where the future thermal performance of the solar house can perhaps be predicted. In this instance, the overall computing effort for the simulation may be substantially reduced.

To arrive at conclusions and reduce simulation time, SAM must contain components common to AI codes, such as the following:

- **Expert Knowledge**, or a library of intuitive knowledge, often in the form of "if-then" statements, such as "If the sun is shining on a wall, the wall ought to be getting warmer";
- **Statistical Analyzer**, or a set of codes for determining correlations, means, and likely results;
- **Logician**, or a code for inferring things logically; and
- **Strategist**, or a code that will permit SAM to address questions about the simulation (for example, to answer the question, "Where can we best place a window?" SAM would have to act like a detective, formulating a "game plan" for using the simulation to reach an answer).

Mathematics in AI

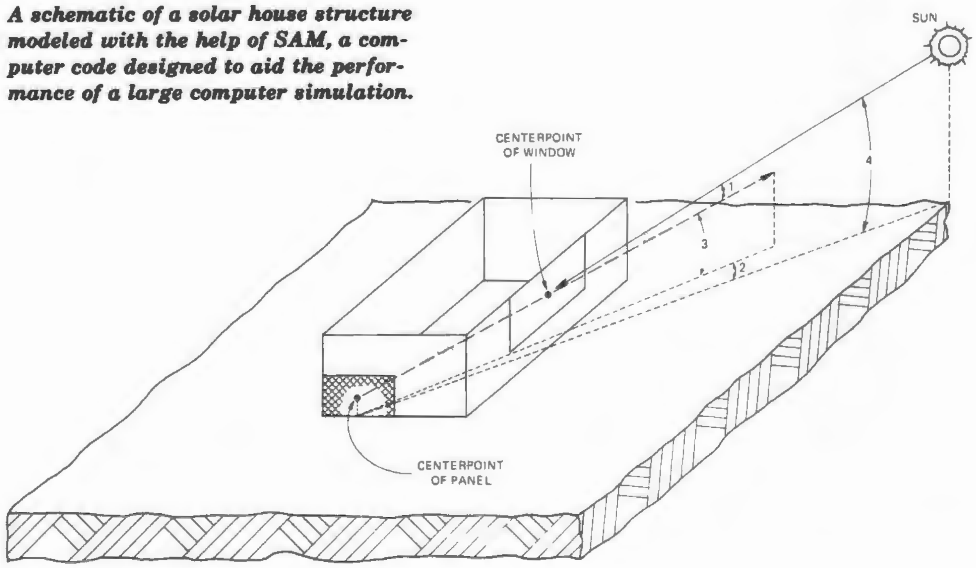
Concepts and methods of mathematics are the essential ingredients of AI because mathematics is the language of real-world processes. Consider some typical AI questions for the program SAM and the mathematical tools needed for their resolution.

Question: SAM is designed to analyze and "know" things about the real world. How can we represent real-world knowledge?

Answer: Knowledge of the real world assumes various forms. Some knowledge is quantitative, like the dependence of the direction of the sun's rays on the time of day. Representing this kind of knowledge requires the familiar idea of a function

$$\left(y = f(x)\right)$$

and leads us to using the tools of calculus. Some knowledge is factual, like the statement "The south-facing wall has a picture window." Representing this kind of knowledge can be done using



A schematic of a solar house structure modeled with the help of SAM, a computer code designed to aid the performance of a large computer simulation.

This computer interacts with itself through the use of a programmed robotic arm.

special mathematical tools called "predicates," or proclamations. A typical predicate formulation of the above statement would be:

TRUE = HAS (SOUTH-FACING WALL, PICTURE WINDOW).

All this says is that the proclamation "The south-facing wall *has* a picture window" is true. The fact that the north-facing wall *did not* have a picture window could be represented as

FALSE = HAS (NORTH-FACING WALL, PICTURE WINDOW).

Use of such predicates leads us to a second kind of calculus, called predicate calculus, which permits predicates to be stored and manipulated in a computer.

Some knowledge is qualitative, such as the statement, "The picture window is large." The word *large* may be quantified by a criterion, such as *large* means that the area is greater than 100 sq ft." On the other hand, we may wish to use tools of set theory, a major branch of mathematics, whereby every object is to belong to one of, say, three collections or "sets": the large objects, small objects, or medium-sized objects.

The statement "In January an average day in Albuquerque has 3 hours of sunshine during the 12-hour period between 7 a.m. and 7 p.m." is said to be a statistical fact. Its representation and use require the tools of another major branch of mathematics, probability theory, which deals with the odds of something happening. The above statement could be reworded in this form: "The probability of sunshine at 12 noon on an average day in January in Albuquerque is 3 chances out of 12, or 3/12 = 0.25."

Of course, other kinds of knowledge also exist. One type is "meta-knowledge," or knowledge of what we know. An example is the statement "I know that Bing Crosby was not 8 feet tall, because if he had been, then this fact would be remembered by me."

Yet another kind of knowledge is "fuzzy" or indistinct knowledge characterized by such words as "I believe" or "it is thought that . . ." Representing and analyzing such knowledge has been the goal of a field called "fuzzy analysis."

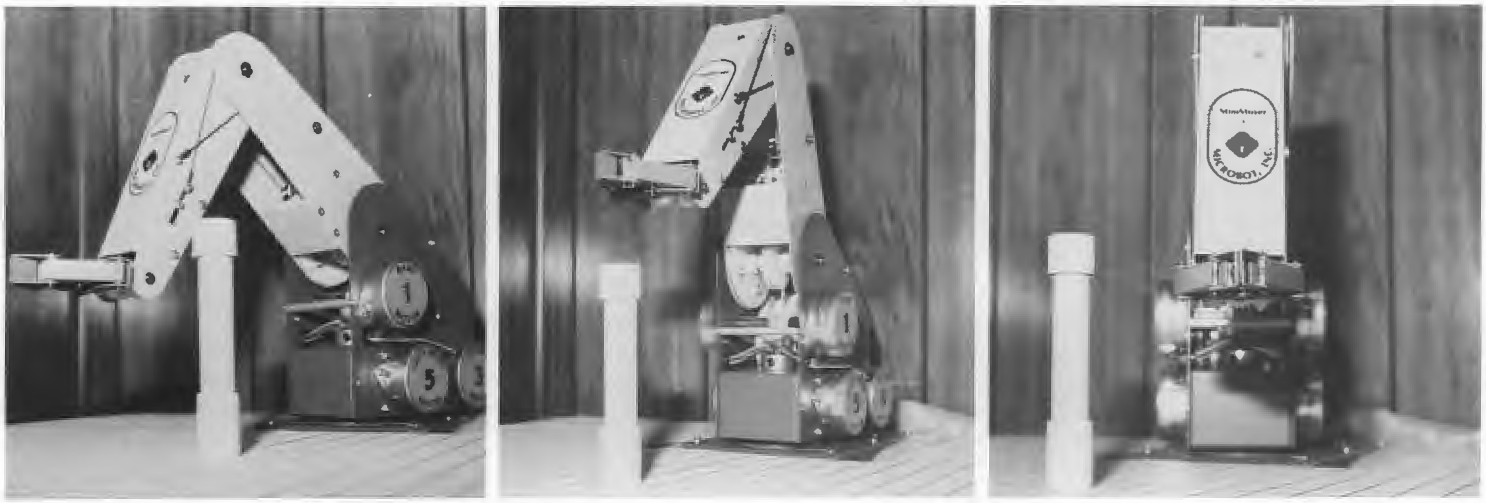
Question: Suppose you wish to ask SAM for the best size of the picture window on the south-facing wall. How can SAM find it?

Answer: *Best* may be defined in a number of ways. It may mean that the selected window costs the least or allows the most light into the room. When this criterion is agreed on—which may not be simple to do—SAM can provide an answer to suit the customer's information needs. Hence, if *best* means lowest cost, then SAM must include a way to compute the cost for a given window size and then use a procedure to find a window size to minimize the cost. A number of different areas of mathematics come into play for this approach. A key area is the "calculus of variations," in which we seek methods for minimizing costs.

Question: How can we infer information from other information?

Answer: Logic, a cornerstone of mathematics, is devoted to consistent thought and inference. The tools of logic can be used to expand our knowledge. Thus from the statement "More sunlight makes the house warmer," we can infer that if the house was colder on Tuesday, there was *not* more sunlight that day.

Logic, however, is a large, complex, and expanding field of



A photo sequence showing the robot arm avoiding an obstacle.

research. It includes branches such as temporal logic, in which the passage of time is taken into account, and modal logic, in which we adjoin such ideas as "necessity" and "possibility" to the standard "true" and "false."

For example, the statement "No body travels faster than light" is supported by so much scientific evidence that we would say "It is not possible for a body to go faster than light." However, this statement is not as logically certain as are statements such as "All bachelors are unmarried" or "It is now either Tuesday or it is not Tuesday." Hence, we are drawn to develop yet finer distinctions of logical certainty than merely "true" and "false." Of course, the resolution of such paradoxical assertions as "This statement is a lie" challenges the human mind even to this day.

Question: The cost of heating and cooling our solar house will depend on many factors. For example, energy costs are influenced by the length of time that the shades are pulled down over the windows, the wall thickness, and the location of the thermostat for auxiliary heating and cooling. If each factor can be varied greatly, then the number of possible things we may do with our house will be enormous. How do we find the alternatives that cost the least now?

Answer: In general the number of possible alternatives is so large that SAM will not be able to examine all of them. Indeed even on the fastest computers available, such searches for least costly alternatives might require hundreds of years of computing effort. To make such searches feasible we restrict the number of such alternatives in various ways, using methods belonging to a branch of mathematics known as discrete mathematics.

