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Energy and Technology Review

Lawrence Livermore National Laboratory

December 1981

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About the Cover

This 200-m tunnel, built to house the accelerator and experimental area of the Advanced Test Accelerator (ATA), dwarfs the human figure inside it. The ATA Facility, now under construction at the Laboratory's Site 300 and scheduled to begin operating in the fall of 1982, will enable Laboratory researchers to test the behavior of intense electron beams in various gases and in the open atmosphere. In the photograph on the cover, you are looking down the tunnel in the direction the electron beam will travel as its energy is boosted to 50 MeV by an 85-m linear accelerator. In some future experiments, the beam will be directed into a gas-filled tank that will be installed in the far end of the tunnel, and in others it will be deflected into the open air. The ATA is described in more detail in the article beginning on p. 1 of this issue.

About the Journal

The Lawrence Livermore National Laboratory, operated by the University of California for the United States Department of Energy, was established in 1952 to conduct nuclear weapons research. Today, about 60% of our resources are devoted to defense programs, which include weapon and laser fusion research. Other major areas are: magnetic fusion energy; laser isotope separation; chemistry, engineering, and physics research; biomedical and environmental sciences; applied energy technology; and work for other Federal agencies. The *Energy and Technology Review* is published monthly to report on accomplishments in unclassified programs. A companion journal, the *Research Monthly*, reports on weapons research and other classified programs. Titles from recent issues of the *Energy and Technology Review* are listed opposite the inside back cover.

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Brief

ZAPP: A New Facility for Studying the Radiation from Hot, Dense Plasmas

We have recently completed construction of a z-pinch facility (ZAPP, for Z-pinch Atomic Physics Project) for detailed studies of the radiation emitted by very hot, dense plasmas. A z pinch

(z is the axial direction) is a cylindrical implosion caused by the self-induced magnetic field of a heavy current flowing through a plasma. Our research, which takes advantage of recent improvements in z-pinch design (J. Shiloh and J. Fisher, *Phys. Rev. Lett.* 40, 515, 1978), pulse power technology, and photon spectroscopy, centers on rapidly developing areas of atomic and plasma physics. At the same time, this facility provides a test stand for the development of new

Fig. 1

Gas-puff and electrode geometry of the ZAPP z-pinch device. A puff of gas released by a fast-acting valve enters the vacuum system through the annular slit centered in the flat surface of the lower electrode. The hollow cylinder of gas crosses the narrow gap to the ring-shaped upper electrode, which receives a high-voltage pulse timed to break down across the gap while the gas puff is still hollow. The heavy current from a capacitor bank induces a magnetic field that interacts with the current to produce a strong inward force. As long as the plasma is hollow, the forces opposing the implosion are reduced.

apparatus and techniques for plasma diagnostics.

At the ZAPP facility, we have already produced an argon plasma with an electron temperature (T_e) of 1 keV and density (n_e) of 10^{22} cm^{-3} , a regime that until recently has been very difficult to achieve in the laboratory. These conditions of density and temperature significantly affect the populations and lifetimes of the electronic states in the plasma. This regime is essentially new to experimental study, and it has possible applications in fields as diverse as astrophysics and laser fusion.

The ZAPP device consists of a large capacitor bank (52 kJ) for storing electric charge, fast electrical switches, and a vacuum chamber into which we can inject a hollow cylindrical puff of gas through an annular slit centered in the face of a flat electrode (Fig. 1). The gas crosses a gap of about 1 cm to a ring-shaped electrode, which is pulsed to 100 kV before the gas has time to fill the center of the puff. A heavy current from the capacitor bank ionizes the gas and then implodes it by interaction with its self-generated magnetic field. Since the gas puff is hollow, the forces opposing the implosion are reduced, and it can reach higher densities and temperatures than those in previous z-pinch devices.

Figure 2 is a pinhole x-ray image of the plasma, which can be formed with any gas or mixture of gases. The plasma column is about 2 cm long, and all radial views are unobstructed by opaque components. Thus, many spectroscopic and diagnostic experiments can view and measure it at the same time.

At present, we use a variety of techniques to obtain information on the plasma conditions and the atomic radiation in the ZAPP device (Table 1). Laser interferometry, electromagnetic probes, x-ray pinhole photographs, and a variety of photon spectrometers indicate the plasma density and temperature. The spectrometers detect the atomic radiation from the infrared to hard x-ray wavelengths, with very high resolution. Under development are laser-based Thomson scattering spectroscopy and time-resolved spectroscopy on a time scale of a few nanoseconds.

The research program emphasizes the study of highly excited atoms. We are measuring the atomic energy levels of the elements from boron to argon (atomic number $[Z]$ from 5 to 18). The helium-like series (ions retaining only two electrons) is of particular interest because these ions, which can readily be produced in the ZAPP plasma, participate in transitions that are useful for inferring plasma temperatures and densities. Future studies will concentrate on heavier atoms such as krypton and xenon, with emphasis on their neon-like ions (ions retaining only 10 electrons) and their use in plasma diagnostics.

Another area of interest involves quantum-electrodynamic effects on energy levels of high- Z systems. These systems have been studied with beams from ion accelerators, but these studies have been hampered by Doppler broadening of the emitted lines. Since the

Fig. 2

A pinhole x-ray image of an imploded argon plasma in the ZAPP device. The plasma, originally hollow and about as broad as it is long, breaks into a series of hot spots (sources of intense x-ray emission) as a result of instabilities associated with the implosion mechanism.

Table 1

Characteristics of diagnostic equipment used to evaluate the density and temperature of plasmas produced in the ZAPP device.^a Spectrometer resolutions ($\Delta\lambda/\lambda$) are typically 10^{-3} to 10^{-5} . Spatial resolutions ($\Delta l/l$) are about 10^{-2} .

Spectrometer	Range				
	Infra-red	Visible	Ultra-violet	X rays	
				Soft	Hard
Normal incidence	-----				
Grazing incidence			-----		
Crystal				-----	
Pinhole photography			-----		
Ruby laser measurements	Quantity measured ^b				
Interferometry		n_e			
Schlieren photography		∇n_e			
Thomson scattering (FY 1982)		n_e, T_e, T_i			

^aWe expect during FY 1982 to use microchannel plates to provide a time resolution of 1 ns for most of diagnostics.

^bOf the quantities listed, n_e is the electron density, T_e is the electron temperature, and T_i is the ion temperature.

ions in the ZAPP device are relatively slow, having only thermal motions, the Doppler shifts should be much smaller. This study is longer ranging and is essential to our understanding of the structure of highly ionized high-Z elements.