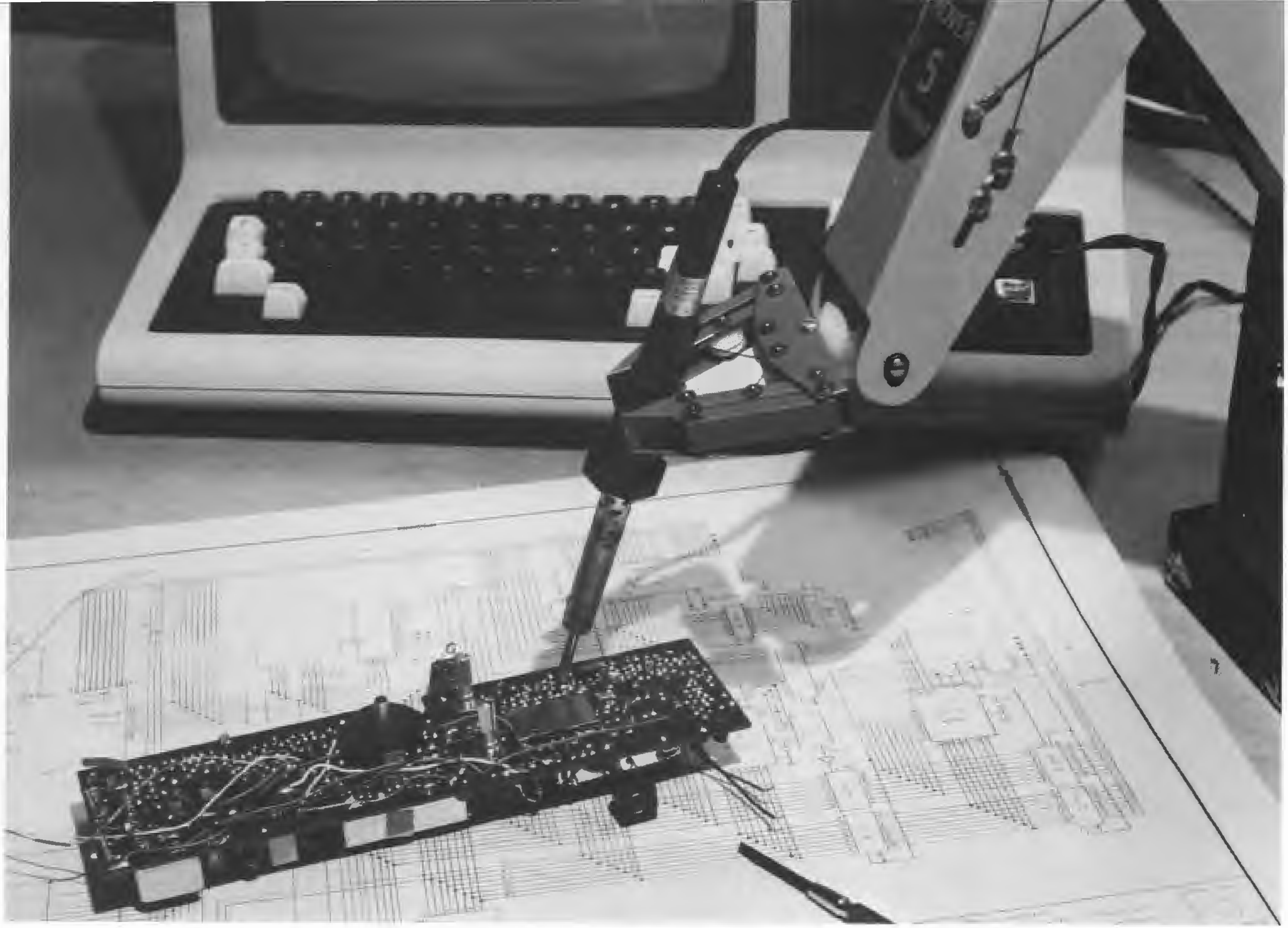
Robotics and AI

A robot may be considered as an extension of the computer into the real world. It enables the computer to interact actively with the physical world and to learn from its observation of how the world responds to its actions. Generally, to fulfill this role, it will have sensors (just as we have eyes to see, ears to hear, and nerve endings to feel) that provide information to the computer. This information must, in turn, be analyzed and stored. Problems in this process can arise from optical illusions such as those in the well-known drawings of M. C. Escher. How even to begin to understand such illusions is currently an open area of AI.

A typical robotic arm consists of a base, shoulder, elbow, wrist, and gripper. Its motion is controlled via simple program steps on a home computer. We wish to have it move from one position to another as quickly as possible without colliding with obstacles in its path. How to do this constitutes the "obstacle avoidance problem" of robotics—and the key difficulty in solving the problem lies in the fact that collision may occur between obstacles (such as the pipe section shown above) and any section of the arm. The problem, however, needs to be solved to ensure (1) the safety of the human workers in the area of the robots and (2) the efficient operation of the robots.

An elegant mathematical problem arising from this enigma is summarized in the question, "How can we quickly know if solid objects intersect?" This problem belongs to a number of mathematical disciplines, including geometry and its meld with calculus, which we refer to as differential geometry.

Most programmable robots used in industry today have only one arm. The single arms can do one



simple task precisely, such as gripping a tool that welds or sprays paint. However, when the complexities of mechanics, electronics, and engineering are worked out, the next generation of industrial robots will have more than one arm. These multiarmed robots will do complex tasks, for example, construction, repair work in hazardous environments such as nuclear fuel reprocessing plants, and extraction of minerals from remote areas such as other planets and the ocean floor.

At ORNL we are exploring the difficult problem of making two arms work cooperatively. We have been using two Microbot MiniMover-5 robot arms controlled by two personal computers—an Apple II and a TRS-80—that can "talk to each other" through suitable connections. We have succeeded in commanding one robot to hand a pipe section to the other. Now, in addition to solving the problem of obstacle avoidance, including the possibility of collision between arms, we are refining our computer programs to coordinate the arms so that one "knows" where the pipe is being held by the other and so that neither one damages the other by wrenching the pipe away.

AI and robotics can, indeed, be enjoyable. Recently, Gary Hall, an Oak Ridge Associated Universities student from Millsaps College, Al Geist of the Mathematics and Statistics Research Section, Thomas Swift, a University of Tennessee student in electrical engineering, and I programmed the two robot arms so that they could play checkers on company time! The robots simply follow the game rules that are stored in the two computers' memories. Although the game programs used by us are very unsophisticated, excellent game-playing programs that use tools of probability, the calculus of variations, logic, and input from experts are available and could be used in this robotics system.

A simple program can be written to make the robot perform a task, such as soldering.

In summary, I have attempted to give a glimpse of AI through the eyes of a mathematician. I have avoided the use of mathematics and have omitted many of the very hard and challenging problems facing us. Like all technical fields, AI will advance through stages of rapid development and quiescence. Indubitably, as it grows, AI will significantly enrich mathematics by leading us to newer areas of excitement, challenge, and practical applications. ornl

BOOKS

***The Information Society as Post Industrial Society**, by Yoneji Masuda, The World Future Society, Bethesda, Maryland (1981), 156 pp + notes and index. Reviewed by William S. Lyon, Analytical Chemistry Division.*

Much have I travell'd in the realms of gold,
And many goodly states and kingdoms seen.

(John Keats, "On first looking into Chapman's *Homer*")

Readers of futurist writings have indeed many goodly states and kingdoms seen, but now for the first time they can travel with a road map. Yoneji Masuda, president of the Institute for the Information Society and director of the Japan Computer Usage Development Institute, is the Japanese counterpart to Alvin Toffler, who in *The Third Wave* popularized the ideas of an information society to the American people. In his book, Masuda programs the advance to the Third Wave from the Heights of Mount Fuji to the Straits of Tsugaru. In a highly stylized, programmed format, Masuda shows what he thinks is the exact route to the Information Society.

Chapman's translation of Homer gave a beautifully written *Odyssey* to the English-speaking world; the translation of Masuda is considerably more prosaic. Perhaps it is characteristic of Masuda or Japanese society, but the translation shows little in the way of graceful writing or a well-turned phrase. Rather, the author writes as if he has received his message from the Great Programmer in the Sky and is passing it on to us in a series of logical computer operations. His "realms of gold" are well papered with self-asserted axioms and sequences of action.

From the first step of the information "revolution" to the ultimate goal of "computopia" ("the rebirth of theological synergism of man and the supreme being"), a process of societal change is laid out. This includes the substitution of automation for manpower, a symbiosis of man and nature to eliminate environmental damage, and the establishment of a public economy (à la Galbraith) instead of private capitalism. A proponent of voluntary action, Masuda believes citizens will band together to share ideas, responsibilities, and facilities. He foresees a time when all our lives will be tied to an information system and when our greatest pleasure, and indeed our only task, will be to develop new ideas and pursue new knowledge. A golden age of information is around the corner.

Well, it sounds great to me. Unfortunately, the author presents no evidence to support his contention that these golden days will actually come. The driving force for these changes is shrouded in inscrutability.

The keystone on which the Information Society rests is the huge data bank that contains information on virtually everything in the world, including the personal lives of all citizens. Keeping personal data private will be difficult, Masuda admits, but he believes that such data will be used only for moral purposes.

But what will prevent persons with evil motives from exploiting such information for their own purposes? Other than a period of public service, he suggests no punishment for transgressors. He believes rather that "the issue of privacy . . . will lose most of its historical significance. . . . The human right to protect secrets will change into a human duty or ethic to share information." How? Why? He never answers.

History suggests instead that such a transformation of human character is unlikely. Will and Ariel Durant, in their summary in *The Lessons of History*, say that "known history shows little alteration in the conduct of mankind. . . . Means and instrumentalities change; natures and ends remain the same." Masuda's computopia would appear to be stymied by human nature.

Nonetheless, American business and the media believe in the Information Society, most of us at ORNL believe in it, and certainly many corporations do. A couple of years ago Frank Jamison, formerly management information systems manager and now personal computer coordinator for ORNL and the other three facilities, presented his thoughts in an ORNL seminar entitled "Information and You: Speculations About the Future." Having spent almost a year in travel, visits, and consultations with many different concerns, Jamison described a scenario much like that of Toffler and Masuda.

Certain puzzling questions continually arise out of such presentations: For example, who will do the dirty work in such a clean society? Who will be responsible for industrial production and mining of raw materials? Is a service and information economy really viable? Most of us, especially engineers, were brought up to believe that industrial production was the key to growth and the "good life."

I asked Jamison his opinion on these matters. His reply went something like this: "I agree with you that the idea of an information society has an intellectual and academic appeal and that a natural question arises as to the need for other societal functions such as basic services and manufacturing. It seems to me that the need for such functions will continue indefinitely. Through robotics and other forms of automation, however, manual labor will gradually be supplanted until a preponderance of jobs in our society will involve 'Third Wave technology.'

"This raises the fundamental and crucial question of how to prepare for such a shift. Public and private education will no doubt be affected, and government must be responsive and adaptive to such a shift," Jamison continued.

"It is my opinion that the inflationary trend since World War II has been, at least in part, caused by this shift of work. That is, we have more and more people engaged in work that produces no salable product. This is not necessarily bad, but it does point out the need for revision in our economic theories."

Certainly, signs of a deindustrialized American society and a rapidly growing information and services sector are abundant. Jobs in heavy manufacturing are disappearing, and automobile and steel factories have shut down. While Americans continue to be voracious consumers, increasingly we are buying our cars, shoes, clothes, and electronic gadgets from Japan and the newly industrialized Third World—Brazil, Mexico, Korea, Taiwan, etc. Because of the Third World's ability to produce high-quality goods at a lower cost than American industry can, our society may appear to have no choice but to become Third Wave.

Our heavy-manufacturing industry is becoming more automated and dependent on robots, thus displacing workers. The demand for computer programmers and engineers is high, yet our unemployment rate exceeds 8%, suggesting a mismatch between skills needed and skills available. Too many people trained for industrial jobs cannot find work in an emerging information, or high-technology, society without retraining.

Another sign is the increased percentage of women in the work force (52% of all American women now work); this increase may be due partly to the recent availability of many new information-type jobs as well as to the feminist movement and the pinch of inflation in the 1970s; in fact, women constitute the bulk of information workers today. Finally, closer to home, we at ORNL see these trends in our work: automated offices and laboratories, paper studies, and a decreased emphasis on hard technology.

A number of business and economic writers have echoed and expanded these ideas. For example, in the November 22, 1982, issue of *Forbes*, James Cook, in a cover story entitled "The Molting of America," points out that, since the 1920s, the best source of new jobs has been the service industries. Of 25 million new jobs created between 1970 and 1982, he writes, only 2.3 million were in manufacturing. In the same issue of *Forbes*, an article entitled "Whatever Happened to Akron?" demonstrates that change from manufacturing to service and information can occur. In 1955, 55,000 people worked in Akron's rubber plants; since 1977, no passenger tires have been made there, and truck tires were phased out in 1982. As the tire business rolled away, information and high-technology industry rolled in.

Numerous books, essays, articles, and television programs have discussed the Information Society and the future. However, I picked Masuda as a focal point because he has some logical sequences in his book and because he is Japanese. For the past few years, emulating a much-admired Japanese industry has become almost a religion in U.S. business circles. Furthermore, his book is highly touted by the World Future Society, which published the first U.S. edition.