Because the ZAPP device operates with extremely short pulses and its plasma is in a violent nonequilibrium state, our results should be applicable to a wide range of transient atomic processes that are not characterized by local thermodynamic equilibrium, such as those in the

explosion of a nuclear device. Our research may help us understand the radiation from stellar atmospheres and the radiative behavior of laser-induced plasmas. (The z-pinch approach is complementary in many ways to producing plasmas by lasers.) Finally, this technology may find application in x-ray-detector development and x-ray lithography or holography or in a facility for calibrating pulsed-x-ray detectors.

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Generating Intense Electron Beams for Military Applications

The ATA Facility we are building at Site 300 will allow us to determine the feasibility of propagating intense, self-focused electron beams in the open air and to answer questions about their potential military applications.

Although charged-particle beams represent a potential breakthrough in military capability, their future use as weapons depends on the feasibility of propagating an intense, electro-

magnetically self-focused electron beam through the atmosphere. LLNL researchers hope to determine this by conducting a comprehensive program of electron-beam propagation experiments

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in a 50-MeV Advanced Test Accelerator Facility (the ATA)¹ that we are constructing at our high-explosives test location, Site 300. This facility, the successor to LLNL's Astron II and Experimental Test Accelerator (ETA), will cost about \$50 million and should be completed by the fall of 1982.

The ATA, together with its associated program of beam-propagation physics, represents the largest single component of the Particle-Beam Technology Program conducted by the Defense Advanced Research Projects Agency (DARPA). This program was formerly sponsored by the U.S. Navy, and the Naval Surface Weapons Center is now the direct funding agency for the DARPA program. The aim of this program of research and exploratory development is to resolve those scientific issues necessary to show that particle-beam weapons are possible. The prime goal of the DARPA Particle-Beam Technology Program is to resolve what is and is not possible in beam propagation. Accordingly, our goal with the ATA is to develop an experimental capability that can answer critical questions about beam-propagation physics in a timely and cost-effective fashion.

Although we can study many aspects of beam stability and propagation in a shielded experimental gas-filled tank, it is

essential to test particle beams in open air to determine how the beam will propagate in various natural environments. In addition, we must be able to fire the beam in a range of directions to test methods of pointing and tracking. Finally, an outdoor facility will allow us to detonate safely substantial quantities of chemical explosives in target-damage studies.

Potential hazards associated with the ATA experiments were considered in choosing our site. LLNL's Site 300 is located in a remote area 25 km southeast of LLNL that is well equipped for managing such experiments. The ATA is being built in a shallow valley to exploit natural shielding. Figure 1 shows the ATA facility under construction.

Applications of Intense Beams

Recent advances in pulsed-power technology, notably the development of repetition-rated electrical components (i.e., ones capable of delivering short pulses of high-voltage, high-current electrical power many times per second), have made feasible a number of high-technology uses for intense electron beams. Of the wide variety of possible applications, flash radiography and nuclear effects simulation have considerable military significance. In their application in the free-electron laser, such

Fig. 1

ATA facility under construction at Site 300. The large corrugated metal tube (a) enclosing the experimental tank—exposed when this photograph was taken in April 1981—is now (b) covered with earth and concrete. The facility is scheduled to begin operation in the fall of 1982.

beams have attracted considerable interest in the scientific and defense communities. The military application that has received the most public attention is the use of intense beams of charged particles as point-defense weapons.

If it proves feasible to propagate charged-particle beams, their first use as weapons will be largely against targets within a few kilometres or less. Since they are so lethal, charged-particle beams can be used at distances too close to allow time for the "second shot" that might be necessary with less effective weapons. Early uses of the beams might be to defend large ships from cruise missiles² or for the close-range defense of hardened sites such as missile silos or national command authority centers. Such missions are called "point-defense" missions in distinction to area-defense missions.

Although the beam must bore its way through the air to the target, this process takes at most a few thousandths of a second. The deposition of megajoules of energy in the target is almost instantaneous. Therefore, beam weapons have the potential for engaging tens of targets per second, depending upon certain constraints in the system that controls their firing. These characteristics make beam weapons particularly well suited for countering small, very fast, highly maneuverable threats.

Accelerators have the demonstrated capability to convert upward of 30% of their prime electrical power into beam energy. If the electricity is produced with conventional generators powered with jet fuel, megajoules of beam energy can be produced with the consumption of approximately 10 litres of fuel. Consequently, even modest fuel supplies translate into a very large reserve of "ammunition" for a particle-beam weapon. This reserve makes it extremely difficult to overwhelm the particle beam with a large number of incoming threats.

Upon striking a target, the beam penetrates deeply and deposits its energy in a long, narrow cone. The high-energy electrons in the beam can penetrate tens of centimetres of solid aluminum, making it very difficult to shield against them. Damage to the target is immediate and severe; it includes structural damage,

destruction or disruption of electronics equipment (e.g., missile guidance systems), and nearly instantaneous detonation of chemical explosives. An example of the damage a charged-particle beam of small radius can inflict on a target is shown in Fig. 2.

Not all the energy in a pulse reaches the target; some of it is lost in the atmosphere at a rate roughly proportional to the density of the air. Under normal conditions, the pulse will lose half its energy after traveling about 200 m. However, this loss rate does not limit the beam's range as much as might be expected. Much of the energy lost from the pulse goes into heating the air along the path of the beam. In a few microseconds, this hot air expands, leaving a channel of much lower density that the next pulse can follow with minimal energy loss. The use of bolts consisting of strings of pulses may allow propagation over long distances.

The electrons in the pulses scatter as they pass through the air, and the beam gradually widens. Because this spreading reduces the beam's power density on the target, the beam is useless as a weapon unless the spreading is inhibited. The large currents in the beam wrap it in a strong magnetic field (proportional to the current) that limits spreading by pinching the electrons closer together. This self-focusing occurs only when a high-current

Fig. 2

An example of the damage charged-particle beams can inflict. This aluminum disk (1 cm thick and 16 cm in diameter) was hit by an electron beam with about the same energy content but one-fifth the particle energy of an ATA beam pulse. The beam penetrated about 0.6 cm into the disk, melting the metal. An intense shock wave traveled through the disk, ripping away chunks of metal when it reached the rear surface (shown in figure). Photograph courtesy of Air Force Weapons Laboratory, Kirtland Air Force Base, New Mexico.

Table 1 Comparison of ATA with earlier induction accelerators.

	Astron II ^a	ERA ^b	ETA ^c	ATA ^d
Beam energy, MeV	6	4	4.5	50
Current, kA	0.8	1.2	10	10
Pulse length, ns	300	30	40	70
Burst rate, Hz	800	2	1000	1000
Average rate, Hz	5	2	5	5

^a Lawrence Livermore National Laboratory, about 1968–1976.

^b Electron Ring Accelerator (Lawrence Berkeley Laboratory).

^c Experimental Test Accelerator (Lawrence Livermore National Laboratory).

^d Advanced Test Accelerator (Lawrence Livermore National Laboratory).

beam is traveling through the air or some other gas. Self-focusing keeps the beam diameter down to a centimetre or so in air. When the focused beam hits the target, it deposits large quantities of energy in a small volume. The high-energy density makes the beam lethal.

The ATA's Role in Particle-Beam Research

The particle energy in a beam weapon determines how deeply the beam will penetrate the target. Although the ATA's energy level of 50 MeV will not cause beam penetrations as deep as more energetic beam weapons would, the ATA should provide data applicable to the designing of beam weapons. The ATA should enable us also to measure the radiation cone that extends far from the beam itself. (This radiation cone can cause significant damage to electronics equipment, thus increasing the effective range of a particle-beam weapon.) In both cases, scaling laws must be applied if we

extrapolate our data to particle-beam weapons. Regardless of how our findings finally are applied, many complex weaponization issues will arise in the transition from the ATA to beam weapons.

Even though the particle energies of the ATA facility are well below those projected for beam weapons, we should be able, with high confidence, to extrapolate positive test results to increased energies. This close programmatic link between theory and experiment makes the ATA a cost-effective means for studying the physics of particle-beam weapons.

The ATA represents several large advances in high intensity accelerator technology. The beam characteristics projected for the ATA (10-kA current, 50-MeV particle energy, 70-ns FWHM pulse, and 1-kHz repetition rate) are far beyond the capabilities of any existing accelerator. Table 1 compares the parameters of the ATA with those of the Astron II, the ETA, and the Lawrence Berkeley Laboratory's electron ring accelerator (ERA) injector.

In our experiments, we would like to test charged-particle beams over as wide a range of physical parameters as possible. The advantages to be gained from such exhaustive testing must, of course, be weighed against both the costs and the physical limits of the accelerator.

The ATA will enable us to determine the conditions under which stable and controllable beams can be propagated in open air. We can also test the beam's capability for inflicting damage and the performance of the nozzles that steer or aim it. As part of our effort to design a system for pointing and tracking, we will measure the degree to which microwaves or optical means can detect the beam path.

The particle energy of the ATA will be ten times that produced by the ETA. This increased particle energy means the ATA will have an order of magnitude more accelerator modules than the ETA, placing great demands on hardware reliability. The large number of components also makes it necessary to keep accelerator instabilities in mind when designing the beam-transport unit,

because the focusing requirements are unusually stringent.

To ensure this high level of component reliability, we are using the ETA to test ATA technology on a small scale. The ETA will be used to verify the projected performance levels of the electron source, the 1-kHz pulsed-power components, and the beam-control system for the ATA. Since its initial operation in mid-1979, the ETA has achieved all design goals and has produced several million beam pulses. The ETA, therefore, has provided the necessary data base for design of high-voltage components for the ATA, which will produce a few

million beam pulses per year in normal operation.

The ATA Design

The 200-m ATA Facility has an 85-m linear electron accelerator and consists of four major units: a pulse-forming network, an electron injector, a series of accelerator modules, and an experiment tank (see Fig. 3). The pulse-forming network provides short, high-voltage pulses that power the electron injector and accelerator modules. The injector produces a 10-kA beam of 2.5-MeV electrons, which are guided by magnetic

Fig. 3

Sketch of the ATA facility showing arrangement of major components: the pulsed-power network, the electron injector, the accelerator, and the shielded, gas-filled experiment tank. (Many of the vertical elements forming the pulsed-power network have been omitted for simplicity.) This whole assemblage will be 200 m long. The cross-section of the accelerator shows the end of one accelerator cavity, located in an underground tunnel, as well as the Blumleins connecting it to the pulsed-power transformers above ground.

fields through an accelerator consisting of 190 separate accelerator modules (cavities). The accelerator increases the electron energy to 50 MeV in 190 separate increments of 0.25 MeV. When they are at full energy, the electrons, still guided by magnetic fields, pass into an experiment tank that contains gas of variable type and pressure and that is covered with a thick layer of earth and concrete to absorb any stray radiation.

Pulsed-Power Network

The pulsed electrical power is provided by a pulse-forming network (Fig. 4) that stores a discrete amount of energy over a period of time in order to compress it and then suddenly releases it as a pulse. The power is stored in components such as an electrical transmission line that is charged to a desired voltage in much the same manner as a capacitor. A transmission line (Blumlein) composed of three concentric, metal conducting cylinders is frequently used in pulsed-electron accelerators.

Figure 4 shows how energy is converted to the beam from primary ac power. The ac power drives a high-voltage dc power supply which, in turn, feeds an energy storage and charging network. The pulse from this network is stepped up to 250 kV by a resonant transformer bolted to the Blumlein transmission line. When the current that has charged the Blumlein drops to zero, a spark-gap switch (filled with gas at high pressure) is fired by a 150-kV trigger circuit. The resulting high-voltage discharge from the Blumlein is then impressed across the accelerating gap. As long as this pulse persists, electrons passing by the gap will be accelerated. The energy stored in the Blumlein in 10 μ s is delivered to the acceleration cavity in 70 ns, almost 150 times faster.

The Blumlein can be recharged (by having an insulating gas flow through the spark-gap switch at high velocity) and then fired again at a 1-kHz burst rate. The voltage-holding ability of this switch (developed for ATA) determines the maximum accelerating voltage per ATA module. Because of limitations in the energy-storage subsystem, the spark gap

is fired in bursts of ten consecutive pulses with two-second rests between bursts.

Electron Injector

The source of the high-current electron beam is the electron injector, or gun. The ATA's 2.5-MeV electron injector, the second major unit in the accelerator, is typical of the pulsed-power technology used throughout the system. Acceleration voltages in both the electron injector and the accelerator module are generated by similar power-supply and pulse-forming networks feeding the ferrite induction cores.

Our coordinated program of theory and experiments with the ATA has the goal of providing a complete understanding of beam-propagation physics.

The 10-kA current produced by the injector requires special design features to control the defocusing effects of the electromagnetic fields the beam itself generates. These fields lead to a net force that is the difference between the electrostatic self-repulsion of the electron cluster and the magnetic pinch forces that tend to hold the beam together. For beams with kinetic energy greater than their rest mass energy, these forces nearly cancel

(the net repulsive force is proportional to the inverse square of the beam energy).