George Orwell once said that intellectuals were prone to see the future "as a continuation of the thing that is happening." Typical of this is Gunther Stent's *The Coming of the Golden Age*. Stent, a California molecular biologist, wrote at the height of campus unrest in the 1960s. He predicted a return to the idyllic existence of Polynesia, with a small intellectual cadre ruling a population divided between two classes, typified by the beatnik and the hippie. This is not exactly the kind of society we are living in today.

Masuda also makes predictions based on a continuation of what he thinks is happening. He gives us an optimistic view of mankind (albeit unproved as of now) and a clean, open path to a computopia. Will his predictions be any better than Stent's? Does a high-technology society in the so-called industrialized nations of the West really mean an end to industrialization?

The cover of his book diagrams the process of the computer-communications revolution and its societal impact superimposed upon a drawing of what appears to be Mount Fuji. In one sense, the voyage that is supposed to lead us to the Information Society is a sort of odyssey. The ideas, the cover, the geographical direction from which this volume comes all call to mind the closing lines of Keats:

Then felt I like some watcher of the skies
When a new planet swims into his ken;
Or like stout Cortez, when with eagle eyes
He stared at the Pacific—and all his men
Look'd at each other with a wild surmise—
Silent, upon a peak in Darien. ornl

Jim Turner (right) is a staff physicist in ORNL's Health and Safety Research Division. A native of Virginia, he grew up in Savannah, Georgia, and received a Ph.D. degree in physics from Vanderbilt University in 1956. After serving as an instructor in physics at Yale University and a radiological physicist at the U.S. Atomic Energy Commission headquarters in Washington, D.C., he joined the staff of ORNL in 1962. In addition to his interest in the physics of metal toxicity, he continues to do research on the interaction of radiation with matter. Bruce Jacobson is a senior research biochemist in ORNL's Biology Division. He was born in Iowa and raised on a farm in New York. He received his Ph.D. degree in biochemistry from Johns Hopkins University in 1956 and held a postdoctoral appointment at the California Institute of Technology for two years. Since coming to ORNL in 1958, Jacobson has conducted research on enzyme regulation, biochemical genetics in the *Drosophila* fruit fly, the relationship of structure to the function of transfer RNA, and the effects of metal ions on proteins and nucleic acids. Wendy Williams is a solid-state physicist in the Health and Safety Research Division.

She came to the United States from her native England in 1960, the year she received a Ph.D. degree in solid-state physics from the University of London. After teaching at Louisiana State University in New Orleans, she came to the University of Tennessee in 1965. Shortly thereafter, she began consulting at ORNL and became a full-time staff member in 1972. She continues to perform research in solid-state physics while carrying out collaborative work on metals in the Biology Division. Here, the three authors discuss the results of experiments in which *Drosophila* fruit flies are exposed to toxic metals.

Predicting Metal-Ion Toxicity

A Collaboration of ORNL Physicists and Biologists

By JIM TURNER, BRUCE JACOBSON, AND WENDY WILLIAMS

The benefits of life in modern industrial societies do not come without an environmental price tag. One cost that has received increasing attention arises from the ever-increasing addition of toxic metals—such as cadmium, lead, and mercury—to the human environment. For example, measurements made in Sweden show that the background concentration of cadmium in wheat has doubled in the past 100 years and that the cadmium concentration in human kidneys has more than tripled. In the past two decades, the news media have deluged us with accounts of deaths and illnesses from the ingestion of lead in paint and of mercury compounds in tuna; we have also heard much about the attempts of governments to control the release of these toxic metals to the environment, such as regulations to reduce the amount of lead in gasoline.

What do these toxic metals do to living organisms, particularly humans? How do they wreak their havoc on body cells? As in the case of ionizing radiation, the regulation of exposure to metals and other chemicals should be based on some understanding of how they alter normal biological processes. However, the scientific foundation for understanding and determining safe levels of human exposure to toxic metals and other chemicals is much less nearly complete than it is for radiation. What can be done? Here is how one group at Oak Ridge National Laboratory got involved.

ORNL physicists and biologists are collaborating in a search for fundamental explanations of the toxic effects of metal ions in biological systems. Their goal is to predict the degree of toxicity of metal ions and other chemical pollutants.

The Challenge

It all started six and one-half years ago when an ORNL researcher was called into his division director's office to hear words to this effect: "You have been successful in doing research for many years to develop a basis for health protection from radiation. What we need now are some analogous scientific principles for predicting risks from exposure to chemicals and for regulating chemical exposures. Why don't you give that a try? We will provide support at a level of two persons, leave you alone for about three years, and then evaluate what you have done. If the chemical problems are too diverse and difficult to yield useful generalities, then that itself is probably worth establishing. One condition, though: The new effort must be undertaken in collaboration with the Biology Division."

This challenge was offered to Jim Turner by Steve Kaye, director of ORNL's Health and Safety Research Division (HASRD) after Kaye returned from a visit to Germantown, Maryland, in August 1977. Kaye and Bob Wood, director of the Pollutant Characterization and Safety Research Division of the U.S. Department of Energy (which supports the basic physics programs in HASRD), had agreed to fund a trial collaborative effort between physicists and biologists for the development of some health-protection principles to be applied in the nonnuclear arena.

Accepting the challenge was easy. The major problem was to find out what to do in a project that seemed to be a long shot. As a theoretical physicist, Turner had little background at that time in biological subjects and only a few ties with members of ORNL's Biology Division—through the Oak Ridge Tennis Club. However, the acceptance of this mutual professional venture by several members of the Biology Division was eventually to change that. During most of the first year, Turner spent his time making systematic contacts within ORNL and elsewhere. Through his discussions with other ORNL staff members, he tried to determine how to turn the lofty ideal of doing nonnuclear "dosimetry" into bench science, especially the publishable variety. Wendy Williams, also a HASRD physicist, joined Turner at this early stage. Rather than trying to build a new program from scratch, they sought to match their objectives to the strengths of ongoing programs in the Biology Division.

Turner and Williams contacted John Storer, then director of the Biology Division, who suggested that they work through Dick Kimball, who knew the division well. An initial exploratory meeting was held in the Biology Division on October 26, 1977. From HASRD the attendees were Turner, Williams, and Harvel Wright; from the Biology Division came Bob Cumming, Dave Doherty, Jim Epler, Michael Fry, Kimball, Salil Niyogi, Gary Sega, Jim Selkirk, and Peter Witschi. A few weeks later a group from the Biology Division visited the experimental physics laboratories of HASRD and then met with Kaye.

In the Biology Division, Witschi was organizing a comprehensive new program in toxicology, and Turner began spending all his mornings there. Turner began a lengthy period of reading, discussing, and attending classes at the University of Tennessee Graduate School of Biomedical Sciences. One obvious problem was communication—that is, finding a common ground for discussion among physicists, biochemists, and toxicologists. Eventually Turner learned to "speak biochemistry."

Metal Ions, Organisms, and Cells

Before long, we decided that metal ions should be the first category of important chemicals to study. Witschi had shown that beryllium interferes with ribonucleic acid (RNA) synthesis. Niyogi and Rose Feldman were conducting studies to determine how metal ions cause RNA polymerase to make errors in transcribing the genetic code. Independently, in early 1978 Bruce Jacobson had received seed money to investigate the effects of cadmium on certain enzymes (substances that stimulate chemical reactions in the body's cells) and transfer RNAs (ribonucleic acids, also called tRNAs, that help translate information from messenger RNAs for the synthesis of enzymes and other protein molecules). At the same time, Abe Hsie had been evaluating the mutagenic effects of metal ions in Chinese hamster ovary (CHO) cells. In addition, Sankar Mitra was interested in studying how metal ions could affect the replication of deoxyribonucleic acid (DNA) by DNA polymerase.

Because of these studies, expertise and facilities were already available in the Biology Division for studying the effects of metals at various levels in diverse biological systems. In addition, from the point of view of the physicists, metals were attractive because a metal atom or ion is a tractable physical system that has well-defined, measurable properties. Also, a metal usually maintains its original identity in association with living systems; by contrast, many organic chemicals undergo molecular transformations.

Regular meetings were organized for those interested in developing the metal ion-toxicity program. In addition to those named above, Elija Johnson, Al Narten, Bill Thiessen, and Roberto Triolo participated from the Chemistry Division. The development of the new program benefited vastly from their interest, guidance, and critical ideas.

Specific objectives for research on metals came clearly into focus. How are the unique properties of each metal ion related to its individual biochemical action? What properties might be the most relevant for inducing adverse effects in living systems—and why? What physicochemical characteristics of a metal ion might give a hint as to its possible toxic effects and, hence, serve as a predictor of toxicity?

As these studies were being planned and conducted, we sought advice and feedback from persons outside ORNL. Robert Hanzlik from the University of Kansas was invited to ORNL. He presented a seminar and gave advice on metal-ligand models. Gunter Eichhorn from the National Institutes of Health listened to plans about the developing program and described his latest research on the interactions of metal ions with nucleic acids. Eichhorn's visit was arranged in October 1979 to coincide with a two-day Symposium on Metals and Carcinogenesis at the University of Tennessee Memorial Research Center. Walter Farkas of the UT Center received funds for the symposium from the American Cancer Society and asked us to help plan it. Results from the ORNL metals work have also been presented as invited and submitted contributions at meetings in this country and abroad.

Toxicity testing requires extensive use of Biology Division facilities. Here Bob Lindenschmidt places mice in an environmental chamber.

Softness and Toxicity

Specific projects began to take shape toward the end of 1979—the second year. Jacobson and Turner undertook a systematic study of the physicochemical properties of cadmium and certain other metal ions in relation to their interactions with proteins and nucleic acids. They assembled and analyzed con-

Salil Niyogi studies the effects of metal ions on RNA transcription reactions.

siderable biochemical data on metals and presented physical arguments on how experimental findings could be understood on the basis of the physics of the metal ions in solution. They applied the chemists' theory of hard and soft acids and bases to link specific metal ions to specific sites of interaction on biological macromolecules. Quite a range of diverse phenomena in the literature could be fit into these ideas.

The "softness" of a metal ion is related to the ease with which its electron cloud can become deformed in an electric field. Soft metal ions, such as cadmium (Cd^{2+}) and mercury (Hg^{2+}), tend to form covalent chemical bonds, or share electrons, at soft sites on biological molecules. In contrast, hard ions such as calcium (Ca^{2+}) and strontium (Sr^{2+}) form ionic bonds at hard sites on biological molecules. Furthermore, chemical softness for a given metal ion can be defined quantitatively in several ways to provide numerical indices for that metal. A softness parameter thus seemed worth exploring as a possible predictor of toxicity. Jacobson and Turner prepared a manuscript on this work. This first paper from the new program appeared as a review article in *Toxicology* in the summer of 1980.

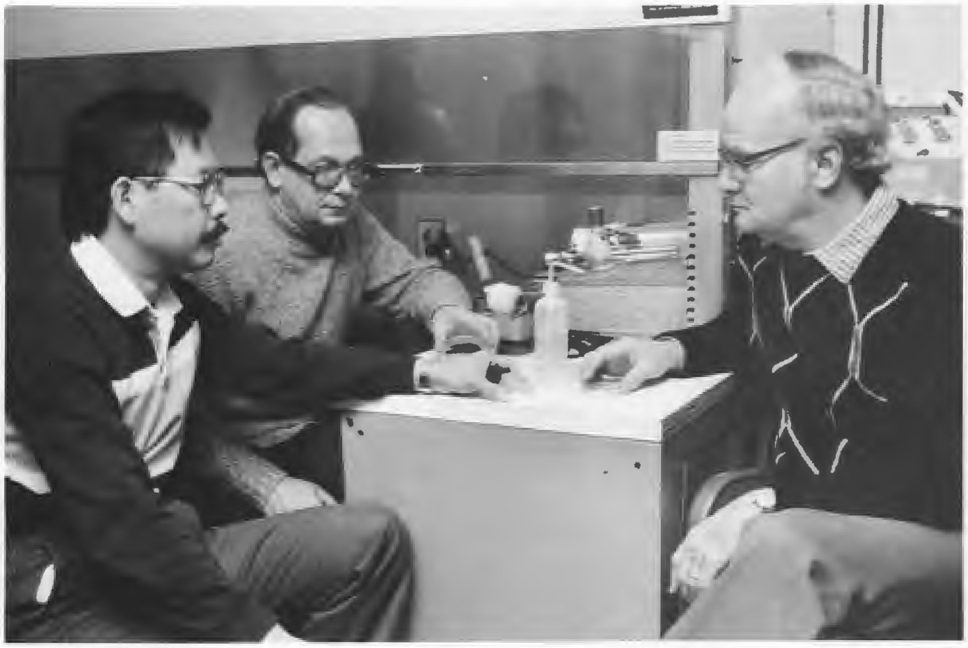
Triolo had first called attention to the fact that one or two papers had been written about correlations between chemical softness and acute lethality in mice and rats, expressed in terms of the 14-day LD50 (the amount of metal, e.g., millimoles per kilogram of body weight, that, if administered to mice, kills 50% of the animals within 14 days). The correlations in the literature were based on data taken at a number of laboratories on different strains of mice and rats that had received the metal ions through various routes of administration.

Would correlations between softness and LD50 improve if data obtained under uniform experimental conditions were used? Because such data did not exist, an experiment was carried out in the summer of 1980 in Witschi's laboratory in the Biology Division to measure the LD50 for 24 metals in mice under uniform conditions. The correlation found between chemical softness and mouse LD50 was significantly improved over that reported by earlier investigators. Of the divalent metals tested, the ranking from most to least toxic is Cd^{2+}, Hg^{2+}, copper (Cu^{2+}), platinum (Pt^{2+}), zinc (Zn^{2+}), barium (Ba^{2+}), beryllium (Be^{2+}), nickel (Ni^{2+}), lead (Pb^{2+}), palladium (Pd^{2+}), cobalt (Co^{2+}), manganese (Mn^{2+}), magnesium (Mg^{2+}), and Sr^{2+}.

The study was simultaneously broadened biologically by Jacobson and Nel Christie, who added aliquots of the same solutions of metal ions to the diet of the fruit fly, *Drosophila melanogaster*. They measured the four-day LC50 (i.e., the dietary concentration that kills 50% of the flies in four days) using a procedure that Lee Opresko helped develop. As with the mice, a correlation between LC50 and the hard-soft properties of the metals was found.

The same metals were subsequently tested in Hsie's lab by Eng Tan, Bob Schenley, and Stella Perdue, courtesy of an ORNL seed-money grant to Turner, Williams, and Hsie. Again, a correlation with softness was found in terms of the reduced cloning efficiency of CHO cells.

Among these three biological assay systems—mice, *Drosophila*, and CHO cells—some similar and some different traits were found. For example, cadmium proved to be the most toxic metal to all three. Strontium, on the other hand, was highly toxic to *Drosophila* but not so to the other two. This difference may be caused by the low levels of



Abe Hsie, Bob Schenley, and Jim Turner (right) discuss the survival of cells after treatment with metals and subsequent plating to determine cloning efficiency.

Peter Witschi examines a hamster. Much can be learned about health effects of essential and toxic metals by studying their effects in animals.

chemically similar elements, such as calcium, in the fruit fly.

Search for Predictors

Although correlations between softness and toxicity were found, the search continues for other parameters that give better correlations. One approach to finding other parameters is being pursued by Dave Gosslee of the Computer Sciences Division and by Tom Hayden, a consultant from the Mathematics Department of the University of Kentucky. They are using multiparameter analysis to test whether linear combinations of various measurable physical characteristics of metal ions can provide significantly better correlations. Sophisticated statistics are being used, and some new leads are appearing.

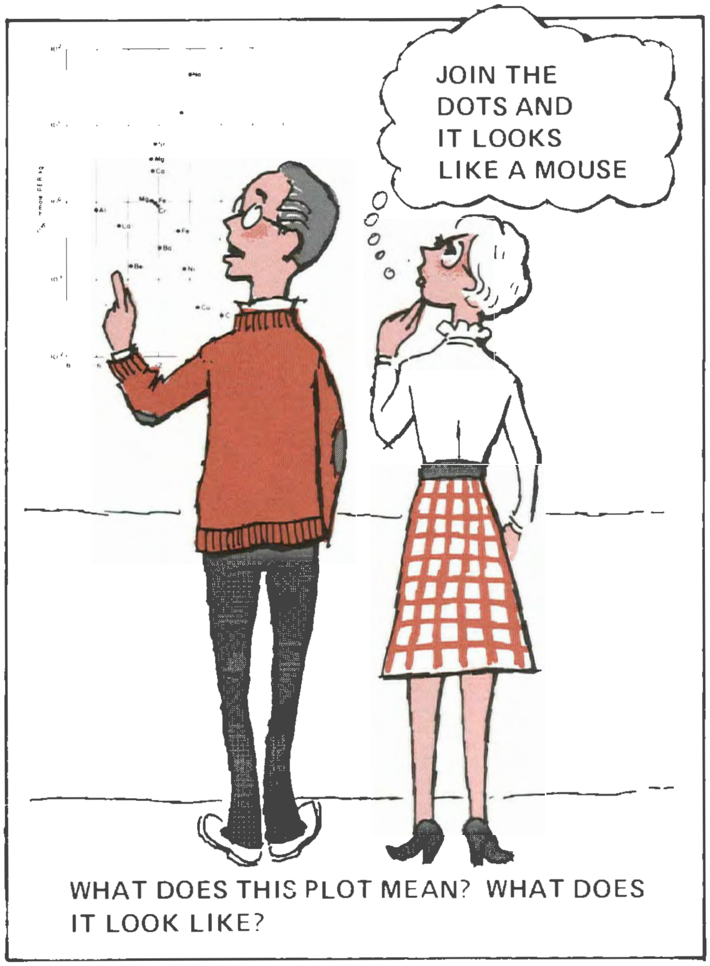
Still another approach is to try to correlate LD50 with the effects of metals on some biological process as a predictor of toxicity. Toward this end, Williams is currently learning experimental techniques in Jacobson's laboratory. After *Drosophila* are fed known concentrations of metal salts, she and Jacobson will try to correlate the amounts of metal-binding proteins produced by the *Drosophila* with the toxicities of the metal ions. It has already been found that exposure to cadmium results in elevated levels of such a protein in the flies.

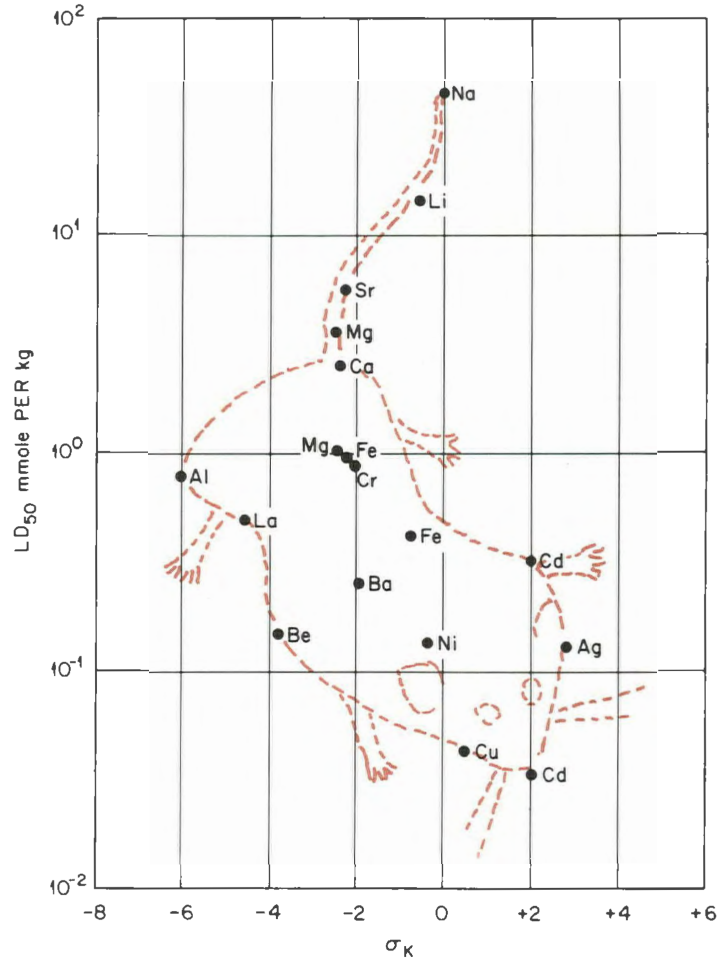
Investigators outside of ORNL are independently using some of our data. A group at the University of Kentucky recently published a study that shows a correlation between the ORNL mouse LD50 data and *in vitro* inhibition of calmodulin activity by different metals. Calmodulin is an intracellular calcium-ion receptor protein that regulates a variety of cellular enzymes and processes.

In all, the ORNL correlation studies have drawn upon a wide spectrum of scientific collaborators. Statistical protocols had to be set up, relevant metals chosen for study, pure metal reagents prepared for biological testing, and the exposed organisms housed and cared for. Collaborators in addition to those mentioned above include Jim Hoeschele of HASRD and Eva Lee, Carol Paton, Keith Owenby, and Lawton Smith of the Biology Division.

Metal Ions and Monte Carlo

Paul Todd of the University of Pennsylvania suggested that Turner look at some new data from Los Alamos National Laboratory (LANL). Ed Hildebrand of LANL had measured the transport and partitioning of cadmium as a function of time in several compartments of CHO cells exposed to this metal. Hildebrand's data were ideally suited to analysis by Monte Carlo procedures long used in HASRD as a tool for calculating radiation transport through matter.





The Monte Carlo approach is applied as follows. A cadmium ion in one cellular compartment (e.g., bound to a protein in the cytoplasm of a cell) has a probability, per unit of time, of moving to any other compartment. Informed guesses are made for the initial set of probabilities for intercompartmental transport of cadmium ions. The Monte Carlo calculations are then carried out, and the computed distributions of cadmium in various compartments as functions of time are compared with data from experiments. The initial probability estimates are refined and adjusted until the experimentally observed temporal transport is reproduced. The final set of probabilities constitutes a numerical representation of the behavior of cadmium in CHO cells.