Figure 5 is a schematic cross section of the ATA's electron injector. Here (as in a conventional vacuum tube) a high-voltage pulse is applied to a grid to extract the required current from the cathode. In the vicinity of the grid, the repulsive self-fields of the beam are shorted by the grid; this causes an initial focusing of the beam (as Fig. 5 shows). When the grid pulses, the anode is pulsed simultaneously to 2.5 MV, rapidly raising the kinetic energy of the electrons as they traverse the region between the grid and anode. Although the defocusing forces are reduced by the increase in beam energy, solenoidal coils are needed to control and focus the beam for transport through the accelerator module.

The electron injector is constructed in two parts, each with five 0.25-MV induction units in series to provide the 2.5-MV anode/cathode voltage pulse. This voltage also appears across two large ceramic accelerator columns that are

divided by metallic rings into ten segments. The segments are connected by power resistors to ensure proper voltage distribution across the column to avoid electrical breakdown. The accelerator columns also serve as the barrier between the vacuum in the beam area and the dielectric oil that fills the induction units. The division of the injector into two parts has several advantages: it provides a vacuum pumping port in the center of the injector, partially shields its ceramic insulators from the electron beam, and makes assembly easier.

The Accelerator Module

The accelerator module is the heart of a multistage accelerator like the ATA. Each accelerator module adds an incremental kinetic energy to the beam. Therefore, by increasing the number of modules that make up the accelerator, one can increase the beam energy to any desired level. In a qualitative sense, the linear induction accelerator is just a series of one-to-one pulse transformers. The primary circuit

Fig. 5

A schematic drawing of the 2.5-MeV electron injector for the ATA. The 2.5 MV between the anode and cathode (provided by 10 induction accelerator units of 0.25 MV each, connected in series) extracts 10 kA from the large cathode. Note that the beam begins to spread soon after

it enters the anode but that focusing coils quickly focus it to a smaller diameter before it is injected into the accelerator. The beam's spreading and focusing is drawn here according to precise calculations of actual beam behavior.

of the transformer is the pulse-forming network previously described. The secondary circuit of the transformer is the electron beam itself. As Fig. 6a illustrates, the primary circuit loops around the magnetic core once, just as the electron beam threads through each core once. In such a transformer, the voltage induced in the secondary circuit is just that in the primary circuit.

The acceleration process can be described more specifically (Fig. 6b). Before the beam enters each cavity, the ferromagnetic cores are magnetized (set) to a maximum magnetic field. Then, a voltage pulse from the coaxial transmission line is impressed upon the accelera-

tion gap. The ferrite torus acts as an inductance, initially preventing a large current from flowing through the structure around the ferrite, thereby keeping the coaxial line from shorting. In accord with the law of magnetic induction, this current increases at a steady rate given by the ratio of the voltage pulse to the ferrite inductance. In addition, the magnetic field in the core decreases from its initial value at a constant rate until the field attains a minimum value. At this time, the applied voltage pulse ends. In addition to the time-varying fields that accelerate the electron cluster, there are static magnetic fields that guide the beam as it moves down the accelerator from one cavity to

Fig. 6

Two diagrams of the accelerator modules in the ATA. (a) Generalized representation of the relationship of the two circuits in the accelerator, the electron beam and the pulsed power that drives it through the ferrite cores. (b) A more specific drawing (longitudinal section) of the accelerator module. This structure is essentially a long metal tube consisting of a series of chambers, or cavities, containing ferrite rings that prevent the current in the coaxial line from shorting. Blumlein transmission lines deliver a high-voltage pulse to each cavity just as one of the electron clusters that make up the beam reaches the cavity. The electron clusters thus pass from one cavity to the next, increasing in momentum each time, until they attain an energy of 50 MeV.

the next. A model of a section of the accelerator is shown in Fig. 7.

In induction accelerators, it is important that the voltage pulse to the acceleration cavity coincide with the arrival of one of the electron clusters that are passing through the accelerator module. Proper timing is ensured by appropriate delays in the triggering circuits of the pulsed-power network.

An important complication in any electron accelerator is that the traveling

electron clusters generate electromagnetic fields that are modified by the accelerator structure. This coupling can distort the accelerating fields and limit the output current. It can also cause small, random perturbations in the beam's position or structure that lead to damaging or current-limiting instabilities. Because the beam fields increase with the beam current, these perturbations are especially serious in high-current accelerators. Furthermore, the current-limiting

Fig. 7

Model of a section of the ATA's accelerator and its pulse-forming power supply. This model represents a structure approximately 16 m high. The components below ground level are embedded in a concrete arch over the tunnel area. The components above ground can be raised for inspection by means of a crane attached to the I-beams.

instabilities grow exponentially with the number of accelerator cavities. The unusually high beam current of the ATA and the ETA necessitates the choice of high-voltage accelerator modules with toroidal ferrite cores rather than lower-voltage, soft iron units such as were used on the Astron. This choice reduces the number of accelerator structures that can interact with the electron beam, suppressing potential instabilities.

Experimental Facility

Once the electrons have acquired their final energy, 50 MeV, they are guided by a series of steering and focusing magnets into a large tank that can be filled with gas of various composition and pressure. This tank is located in an 80-m-long underground tunnel (Fig. 8).

The first experiments to be performed after the accelerator construction is completed will be measurements of beam current and voltage waveform and their

variation on a pulse-to-pulse basis. We will measure also the spatial distribution and angular divergence of the beam. When we understand the nature of the beam and its pulse-to-pulse reproducibility, we will begin studies of beam stability and dynamics as well as beam interactions with gas and plasma.

For all such studies, we require accurate measurements. Line-of-sight holes from the ground surface into the tunnel provide a means of removing sensitive diagnostic equipment from the intense radiation environment the ATA beam will produce. A diagnostic bunker near the entrance to the tunnel will accommodate the fast diagnostics needed to study the physics of beam/gas interactions. Both the diagnostics for beam/gas interaction studies and the specially designed voltage and current probes will be monitored by the ATA control system to provide accurate and complete records of all experiments.

Fig. 8

The completed ATA facility will permit experiments on beam behavior both in a shielded gas tank and in open air. The artist's sketch shows the underground experiment tank into which the beam will be directed. The thick earth cover will absorb stray radiation, and the pipes from the ground surface down to the tunnel will allow access for diagnostic instruments. The instruments can be monitored from the shielded bunker above the tank. To the left of the tank, a door can be opened to allow the beam to be aimed into the atmosphere. In the photograph (taken in October 1981), the concrete door has been rolled aside.