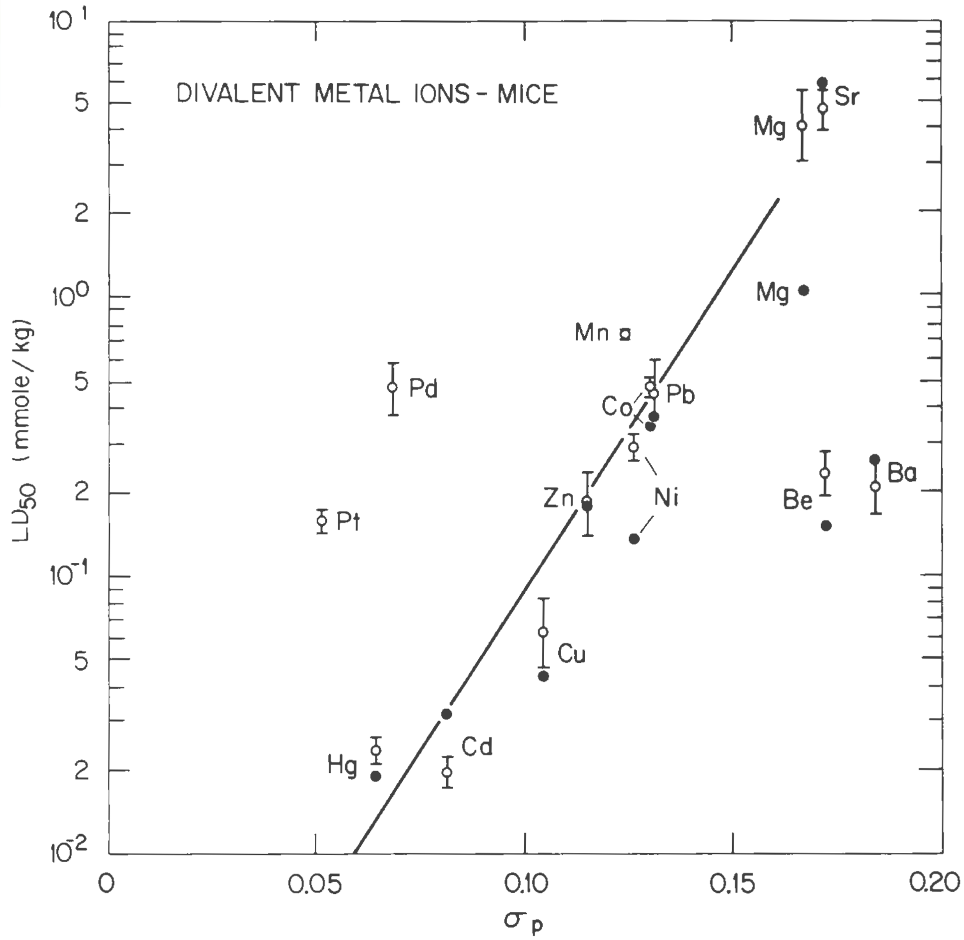
The idea was to carry out such Monte Carlo calculations for a variety of toxic and nontoxic metals and see whether any predictive patterns emerged in the values of the probabilities. Paul Furcinitti collaborated with Turner in some early programming to analyze the cadmium data of Hildebrand. As specific questions began to arise in their modeling, John Cook and Hsie provided essential input and advice. Hayden, with assistance from the others, completed the modeling of the LANL cadmium data. This work has been published.

Using a set of well-defined growth conditions and another subclone of CHO cells, Tan has also obtained data on cadmium transport. Although this line of investigation shows promise, the required metal transport experiments are difficult and costly, and further laboratory work with other metals has not yet been carried out. The Monte Carlo calculation provides a tool for studying the route of invasion of a cell by metal ions, but it is too early to say which step in that route is lethal to the cell.

Search for Basic Understanding

The studies described above focused on one aspect of the metal toxicity program by using the whole organism to search for a numerical index or predictor that correlates with chemical toxicity. It may be more important in the long run to understand basic mechanisms by which the metals can kill. Why does a cell or organism die from an excess of cadmium or zinc or barium? What specific targets

An example of mouse LD50 values for divalent metal ions as a function of a softness parameter, σ_p. The open circles represent data obtained at ORNL under uniform experimental conditions, and the closed circles represent data taken from the literature by other investigators. For the 11 metals common to both groups, the correlation coefficient for the ORNL points is 0.77 compared with 0.60 for the data from the literature. Error bars show the 95% confidence limits for the ORNL data.



are attacked by metal ions? How do metals interfere with vital processes and lead to death? Clearly, it is essential to "look inside the black box" and try to discover the details of changes induced by metals at the molecular level. In fact, the correlations we seek may be most clearly demonstrable there.

Jacobson has been using a variety of experimental techniques to study the interaction of metal ions with nucleic acids and certain enzymes. One of his tools is the RPC-5 chromatography system developed several years ago by Don Kelmers and Dave Novelli. Chromatography, in this case, is a fractionation procedure in which a mixture of substances (tRNAs) is run through a column where they separate according to their individual affinities for the matrix (in this case, RPC-5) in the column. Jacobson and Vivian Hiatt of Knoxville College studied the chromatographic behavior of different tRNAs in the presence of various metal ions, and they found an unexpected result. In contrast to cadmium, magnesium, and nickel, increasing concentrations of zinc caused the tRNAs to elute—leave the material on which they were adsorbed—at progressively higher ionic strengths. This finding does not appear to be consistent with the theory of hard and soft acids and bases.

How do zinc ions affect tRNAs and how do they interact with other nucleic acids, such as DNA and messenger RNA? These questions are being studied by John Flanagan of the University of Tennessee. Identifying binding sites of metal ions and subsequent structural changes is fundamentally important in determining how metal ions cause detrimental effects. Nuclear magnetic resonance (NMR) spectroscopy offers a powerful technique for investigating these questions. Brian Reid at the University of Washington is using NMR to help delve further into the ORNL finding on the behavior of zinc. His instrument is one of the few in the world that has the high resolution needed to study the structure of small nucleic acids.

Other studies have focused on the anticodon of tRNA, the sequence of three nucleotides in tRNA that reacts with messenger RNA to translate genetic information into the structure of protein molecules. Using the RPC-5 column, Jacobson found that cadmium perturbs those tRNAs that have either guanosine (G) or queuosine (Q, a rare nucleotide) in the first position of their anticodon. This finding led Brian Hingerty to examine a mathematical model for the binding of a metal ion to GpC [guanylyl-(3′,5′)-cytidine], a dinucleoside monophosphate, a portion of the anticodon of some tRNA molecules. Hingerty calculated the most stable binding sites for the hard ion, calcium, and the soft ion, cadmium, both of which have the same size and charge. Calcium causes minor perturbations in the three-dimensional GpC structure; in contrast, cadmium causes both the guanine and cytosine rings to shift and rotate 90° in the GpC struc-

Lawton Smith, left, and Witschi discuss toxicological protocols in front of a scintillation counter.

ture. A similar calculation is being performed to see whether cadmium interacts differently with QpU than with GpU (U = uridine) to shed some light on the cadmium-tRNA experiments. Hingerty is also working to set up a program that will allow calculations to be done for a complete tRNA molecule and a complete turn of double-stranded DNA, a very challenging task. Recently, Jacobson has also obtained the X-ray diffraction coordinates for the crystal structure of queuosine monophosphate from colleagues at the University of Tokyo.

Niyogi and David Hoffman published a paper in *Science* in 1977 on the relationship between the known carcinogenic and mutagenic properties of metals and their effect on RNA synthesis rates. The genetic information from DNA is copied through the synthesis of messenger RNA, which in turn specifies the structure of proteins—the ultimate expression of the genetic information. If this copying process, or transcription, is inhibited, the amounts of certain proteins will be diminished or they will be lost altogether. Niyogi, Feldman, and Hoffman demonstrated that a number of metal ions tend to inhibit synthesis of messenger RNA. Such inhibition may be caused by a failure in the formation of the very first bond (initiation) that joins the first two nucleotides or in the elongation process farther down the chain (messenger RNA may contain several hundred nucleotides). Seven metal ions did inhibit this initiation process at concentrations that inhibited the overall transcription. These metal ions, all hard, are lithium (Li^{+}), sodium (Na^{+}), potassium (K^{+}), Ca^{2+}, Mg^{2+}, Sr^{2+}, and Zn^{2+}; they are believed to play no role in causing mutations or cancer. Certain other metal ions that have been shown to cause mutations or cancer or both are Pb^{2+}, Cd^{2+}, Co^{2+}, Be^{2+}, and Mn^{2+}—all soft metal ions; this latter group of ions stimulated the initiation process at low concentrations. Some of this latter group of metal ions simultaneously stimulated initiation and inhibited the overall synthesis of messenger RNA, suggesting multiple sites of initiation on the DNA template. Whether this activity may relate to cancer production by these metal ions is not clear.

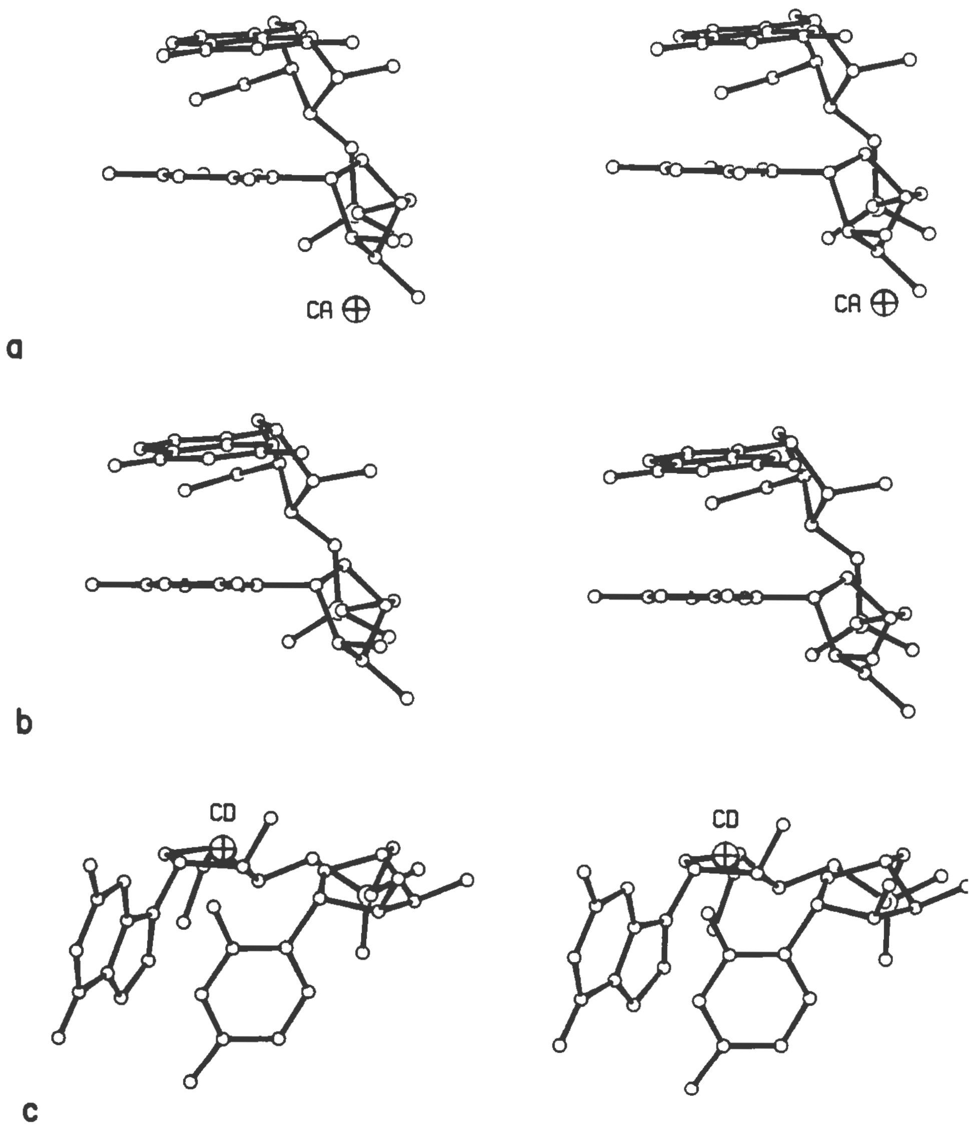
Outlook

What have we learned? How do any findings thus far help us predict or understand metal toxicity? Where do we go from here? Each of the studies mentioned above has provided some information and insight. Let us speculate on one of them.