Conclusion

Our coordinated program of theory and experiments with the ATA has the goal of providing a complete understanding of beam-propagation physics. Our DARPA-funded program in particle-beam technology has yielded a body of knowledge about high-intensity accelerators and beam physics that we can build upon with the ATA. We expect to obtain high-quality, high-current electron beams with energies at least an order of magnitude greater than any other repetition-rated accelerator has produced. The ATA promises advances in high-intensity accelerator technology that are essential to a wide range of applications, including beam weapons. ☐

Key Words: accelerator; advanced test accelerator; ATA; charged-particle-beam weapon; electron accelerator; electron-beam propagation; experimental test accelerator; ETA; linear accelerator; particle beam.

Notes and References

1. For information on the dedication of the Advanced Test Accelerator, see *Energy and Technology Review* (UCRL 52000-81-1), January 1981, p. ii.
2. W. E. Wright, "Charged Particle Beams. Could We? Should We?," *Proceedings of the U.S. Naval Institute*, U.S. Naval Institute Press, Annapolis, MD (November 1979).

SYNROC: Permanent Radioactive-Waste Storage

We are developing a synthetic rock whose loading capacity and resistance to leaching and radiation damage, vital for proper performance in immobilizing high-level nuclear waste (radwaste), surpass those of borosilicate glass.

The long-term storage of high-level reactor waste poses a major problem for the defense industry today and a future problem for the civil power sector when commercial reprocessing of spent fuel begins. Work done to solve the defense-waste problem will apply to the eventual commercial-waste problem.

High-level waste contains virtually all the fission products and most of the actinides not separated during reprocessing. Current plans call for immobilizing this waste in borosilicate glass and then burying the glass in an underground repository. Although this process is well developed from an engineering standpoint, several alternative waste forms have better performance properties than those of borosilicate glass.

One of these alternatives is SYNROC, a synthetic mixture of minerals that mimics the way certain natural rocks immobilize radwaste-like elements over geologically significant time spans. The SYNROC concept was developed in 1978 by A. E. Ringwood,^{1,2} of the Australian National University, for storing waste from commercial nuclear power production. At LLNL, we are extending the initial SYNROC concept (called

SYNROC C, for civil) to include storage of high-level waste from defense-related activities (SYNROC D, for defense).^{3,4}

We will examine here the various types of waste, the SYNROC concept as applied to those wastes, and SYNROC fabrication and performance testing.

High-Level Nuclear Waste in the United States

Table 1 summarizes the major categories of nuclear waste and their primary sources,^{5,6} and Table 2 lists the quantities and types of high-level waste on hand in 1980. Policy until October 8, 1981, prohibited commercial reprocessing; on that date, President Reagan directed that the indefinite ban on commercial processing be lifted. As a consequence of the ban, however, nearly all of the high-level liquid waste currently in storage came from the reprocessing of spent fuel assemblies originating from defense activities. Spent commercial fuel assemblies are being stored intact, for the present.

High-level defense wastes are generated in the production of plutonium and tritium for nuclear weapons and in

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Table 1 Sources of major classes of nuclear waste (see Refs. 5 and 6).

Class	Source(s)
High-level waste	<ul style="list-style-type: none"> Spent fuel assemblies Reprocessing spent fuel
Transuranic waste (>10 nCi/g transuranic material)	<ul style="list-style-type: none"> Reprocessing spent fuel Plutonium fabrication
Low-level waste (<10 nCi/g transuranic material)	<ul style="list-style-type: none"> Reactor operation Fuel fabrication
Uranium mine/mill tailings	<ul style="list-style-type: none"> Uranium mining
Gaseous effluent	<ul style="list-style-type: none"> Reactor operations Reprocessing spent fuel

Table 2 Approximate quantities of high-level waste (1980).

Source	Quantity
High-level waste from reprocessing ^a	
Commercial, m ³	2 500
Defense, m ³	286 000
Spent fuel from commercial reactors, Mg	5 700

^a One cubic metre of water has a mass of 1 Mg, or 1 metric tonne.

Table 3 Quantities of U.S. high-level defense waste (1980).

Source	Form of Waste	Volume, m ³	Activity, MCi ^d
Savannah River Plant	Salt/sludge/liquor	94 600	700
Hanford Plant	Salt/sludge/liquor	181 700	220 ^b
Idaho Chemical Processing Plant	Acid liquor ^c	9 460	17

^a The official SI unit of radioactivity is the becquerel (Bq), defined as the activity that produces an average disintegration rate of 1 s^{-1} . This unit is inconveniently small for expressing the activities routinely encountered in waste management. For example, $1 \text{ nCi} = 37 \text{ Bq}$ and $1 \text{ MCi} = 37 \text{ PBq}$ or $37 \times 10^{15} \text{ s}^{-1}$.

^b An additional 335 MCi are stored as separated strontium-90 and cesium-137.

^c Another 2000 m³ (36 MCi) of Idaho liquid wastes have already been calcined into granular solids.

Table 4 Major components (expressed as oxides) in Savannah River Plant waste sludge (see Ref. 8). The columns labeled "high iron" and "high aluminum" represent the compositional extremes that might be expected during processing of the sludge into a solid-waste form (e.g., SYNROC). The "composite" composition is an average of the inventory at the Savannah River site.

Component	High iron, wt%	Composite, wt%	High aluminum, wt%
Fe ₂ O ₃	53.17	36.13	5.32
Al ₂ O ₃	4.89	28.26	76.05
MnO ₂	3.56	9.94	4.37
U ₃ O ₈	12.34	3.26	1.28
CaO	3.62	2.69	0.35
NiO	9.08	4.47	0.78
SiO ₂	0.40	0.85	0.56
Na ₂ O	4.52	5.08	1.96
Na ₂ SO ₄	0.50	0.93	0.50
Ion-Siv IE-95 ^a	8.82	8.93	9.33

^a Mixture of $\text{CaAl}_2\text{Si}_4\text{O}_{12} \cdot 6\text{H}_2\text{O}$ and $(\text{NaKC})_3\text{Al}_3\text{SiO}_{24} \cdot 8\text{H}_2\text{O}$.

the processing of spent naval reactor fuels. These wastes are presently in tank storage at the Hanford site near Richland, Washington, the Savannah River Plant near Aiken, South Carolina, and the Idaho Chemical Processing Plant near Idaho Falls, Idaho. The current total inventories are summarized in Table 3.⁷

The Savannah River Plant Defense Wastes

The first high-level defense waste that will be immobilized is that in storage at the Savannah River Plant. Defense waste in tank storage at this facility consists of three components—sludge, salt cake, and supernatant liquor (Fig. 1). The sludge precipitates from the acid waste stream upon neutralization with sodium hydroxide, carrying with it large fractions of the radionuclides present (except cesium, which remains in solution in the supernatant liquor).