Chemical softness provides a useful numerical parameter to consider in evaluating the potential hazards of metal ions to humans. We are searching for other properties, particularly of a more biological nature, that will provide better correlations with acute toxicity as measured in our test systems. Apart from correlation studies, the physics of hard and soft metal ions suggests some reasons for the different degrees of toxicity of different metals.

A chemical system as complicated as a living cell is replete with sites that bind multivalent ions. Soft ions form bonds of a highly covalent character in which electrons are shared by the ion and the molecule to which it binds. Symmetry requirements restrict the directions in which covalent bonds can form about a soft metal ion. Therefore, such an ion may well introduce structural changes in the molecule to which it binds. In contrast, the predominately ionic bonds of hard ions do not involve as much electron sharing and potential conformational disruption. Soft metal ions such as Cd^{2+} will find numerous sites for covalent bonding with biological macromolecules. In so doing they are likely to cause detrimental conformational changes, some of which may be

Stereoscopic views of the dinucleoside monophosphate GpC in water (a) with bound calcium (Ca), (b) in its native state, and (c) with bound cadmium (Cd). The hard calcium ion forms ionic bonds with the phosphate linkage. The soft cadmium ion, which has the same charge and ionic radius as the calcium ion, forms covalent bonds with both the nitrogen (N_3) atom of guanine and the oxygen (O_3) atom of the ribose. The directional symmetry required of covalent linkages causes considerable distortion of the GpC structure in (c).



critical to a biological system. Thus, the metal is toxic. We are currently testing the hypothesis that the degree of covalent-to-ionic nature of the bonds that a metal ion can form gives a general prediction of its toxicity.

The time has passed rapidly since the initial challenge was extended in August 1977. As originally intended, the program was reviewed after three years, once in Washington and once on a site visit in Oak Ridge. Both DOE and ORNL agreed to continue the collaborative metal-toxicity program. Most recently, the participants discussed the program's objectives and future with the two division directors most closely involved, HASRD's Kaye and Dick Griesemer of the Biology Division. If continuing DOE and ORNL support and the enthusiasm of the many participants are a measure, then the program is a success. This collaborative effort has developed an approach, an outlook, and a forum for communication and understanding among persons with different training but with a common scientific purpose.

Is it necessary to understand metal-ion toxicity at fundamental levels? Can we not control the entrance of toxic metal ions into our biosphere and simply avoid the exposure of humans to toxic metals? An article in the August 1983 issue of *Nature* emphasizes the importance of these questions. "Despite increasing attempts to control environmental pollution, changes in the distribution and availability of toxic metals like mercury and cadmium are still occurring. Apart from natural processes, other contributory factors include the gradual spread of industrialization, the use of sewage sludge as fertilizer, and the acidification of Northern Hemisphere groundwater. Animals (including man and domestic varieties) can accumulate harmful concentrations of toxic metals."

Because toxic metals and other hazardous chemicals are not being adequately contained, we had better learn all that we can about them. Much work remains to be done to establish the necessary scientific basis for predicting risks from exposure to chemicals and for regulating chemical exposures. ornl

Paul A. Haas, a native of Rolla, Missouri, holds chemical engineering degrees from the University of Missouri at Rolla, Montana State University, and the University of Tennessee. He came to ORNL to attend the 1951–52 Oak Ridge School of Reactor Technology and has been with ORNL's Chemical Technology Division since 1952. His Ph.D. thesis was done at ORNL on the foam separation process for treatment of low-level radioactive waste solutions. He has worked on engineering development for most parts of the nuclear fuel reprocessing and recycle operation, isotope separation processes, waste treatments, and transuranium processing. Haas holds ten patents in these areas, including five on gel-sphere technology. He is a registered professional engineer in Tennessee and a member of the American Institute of Chemical Engineers, American Nuclear Society, and American Chemical Society. Here, Haas (right) confers with Sam Clinton about gel-sphere technology.

Sol-Gel and Gel-Sphere Technology:

Powders for Power

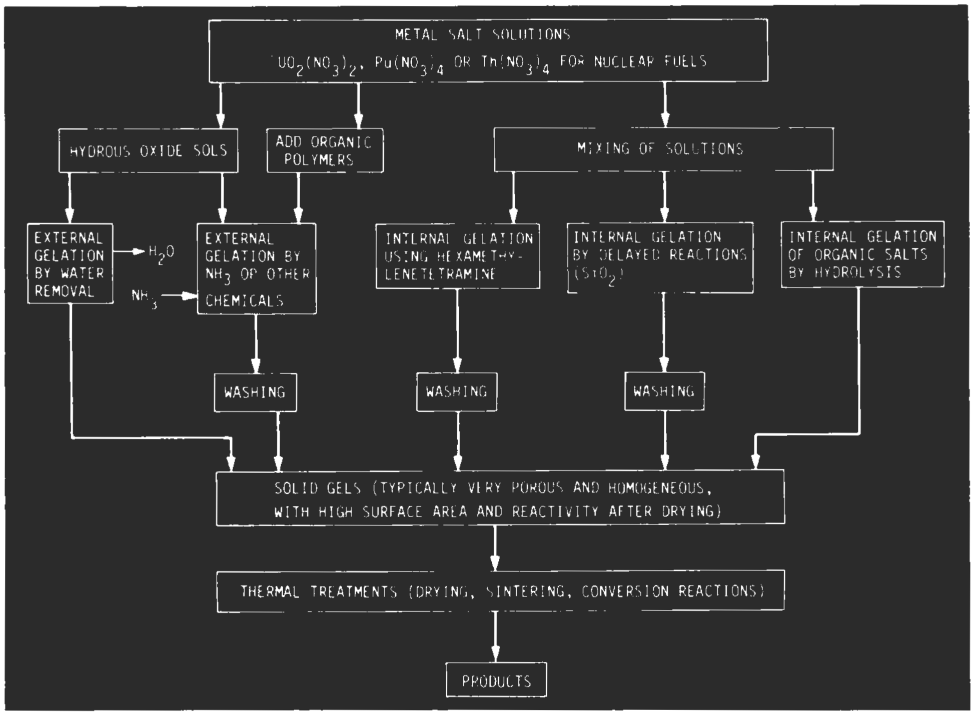
By PAUL A. HAAS

Powders for ceramics, cosmetics, pigments, and other uses are usually produced with little control of the particles' size, shape, or microstructure. Oak Ridge National Laboratory, however, has been making powders for years with close attention to and control of the size, shape, and structure. Many of these products consist of spherical, beadlike particles, formed by solidifying (gelling) droplets or particle suspensions. ORNL has used these particles for nuclear reactor fuels and a new nuclear waste form; we are now employing them for such nonnuclear applications as tougher ceramics for possible use in energy-efficient heat engines and as improved varistors that could be used to protect high-voltage lines from destructive power overloads. These special "powders for power" may give a facelift to nuclear and nonnuclear technologies.

We have had active programs to develop sol-gel or gel-sphere

Sol-gel technology, developed over a 25-year period at ORNL, has been used to make spherical, beadlike particles for nuclear reactor fuels. Today industry is showing interest in the technology for making ceramics of uniform composition for electronic and other nonnuclear applications. A researcher involved in the development of the prizewinning technology tells its history.

This flow sheet shows sol-gel and gel-sphere processes for conversion of metal-salt solutions to ceramic products.



processes for making these special powders almost continuously since the late 1950s. The primary applications were in the preparation of nuclear reactor fuels, but other fuel cycle applications and nonnuclear uses have also been studied. Exxon Nuclear Corporation and other nuclear-oriented companies have shown interest in using sol-gel technology, but this interest has waned because their industry is in the doldrums. However, several companies are interested in sol-gel technology for making zinc oxide (ZnO_2) and other ceramic materials for nonnuclear applications, particularly in electronics.

Gel Processes

The sol-gel and gel-sphere processes developed at ORNL convert metal-salt solutions to ceramic solids. The sol-gel process gets its name from the fact that the process converts the metal-salt solution to a sol—a colloid in liquid form—and then into a gel, which is a colloid in a more solid form than a sol. (A colloid is a suspension of finely divided particles that tend not to settle out of suspension.) The chemical compositions of our gels are those of hydrous metal oxides. The sol-gel name applies when the hydrous metal oxides are first prepared as a sol of colloidal particles in a liquid (metal-salt solution) and are then solidified, or gelled. The gel-sphere name refers to the formation of the gel from drops (which may be solutions or sols) to give solid spheres that resemble tiny beads. Some of the processes are both sol-gel and gel-sphere types, while others are only one of these.

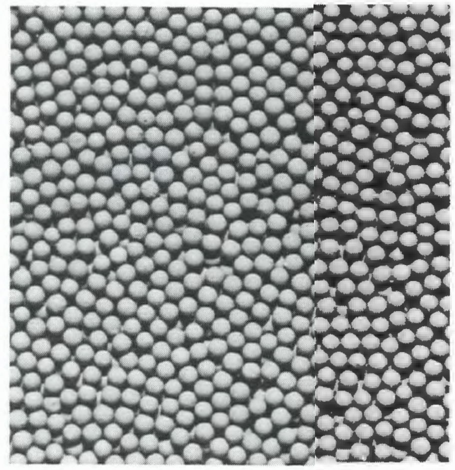
Gel processes have several advantages over conventional ceramic processes. They produce no dust and can be run at lower temperatures, thus saving energy and allowing use of simpler equipment. Gel processes start with solutions that can be highly purified and easily mixed to give homogeneous compositions, and the products will usually be as homogeneous and pure as the starting solution. Furthermore, gel processes can produce oxide gels that are particularly useful, not only because they have a uniform composition but also because they are highly porous, highly reactive, and spherical, unlike the products of conventional processes.

The limitations of gel processes are that they involve more complex chemistry and require more chemicals than do conventional processes, which rely largely on precipitations and thermal decompositions to convert metal salts to ceramic solids. These limitations, however, are overshadowed by the advantages of using gel processes for specific applications that require the unique gel characteristics.

Gel processes can be grouped and described in terms of the reactions that change the solutions or sol into a solid gel. One method of forming a gel is to remove water from a colloidal sol—a method used at ORNL from the late 1950s through 1972. By extracting the water from sol drops into an immiscible alcohol, ORNL researchers formed gel spheres. Patents based on this dehydration method were received by Sam Clinton, Orlen Dean, Don Ferguson, Todd Kleinsteuber, Milt Lloyd, John McBride, Ken McCorkle, and me. This method is a type of "external gelation," because mass transfer to the surrounding second fluid is used to produce the gel. Another type of external gelation uses mass transfer of ammonia from the surrounding gas or solution; the ammonia reacts chemically with the metal-salt solutions to form the hydrous oxide gels. This process was developed to prepare nuclear fuel spheres in Italy, Germany, and England.

Unfortunately, the mass transfer for external gelation limits the gelation rate and can result in

The formation of mixed uranium and plutonium oxide spheres is accomplished when solution drops are solidified in a hot organic liquid, washed, and dried.



nonhomogeneous, or shell, structures. To ensure a more homogeneous product, researchers have used slow chemical reactions for gelation so that the liquids can be mixed thoroughly and then allowed to gel without mass transfer. This method of reacting metal-salt solutions with special chemicals is called "internal gelation" because gelation occurs without mass transfer to a second "external" fluid.