The supernatant liquor is further concentrated by evaporation. Upon cooling, salts (mostly nonradioactive) precipitate from the resulting concentrated solution to form the salt layer or "salt cake" on top of the sludge.

The sludge from this waste, consisting of about 95% fluid and 5% solids, looks and handles like black axle grease, and is chemically diverse. Table 4 lists the composition of sludges from specific tanks that have order-of-magnitude differences in the major elements, i.e., iron and aluminum.⁸ Also listed is the composite

or average composition for the total inventory. Minor-element chemistry is also varied and diverse. A matrix intended to immobilize defense wastes must have the flexibility to incorporate these extreme differences in sludge composition.

The principal radionuclides in the waste are cesium-137 and strontium-90, followed by ruthenium and europium.^{8,9} Almost all of the cesium-137 is in the supernatant liquid and almost all of the strontium is in the sludge. Alpha activity (indicative of actinide-element content) is quite low (Table 5).

The SYNROC Concept

As far back as the early 1950s, it was suggested that radioactive waste could be put into a synthetic mineral form. Ringwood's SYNROC concept is based on the extremely insoluble titanate minerals and incorporates the radionuclides into these host minerals as dilute solid solutions.

The host minerals in SYNROC C have the structure of zirconolite, perovskite, and hollandite (Fig. 2). Natural analogues of these minerals exist. Many of them contain uranium and thorium and have retained these elements in their crystal structures for up to 500 million years.¹⁰

Defense wastes differ chemically from commercial wastes (because of differences in fuel reprocessing steps) and so require a different SYNROC composi-

Radionuclide	Activity, ^a mCi/g			
	Tank 5	Tank 7	Tank 13	Tank 15
Strontium-90	74.7	27.0	15.5	25.6
Cerium-144	4.8	0.2	2.0	16.9
Ruthenium-106	2.7	1.4	0.4	1.7
Europium-154	0.5	—	0.3	1.2
Cesium-137	1.3	1.3	0.3	0.1
Gross alpha	0.1	0.1	0.3	0.1

^a See footnote a, Table 3.

tion. In our modified composition (SYNROC D), such nonradioactive elements as iron, aluminum, and manganese, which are present in large quantities in the defense sludge, form an inert spinel mineral phase (Fig. 2). Second, because of the presence of sodium and silicon in the defense waste, we incorporate cesium into a nepheline rather than a hollandite host. The radionuclides other than cesium substitute for various cations in the zirconolite and perovskite minerals (Fig. 2).

SYNROC Fabrication

On a laboratory scale, SYNROC synthesis is straightforward.⁴ The additives required to make the final minerals

Fig. 1

Schematic view of a Savannah River Plant high-level defense waste storage tank, showing the three major waste components: supernatant liquid, salt cake, and sludge. The sludge layer is composed mainly of hydroxides, formed when the acid-waste stream from the reprocessing facility is neutralized with sodium hydroxide before being put into tanks. To reduce the volume of liquid waste stored, the supernatant liquid is drawn off, heated to evaporate some of the water, and returned to the tanks. As this highly concentrated supernatant solution cools, a low-activity layer of salt crystallizes out on top of the sludge.

All the radionuclides, with the exception of cesium, are strongly partitioned into the sludge. Cesium is concentrated in the supernatant liquid.

Fig. 2

Crystal structures of the minerals in SYNROC. SYNROC C (containing waste from commercial reactors) consists of zirconolite, perovskite, and hollandite. SYNROC D (containing wastes from defense reactors) is composed of zirconolite, perovskite, spinel, and nepheline. The spinel phase accommodates the large concentration of nonradioactive transition metals (iron, aluminum, manganese, and nickel) in the defense sludge.

Because of the presence of sodium and silicon in the Savannah River wastes, we have chosen to incorporate cesium in nepheline rather than hollandite. The fission products such as strontium and the rare-earth elements (REE) are immobilized primarily in perovskite and the actinides primarily in zirconolite.

(TiO_2 , ZrO_2 , CaO , and Al_2O_3) are combined with a water slurry of the sludge and then mechanically mixed. The resulting aqueous slurry is sprayed into a heated air stream that evaporates the water, leaving a free-flowing powder composed primarily of the SYNROC additives and hydroxide and nitrate salts of the waste sludge.

Before SYNROC can be made from this powder, the salts must be decomposed to metal oxides (CaO , U_3O_8 , Fe_2O_3 , Al_2O_3 , etc.) by calcination at temperatures in the range of about 600 to 800°C. Finally, the powder is "mineralized," that is, it is reacted at an elevated temperature (1100°C) and pressure (14 MPa) to form the desired SYNROC mineral phases. This step locks the radionuclides into the crystal structure.

On a laboratory scale, the mineralization step takes place in a conventional ceramic hot press. The calcined powder is loaded into a graphite die, heated to the desired temperature, and compressed between two axially opposed rams. This method forms the SYNROC into a large disk (Fig. 3). To date, we have produced high-density SYNROC disks up to 15 cm in diameter and 2.5 cm thick. We are currently investigating scaling these laboratory operations to determine the feasibility of large-scale production.

Figure 4 summarizes the composition of the oxide starting materials and the final mineral phases formed during the mineralization step. The chart is color coded to distinguish between the types of starting material (i.e., SYNROC additives or high level waste) and the mineral phases formed (inert vs radionuclide hosts). The mineral grain sizes in SYNROC are mostly less than 1 to 2 μm in diameter, giving SYNROC excellent mechanical strength.

We use conventional geochemical tools to verify the composition of the SYNROC material, determining the mineral phases present by their characteristic x-ray diffraction patterns (Fig. 5). The actual elemental composition of the various minerals present was determined by electron microprobe analysis, using samples specifically processed to give the

grain sizes required for accurate microprobe analyses.

SYNROC Performance

The five major criteria for judging waste-form performance are:³

- Waste loading (the quantity of waste that can be loaded into a given volume of the waste form).
- Leach resistance.
- Mechanical properties, particularly the tendency of the solid to form respirable dusts on impact.
- Thermal properties, particularly thermal conductivity and thermal expansion.
- Resistance to the effects of radiation damage.