Current gel-sphere processes used at ORNL are internal gelation processes initially developed in the Netherlands by M. E. A. Hermans and others. In these processes, hexamethylenetetramine (HMTA) is mixed at about 0°C with uranyl nitrate [$UO_2(NO_3)_2$] and plutonium nitrate [$PuO_2(NO_3)_2$] solutions. Hydrous uranium-plutonium oxides [$(UPu)O_x$] are formed because HMTA is a weak base and decomposes to ammonia and formaldehyde. Because these reactions proceed slowly at 0°C, good mixing and drop formation result. The drops are then suspended in a hot organic liquid to increase the rates of chemical reactions so that gel spheres are formed.

The gel-sphere processes first described in technical literature were internal gelations used to prepare silica or aluminum silicate spheres for petroleum-cracking catalysts. Sodium silicate solutions were mixed with acids or acidic salts in concentrations selected so that the gelation would occur 0.3 to 5 min after mixing. The most widely studied sol-gel processes in recent years are internal gelation methods in which metal alkoxides (such as tetramethoxysilane) are hydrolyzed—that is, chemically decomposed by splitting chemical bonds and adding the elements of water. In these processes, the organic metal salts are dissolved in an organic solvent such as ethanol. Water is mixed with the solution to allow a slow, controlled hydrolysis of the metal salts to hydrous polymer chains that gel. Depending on the starting compositions and the treatments applied to the gel, many different carbide, oxide, and nitride ceramics can be prepared. The sintered products (ceramics heated to a coherent mass without melting) may be amorphous and glasslike or may be crystalline.

Sol and gel processes are not at thermodynamic equilibrium, nor are they thermodynamically reversible. Therefore, the development of the flow sheets is highly empirical, and small flow-sheet changes sometimes produce large, unexpected effects that are difficult to explain. Sam Clinton's earliest attempt to prepare thorium dioxide (ThO_2) spheres at ORNL went well for the first few tests. However, his later tests produced "junk"—namely, drops that coalesced or coated the equipment wall before gelling. After a number of ideas were tested and proven false, Clinton found that a surface-active compound was needed to get good results. Such compounds were leached from the connecting polymer tubes. When he had no fresh tubing in his experimental apparatus, he did not have enough surfactant, and the results were poor. After testing dozens of compounds, Clinton finally identified a surfactant (a sorbitan monooleate ester) that is particularly effective for preventing coalescence and sticking of sol drops or gel spheres. We still use it today without knowing why it is superior to other, chemically similar, surfactants.

Uses for Gel Processes

The earliest sol-gel work at ORNL was part of the Thorium Fuel Cycle Program. In the late 1950s ORNL researchers attempted to develop thorium blankets to breed fissionable uranium-233 (^{233}U) by absorbing neutrons in the Aqueous Homogeneous Breeder Reactor, under development at the

An ORNL sol-gel process exhibit and the persons who prepared it for the Third United Nations Conference on the Peaceful Uses of Atomic Energy in 1965 are shown here. From left in back are Jim Bresee, John Sease, Sam Clinton, Glen Williams, Gerry Pierce, Tom Gayle, Pete Lotts, and Charlie Angel; from left in front, Orlen Dean, Bob Brooksbank, Jim Snider, Paul Haas, and Fred Heddleson.

Information Exchanges on Sol-Gel Technology

Information on gel processes for nuclear fuel cycles was exchanged at a series of international conferences. The first sol-gel papers were presented at conferences for the thorium fuel cycle by Don Ferguson at Rome in 1961 and by Pete Lotts, Orlen Dean, and John McBride at Gatlinburg in 1962. Other organizations besides ORNL presented sol-gel papers at Gatlinburg in 1966.

A highlight for ORNL's early sol-gel researchers was the Laboratory's participation in the 1965 Third United Nations Conference on the Peaceful Uses of Atomic Energy in Geneva, Switzerland. Jim Snider was in Geneva for eight weeks setting up, operating, and taking down this exhibit illustrating ORNL work. At Turin, Italy, a 1967 conference on sol-gel processes for nuclear fuels included 15 papers, 4 of which were presented by Arnie Olsen and me. Larger symposiums on sol-gel processes for nuclear fuels were held in Vienna in 1968 and 1973.

ORNL's involvement in developing and applying sol-gel technology resulted in many exchange assignments between the Laboratory and other research organizations. Most visiting scientists worked at a host laboratory for one to two years, becoming intimately involved in its research. Karl Notz, Walt Bond, Ron Beatty, and Milt Lloyd were visiting scientists at the Swiss Federal Institute for Reactor Research (EIR). Two EIR scientists, Hans Beutler and Guido Ledergerber, conducted gel-sphere studies at ORNL. Information developed during these exchanges has led to the current ORNL program devoted to developing uranium and plutonium oxide fuels for DOE's Fast Breeder Reactor Program. Other exchange visitors at ORNL have included M. J. Bannister of Australia, A. P. Luina of Spain, Helmut Ringel of Germany, and Clarence Hardy from England. Jack McWherter of ORNL has been an exchange scientist in the Netherlands. Roy Norman first came to Oak Ridge to perform gel-sphere research while on assignment from the General Atomics Laboratories in San Diego; he is now a task leader for current ORNL gel-sphere studies.

Since the 1970s, the interest in nonnuclear applications has increased greatly, and technical programs show this change. Recent gel process conferences have been sponsored by the Materials Research Society (Boston in November 1981 and November 1982) and the U.S. Air Force (Gainesville, Florida, in February 1983).

In January 1984, ORNL's Power Systems Technology Program and DOE's Electric Energy Systems Division sponsored a technology-transfer workshop in Knoxville and Oak Ridge. According to Robert Lauf, who helped organize the workshop, at least two companies expressed interest in using sol-gel technology for making zinc oxide and other ceramic materials for electronic applications. He said that these companies are particularly interested in the ability of sol-gel technology to produce a uniform product.—*P. H.*.

Laboratory. Both thorium nitrate [$Th(NO_3)_4$] solutions and ThO_2 slurries were tried but were rejected because they either corroded and eroded the blanket structure, thus threatening its stability, or were unstable under blanket conditions. Thorium oxide sols were prepared but were not stable when subjected to neutron radiation at 280°C. It was therefore proposed that ThO_2 spheres, or "pebbles," be made and that heavy water be circulated through the "pebble bed" to slow down the neutrons and remove heat.

My first sol-gel assignment was a request by Don Ferguson in February 1960 to review the technical literature on commercial preparation procedures for catalyst pellets. I recommended that the procedures used for silica-alumina (SiO_2-Al_2O_3) gel beads be applied to preparation of ThO_2 spheres. The Houdry Process Corporation prepared ThO_2 spheres of high density using gel sphere processes but was unable to supply the larger 0.3-cm-diam spheres needed for the pebble bed blanket. Experience gained from these investigations was later applied to preparing other thorium fuels.

Under the kilorod program in the 1950s, the first sol-gel pilot plant at ORNL was operated by Claude Haws, John Sease, and coworkers. It fabricated 1000 nuclear fuel rods using vibratory compaction of a sol-gel product that was 97% ThO_2 and 3% uranium oxide ($^{233}UO_2$). The mixed sol was prepared from $Th(NO_3)_4$ and $UO_2(NO_3)_2$, gelled by evaporation of water; and then sintered, crushed, and milled to give three sizes of dense oxides. The three fractions were mixed in measured proportions and compacted by vibrating them in zirconium-alloy tubes to 85–89% of theoretical density.

Fuel elements for a thorium fuel cycle would be fabricated remotely because the reprocessing stream of ^{233}U and recycled thorium contain ^{232}U and ^{228}Th, which are too radioactive to handle by direct fabrication. The sol-gel and vibratory compaction are simpler and more suitable for remote fabrication than are the conventional preparation of ceramic ThO_2-UO_2 powders and fabrication of pellets. This type of gel process will probably be used again if future energy needs result in the use of a thorium fuel cycle.

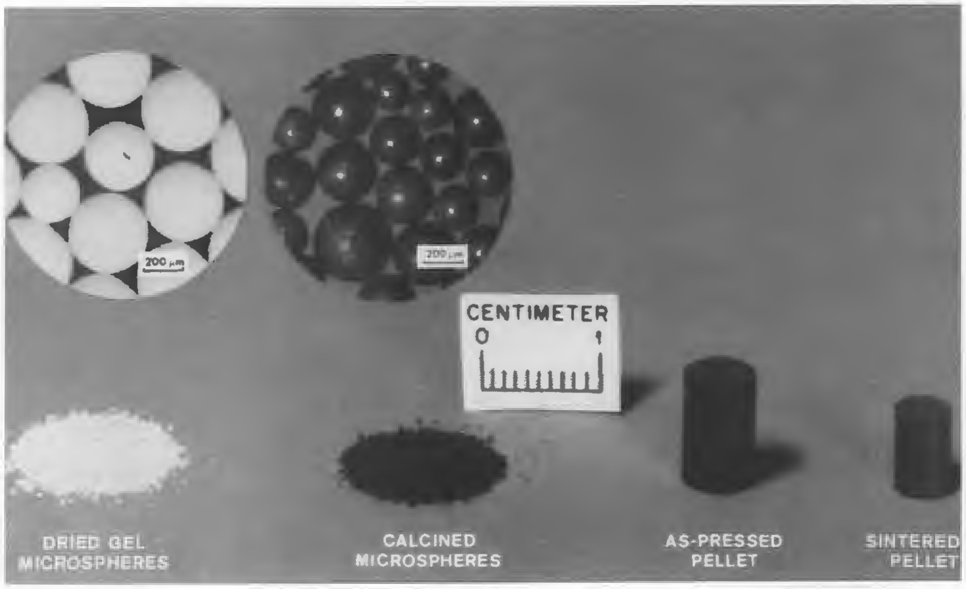
Because gel-sphere technology was needed in the development of fuel for high-temperature gas-cooled reactors (HTGRs), the national HTGR program supported about ten years of gel-sphere work at ORNL. The HTGR fuels are particles embedded in a graphite matrix. The spherical shape of the gels is important because the pyrolytic carbon and silicon carbide coatings act as miniature pressure vessels to contain fission products in HTGR fuels. Spherical coatings make much better pressure vessels than irregularly shaped coatings. Gel-sphere technology also makes possible homogeneous mixtures of thorium and uranium in the fuel pellets; such homogeneity is important to fuel performance and to reprocessing. In addition, the technology allows sinterability to high density and partial or complete conversion to carbides at much lower temperatures than are required for conventional ceramic processing using mixtures of powders, thus saving energy. Gel-sphere processes also can be adapted to remote operation, which is required for the safe fabrication of new, radioactive fuel from recycled thorium and ^{233}U. Gel-sphere processes have been used to make fuel for the HTGR now operating in Fort St. Vrain, Colorado, and may be employed more widely if orders for HTGRs resume.

How to dispose of radioactive wastes permanently and safely has been a challenge in recent years for nuclear-fuel-cycle specialists. One technical solution for underground burial uses gel-sphere processes to produce crystalline waste forms that resist leaching by groundwater. These crystalline spheres are ideal because they can be heavily coated to ensure containment of the fission products within. The HMTA internal gelation process was used at ORNL to prepare spheres of synthetic rock compositions and zirconium oxide (ZrO_2)-based compositions containing 20 to 90 wt % waste loadings. The sintered product spheres have excellent characteristics for waste disposal applications, but the practicality and economics of scaling up the production process have not been proven.