In general, none of the performance standards that the waste form must meet in the above categories is completely defined. Consequently, in common with most waste-form developers, we take borosilicate glass as a standard and measure performance in terms of the degree of improvement in relation to this standard. In this section, we compare the performance of our SYNROC D with the available corresponding data on borosilicate-glass.

Waste Loading

Waste loading affects all phases of the waste isolation scheme—processing,

Fig. 3

A 15-cm-diam disk of SYNROC D produced in our kilogram-size processing equipment. This sample contains simulated waste very similar in composition to the radioactive sludge found in tank storage at the Savannah River Plant in South Carolina.

interim storage, transportation, and repository (underground) storage. In general, the higher the waste loading, the fewer waste canisters will have to be produced, transported and, ultimately, stored. This helps to reduce both cost and risk, as long as the processing scheme (to

Fig. 4

Pie chart summarizing the starting materials and mineral products of the SYNROC D process. The emerald green areas represent the additives required to form the final SYNROC phases from the Savannah River Plant waste sludge (blue). Note that this sludge contains a large proportion of nonradioactive material, primarily compounds of the transition metals iron, aluminum, and manganese. The final SYNROC mineral phases can be divided into two groups, those that contain radionuclides (yellow green) and those that do not (orange).

Fig. 5

X-ray diffraction pattern of a special formulation of SYNROC D that contains hollandite, showing the mineral phases present. Only the major peaks are identified in this figure, although we have been able to assign all peaks except for the two small ones denoted by "X." Thus, the quantity of unknown material in SYNROC is very small.

achieve high loading) is not so complex or costly as to offset these gains.

The loading of Savannah River Plant defense wastes that can be achieved in SYNROC is much greater than that in borosilicate glass because of two factors, a higher waste proportion and a higher matrix density. Generally, borosilicate glass will accept only about 25 to 30 wt%

waste whereas in SYNROC, we typically load at levels of 60 to 70 wt%.^{3,10} Also, SYNROC is 35% more dense than glass, so the actual volumetric waste loading (concentration) is about three and a half times greater.

Leach Resistance

As long as the waste package remains dry, there is virtually no way for the stored radionuclides to reach the biosphere. However, it is impossible to guarantee that the waste package will remain dry forever. Therefore, the most important performance characteristic of any waste form is its ability to withstand leaching when exposed to groundwater.

We have evaluated the leaching characteristics of SYNROC D under both flowing (dynamic) and nonflowing (static) groundwater conditions and over a temperature range of 40 to 200°C. Some of our tests have been conducted for periods of hundreds of days.

Figure 6 compares selected SYNROC D leaching data with the corresponding data for borosilicate glass.¹¹ These results are from a standardized test (called the MCC-1) carried out in deionized water. The data are expressed in terms of leach rates, i.e., the quantity of material removed per unit of

Fig. 6

Experimental leach rates (quantity of material lost per day per square meter of exposed surface area) for SYNROC D and borosilicate glass in deionized water at 90°C. The data were collected over a period of 28 days. The values have been normalized to the elemental composition to account for the difference in radwaste loading.

exposed surface area over a given time ($\text{g}/\text{m}^2 \cdot \text{d}$).

Two types of leach rate are represented, the rate of loss for the key matrix element of the waste form (silicon for glass and titanium for SYNROC) and leach rates for two of the radionuclides in the waste, uranium (an actinide) and cesium (the most soluble fission product). For all radionuclides except cesium-137, leach rates for SYNROC D are less than one tenth those for borosilicate glass.

The cesium leach rate for SYNROC depends on the host mineral. If nepheline (a silicate) is used, as in SYNROC D, the cesium leach rate is about 50% that of borosilicate glass. If hollandite (a titanate) is used, as in the case of SYNROC C or special formulations of SYNROC D, the leach rate for SYNROC is less than one tenth that of glass.

Thermal and Mechanical Properties

Table 6 compares some important thermal and mechanical properties of SYNROC D with those of borosilicate glass. In general, the properties of SYNROC are equivalent to or better than those of the glass. The only exception is the compressive strength, which is better for borosilicate glass.

Radiation Damage Effects

The fifth major performance criterion is the ability of the waste form to maintain its integrity despite radiation damage. There are two aspects to our investigation of radiation damage in SYNROC. First, we dope SYNROC D samples with enough of a short-lived plutonium isotope to create, in a few months, the expected long-term storage alpha decay. We also deliver a high ionization dose to the samples at the beginning of the experiment to simulate the effects of beta-decay of fission products over the first few hundred years of storage. This work will be completed in FY82. Second, we compile published damage information on both naturally occurring and synthesized SYNROC minerals or mineral analogues. Much of the information on natural samples comes from data published by Ringwood and his coworkers (see, for example, Ref. 9).

Table 6 Thermal and mechanical properties of SYNROC D and borosilicate glass.

Property	SYNROC D	Glass
Thermal conductivity, $\text{W}/\text{m} \cdot \text{K}$	2	1
Density, g/cm^3	4.0	2.9
Thermal expansion coefficient ($20\text{--}425^\circ\text{C}$), $10^{-6}/\text{K}$	11	12.2
Compressive strength, MPa	352	690
Bulk modulus, GPa	107	33
Shear modulus, GPa	54	24
Young's modulus, GPa	139	63
Respirable particles on impact, wt% ^a	0.16	0.16

^aRespirable particles are those smaller than $10\text{ }\mu\text{m}$. To measure this quantity we use a standard impact having an energy density of $10\text{ J}/\text{cm}^3$. This test is designed to simulate the effects of a possible impact-accident scenario on the waste form.

The most important performance characteristic of any waste form is its ability to withstand leaching when exposed to groundwater.

There are natural specimens of zirconolite and perovskite containing uranium and thorium that have, over their lifetimes, accumulated radiation doses greater than those that would occur in SYNROC D stored for a million years. Ringwood studied the volume expansion due to this radiation damage and showed that, at most, it was about 1.8% for the perovskite and 2.8% for zirconolite.

Studies of lead isotopes in natural zirconolites and perovskites¹⁰ show that these minerals possess a remarkable ability to retain uranium and thorium and their decay products even after suffering a large degree of radiation damage. Measurements of lead-207/uranium-235, lead-206/uranium-238, and lead-207/lead-206 ratios permit three independent age calculations. These calculated ages agree closely, which means that the system has been "closed" with respect to uranium, lead, and intermediate daughter products since the mineral first crystallized, millions of years ago.

SYNROC Scale-Up: Problems and Potential

Over the past several years, we have demonstrated that SYNROC D has excellent performance properties and can be made on a laboratory scale. We are now

beginning the engineering research to determine whether SYNROC can be made on a large scale (1000 to 2000 kg per day in a remotely operated environment).