The need to protect nuclear materials from diversion or misuse (proliferation resistance) resulted in a major change in gel-sphere programs in 1977. One way to protect against diversion is to avoid separating highly purified fissionable materials. Coprocessing of uranium and plutonium and low enrichments of ^{235}U are examples of this approach to producing nuclear fuels that are not suitable for making bombs. Another way is to manage the fuel cycle so that such materials are always too radioactive for direct handling. Because gel-sphere processes can be used for remote refabrication, they are particularly suited for converting mixtures of fissionable and fertile materials to new fuel elements.

The Sphere-Pac process, developed by ORNL in the late 1960s as a joint effort of the Chemical Technology and Metals and Ceramics divisions, can be harnessed for remote refabrication of nuclear fuel, unlike the usual com-

ORNL has developed an internal gelation process (called the Direct Press Spheroidized Feed, or DIPRES process) for preparing fast-breeder reactor fuels. The process, which produces spheres of uranium and plutonium oxides that can be used as feed for making pellets, forms the basis for scaled-up sphere fabrication processes developed at the Battelle Pacific Northwest Laboratory and Hanford Engineering Development Laboratory.



mercial fabrication processes using ceramic powders to prepare pellets for fuel rods. Arnie Olsen and Bob Fitts, both of ORNL, and J. Komatsu, on assignment to ORNL from Japan, initially developed a fuel-fabrication procedure in which uniform large spheres are infiltrated first with medium spheres and then with small spheres.

This process, which received an I•R 100 award in 1979 from *Industrial Research & Development* as one of 100 innovative products or processes, uses three sizes of high-density spheres that can be loaded into the fuel cladding to 83–88% of the theoretical density of the fuel oxides or carbides. In 1977 ORNL's Chemical Technology Division prepared dense UO_2 spheres of 1200, 300, and 30 μm in diameter using the HMTA internal gelation process; and the Metals and Ceramics Division developed the fuel-rod-loading procedures to achieve simultaneous loading of the three size fractions.

About 100 kg of enriched UO_2 spheres was prepared for fabrication of irradiation specimens in the form of light-water reactor (LWR) fuel elements. Tests in which fuel rods containing UO_2 spheres were subjected to neutron radiation showed that these rods can survive higher burnups and do not suffer cladding failures as soon as LWR rods containing UO_2 pellets. The reason for this improved performance is that the spheres, unlike the pellets, exert reduced or more uniform forces on the fuel cladding during thermal cycles.

Exxon Nuclear Corporation has developed continuous pilot plant equipment for sphere preparation based on the ORNL work. Future applications of Sphere-Pac fabrication will depend on whether the irradiation tests now in progress yield favorable results.

Current gel-sphere studies at ORNL are dedicated to fabricating fast-breeder reactor (FBR) fuels made of spheres of $(UPu)O_x$ that can be used as feed for making pellets. At ORNL, $(UPu)O_x$ spheres are prepared by an HMTA internal gelation method, known as the Direct Press Spheroidized Feed (DIPRES) process. This ORNL process forms the basis for scaled-up sphere fabrication processes developed at the Battelle Pacific Northwest Laboratory and Hanford Engineering Development Laboratory (HEDL). The results to date indicate that the overall process is both technically and economically feasible. So far, the pellets fabricated by the scaled-up DIPRES process are of high quality and meet FBR specifications. The $(UPu)O_x$ spheres have a uniform composition and low concentrations of impurities and can be simply fabricated into fuel pellets. Furthermore, sphere fabrication produces less dust contamination than conventional processes and does not require the ball-milling, preslugging, and granulation operations commonly used for ceramic powders. We are continuing work on this process, with a goal of developing a second-generation process for fabricating FBR fuels in the Breeder Reprocessing Engineering Test to be located at HEDL's Fuels and Materials Examination Facility.

Nonnuclear Uses

Increasingly stringent requirements for ceramics and glasses have created an interest in sol-gel preparation processes for nonnuclear as well as nuclear uses. This growing interest is shown by the large number of sol-gel sessions at meetings of the American Ceramic Society, the Materials Research Society, and special conferences.

Ceramics and glasses are strong and hard at high temperatures but

Clinton pours uranium oxide spheres from a beaker as Haas looks on.

have had limited use as structural materials in high-temperature energy systems because of brittleness and a tendency to crack and break easily. However, Paul Becher, Robert Lauf, George Wei, and Walt Bond have recently applied sol-gel technology to make fine particles with microstructures that increase the ceramic's resistance to tensile stresses. As a result, these particles, when incorporated in the ceramic, absorb energy at crack tips and thus slow the growth of cracks when the material is stressed at high temperatures. They have used sol-gel technology to make powders for fabricating dense ceramics that are exceedingly tough. With this technology, the researchers can control the size and uniformity of zirconia, hafnia, and yttria particles used to toughen alumina. By using this technology, the ORNL ceramists have made alumina-zirconia composites that have a fracture toughness two to three times as high as that of alumina alone.

Sol-gel technology produces materials with desirable microstructures or microscopic properties that cannot be duplicated by conventionally produced ceramic powders. Among these desirable properties are homogeneous compositions, unique crystalline forms, better glass characteristics, and the ability to be processed at low temperatures without volatility or phase-separation problems. Such improved ceramics might be used for high-temperature structural materials (turbine blades, heat exchangers, or heat recuperators), electronic devices, optical glasses, coatings, films, or fibers. Ceramic gas turbines or diesel engines could operate at higher temperatures and higher efficiencies.

Compositions currently being studied at ORNL include pure compounds and mixtures of alumina, zirconia, silica, india, iron oxide, silicon carbide, silicon nitrate, titanium dioxide, tin oxide, and more complex ceramic compounds.

Led by Robert Lauf, ORNL researchers also have used sol-gel technology to make a more compact and versatile varistor. Varistors are ceramic surge arresters used in high-voltage switchyards, transformers, and transmission lines to short out electrical overloads resulting from lightning and other causes. The ORNL varistor is smaller than current commercial devices and can be more easily sited in congested urban areas. Because of its reduced size, the new varistor could also be used in underground transmission lines and energy-saving high-voltage cables insulated with gas.

To produce a smaller varistor for high-voltage applications, Lauf had to reduce the size of the ZnO_2 grains and its dopants so that a given volume of material has more grain boundaries. Using sol-gel technology, Bond produced extremely fine powders of ZnO_2 and the dopants—oxides of bismuth, cobalt, chromium, and antimony. Lauf used these gels to obtain grain sizes of about 3 μm, compared to 10–30 μm in current commercial varistors.

At least ten manufacturers are showing interest in the ZnO_2 varistor materials, according to Lauf. Representatives from companies such as GTE, McGraw-Edison, Ohio Brass, Raychem Corporation, and Westinghouse Electric Corporation, and from utilities such as Consolidated Edison attended a technology-transfer workshop held January 11-12 at the Knoxville Hilton Hotel and at ORNL. The workshop was sponsored by DOE's Electric Energy Systems Division and by ORNL's Power Systems Technology Program. At the conclusion of the conference several companies expressed interest in using sol-gel technology for making ZnO_2 grains for electronic applications.

Application of sol-gel processes to making toughened ceramics and improved varistors is empirical and somewhat of an art. If the product characteristics needed are carefully defined, then the ORNL experts can make important contributions to meeting nonnuclear needs. Indeed, the special powders that ORNL researchers make may solve some problems of tomorrow's power industry. ornl

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	Fur-Bearing Traffic Hazards: The Deer Problem	Barbara Lyon
	The Spin Spectrometer	Mel Halbert
	Energy Development and the Religious Freedom Act	Harry Arnold
1981 Winter	Scanning Electron Microscopy at ORNL: A Picture Story	
	Golden Days—or Brass? (relative energy prices)	Herb Inhaber
	Skimming the Surface (surface physics)	Mark Mostoller et al.
	Trends in Scientific and Technical Information	Carolyn Krause
	Pressel Vessel Integrity (thermal shock research)	Dick Cheverton
	Biomass into Energy: What Are the Environmental Effects?	Carolyn Krause
	CO_2 and Acid Rain: A Briefing at ORNL	Carolyn Krause
1981 Spring	State of the Laboratory–1980	Herman Postma
	Spiraling Down the Watershed	Jerry Elwood
	Artificial Intelligence Is Coming: Uses at ORNL	Carroll Johnson
	Health Risk Analysis: Science and Politics	Emily Copenhaver, Phil Walsh
1981 Summer	The Human Factor of Reactor Safety	Paul Haas, Carolyn Krause
	Reconstructing Past Climate	Carolyn Krause
	Putting It on the Line (UNIX at ORNL)	R. D. McCulloch
	Toward a Desirable Energy Future	Truman Anderson
	Genes and Cancer	Carolyn Krause

1981 Fall	Eastern Oil Shale and the Environment Personal Transportation of the Future Images of the Heart: Isotopes for Disease Detection Projected Man (robotics and teleoperations)	Jim McBrayer John Googin Carolyn Krause Mel Feldman
1982 Winter	ORNL's Quick Response Team: Analytical Chemists on Call Mining and Land Reclamation: ORNL and Abandoned Mines The New Hydrofracture Facility (nuclear waste disposal) Decommissioning Reactors Pocket Monitors for Airborne Pollutants	Carolyn Krause Jon Jefferson Barbara Lyon Robert Blumberg Carolyn Krause
1982 Spring	State of the Laboratory–1981 Iodine and Reactor Accidents	Herman Postma Carolyn Krause
1982 Summer	Energy Transitions Electricity: Faithful Servant at the Flip of a Switch Four Decades of ORNL Achievements	Truman Anderson Don Trauger Carolyn Krause
1982 Fall	Silver Recovery from ORNL Wastes UNISOR: The First Decade Minerals in the Wilderness: How Important Are They?	Carolyn Krause Russ Manning Jon Jefferson
1983 Winter	A Novel Way to Grow Anaerobic Bacteria Modeling Air Pollution Fuel Reprocessing: A Futuristic Look Hot Water District Heating for St. Paul Incentives for Hosting Nuclear Waste Repositories	Howard Adler Alan Witten Carolyn Krause Carolyn Krause Sam Carnes, Emily Copenhaver
1983 Spring	State of the Laboratory–1982 Life at the End of the Periodic Table Neutron Scattering: A Tool to Probe Biological Structures Safeguarding Reprocessing Plants	Herman Postma Richard Hahn V. R. Ramakrishnan Carolyn Krause
1983 Summer	Paradox of the Striped Bass: ORNL Fishes for Answers Cable-in-Conduit Superconductors Monoclonal Antibodies and Cancer Clean Water from Synfuels Plants The Staying Power of the HTGR Health Risks of Energy Technologies: The Experts' Views	Charles Coutant, Webb Van Winkle Lawrence Dresner Stephen Kennel Carolyn Krause Paul Kasten Carolyn Krause
1983 Fall	Special issue on materials research at ORNL, including development of new alloys, tougher ceramics, better graphites, more efficient solar cells, surface modification techniques, and solid-state and alloy theories.	30 ORNL authors

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Linda Spurling uses a net to catch a largemouth bass that has been stunned by an electroshocking probe held by Joe Malone. Wayne Parsons is driving the boat at Melton Hill Dam, where control studies are done. Gamma scans are performed on the fish samples to determine whether there is radioactive contamination in lakes and streams in ORNL's vicinity. See article "How Does ORNL Affect the Environment?" on page 2.