Option one, Fig. 7, presents the engineering flow sheets for a direct scale-up of our current laboratory operations. According to this analysis, the SYNROC process requires four times as many process steps and twice as much work volume as the borosilicate-glass process. The cost of the processing section of the waste immobilization facility (whether it is to make glass or SYNROC) is closely coupled to the floor space needed. Thus, the cost of a SYNROC-processing cell is about \$600 million vs \$300 million for a glass-processing cell. (The total estimated plant costs are between \$1.0 and \$1.5 billion).

Because the entire facility is operated remotely, the equipment used in the waste-processing plant must be reliable and easy to maintain. Currently, only two tools can be used to install, disassemble, or repair the equipment, an impact wrench and a crane. In other words, inside the processing cell one can only twist or lift. To design equipment that can be maintained with only these two operations, a separate piece for each step in the process, is a formidable assignment.

Our approach will be to combine and simplify the number of steps, thus reducing the size and complexity of the process. For example, we now feel we may be able to combine the mixing, drying, and calcining steps into a single operation by using a slurry-fed fluidized-bed calciner (see option two in Fig. 7). This unit has no moving parts and so may be expected to require little maintenance. Furthermore, this change could save considerable space.

The current engineering flow sheets also call for the use of a hot isostatic press to carry out the mineralization step. Storage for the pressurizing fluid occupies a large area inside the remote cell. Recent laboratory studies have led us to believe that we may be able to use either sintering or uniaxial hot pressing (in a die) as alternatives to the isostatic press. Both processes give excellent sample quality and have the potential advantage of

Fig. 7

The major processing steps for scale-up of SYNROC D production. Option one is based directly on our current laboratory experience. Option two represents a simplified production stream that we consider achievable with further engineering research.

reducing the complexity (and size) of the mineralization step.

Summary

We are developing SYNROC D, a synthetic mineral capable of immobilizing high-level radioactive waste for geologically significant time spans. We have essentially completed laboratory-scale tests, producing SYNROC disks loaded with mock waste as large as 15 cm in diameter and 2.5 cm thick. The thermal, mechanical, chemical, and radiation-resistance properties of this material equal or exceed those of borosilicate glass, the material used in reference waste-solidification designs.

We are investigating the means for producing SYNROC D in the quantities necessary for the waste-storage task. Preliminary designs indicate that plant costs and operating expenses may be roughly comparable to those for borosilicate glass or perhaps a little higher. We are investigating process simplifications that would reduce costs and maintenance.

Key Words: radioactive waste storage; radioactive waste—disposal, management, solidification.

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Ozone, Respiration , and “The Bends”

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In our studies of smog-related breathing problems and decompression sickness ("the bends"), we are using a positron camera and radioisotopes produced in our 100-MeV electron linear accelerator to determine the effects of ozone on respiration and the relationship of nitrogen transport to decompression sickness.

Physiologists investigating respiratory diseases and decompression sickness have long been hampered by their inability to measure accurately and in detail the distribution of such gases as oxygen and nitrogen in the body. Nuclear medicine, which can measure an extremely small quantity of radioactive material and trace its progress through the body with imaging devices, offers the hope of circumventing this limitation. The equipment and techniques of conventional nuclear medicine are built around commercially available radioisotopes, however, and the radioisotopes of oxygen and nitrogen cannot be stocked and distributed because none has a half life longer than ten minutes.

In work supported by the National Institutes of Health and in cooperation with the University of Washington School of Public Health and Community Medicine, we developed a sensitive technique¹ for measuring lung ventilation. Using this technique, we have been investigating the changes that occur in response to low concentrations of ozone. These studies were made possible by our ability to produce short-lived radioactive tracers for both oxygen and nitrogen as needed. We do this by irradiating the pure gases with x rays at our 100-MeV electron linear accelerator (see box on pg. 27). The oxygen-15 and nitrogen-13 radioisotopes decay by positron emission; the positrons collide with electrons, producing 0.511-MeV gamma rays that escape from the body with minimal attenuation. These are readily identified and detected by our positron camera (see box on p. 29), allow-

ing us to determine their point of origin in the body.

The short half lives of these tracers (two minutes for oxygen-15 and ten minutes for nitrogen-13) make possible high counting rates without excessive radiation dose to the subject. During the entire course of our ventilation experiments, for example, each subject was exposed to less than 50 mR of radiation, a dosage equivalent to that received from two typical chest x rays.

Integrated Gas-Exchange Measurements

Studies have shown that exposure to ozone at high ambient concentrations changes the pulmonary mechanics of healthy lungs, causing bronchial inflammation and pulmonary edema (swelling).^{2,3} The effects of lower concentrations (such as those present in an industrial environment) could not be investigated, however, because conventional tests are too insensitive.

What we really want to know in studies of respiratory difficulty is how much air gets into the tiny sacs (the alveoli) in the lung. Standard techniques measure total ventilation, i.e., the total volume of air entering the lungs in one minute. Measurements of total ventilation are straightforward,⁴ but the interpretation of results has been clouded by complicating factors. For example, the amount of air leaving the lungs is slightly less than the amount inhaled, since the body absorbs more oxygen from the air than it returns to it as carbon dioxide.

Nuclear medicine . . . can measure an extremely small quantity of radioactive material and trace its progress through the body with imaging devices.

In addition, about 30% of each breath simply fills conducting airways (the complex branching bronchi and bronchioles which become narrower, shorter, and more numerous as they penetrate deeper into the lungs) without ever reaching the respiratory zone of the lungs to participate in gaseous exchange. It has been difficult in the past to measure this total "dead volume" accurately and thus to determine actual ventilation. Furthermore, such overall measurements could provide no estimates of differences between one part of the lungs and another.

Similar problems have arisen in relation to interpreting the effects of working under high compression, as in deep-sea diving. It is known that nitrogen dissolves in the body fluids and that rapid decompression causes extreme pain and various disabilities. It has been suspected that different tissues might have widely differing abilities to absorb nitrogen under pressure and differing release rates upon decompression. However, these differences were almost impossible to measure, and, in the absence of such information, it has been necessary to conduct deep-sea diving operations on the basis of calculated diving and decompression schedules that are not supported by experimental evidence.

Radioisotope Gas-Exchange Measurements

Ozone and Respiration

The work on ozone-concentration effects on lung ventilation was done mainly by exposing healthy dogs, under strictly controlled conditions, to concentrations of ozone similar to those found in the smogs of California's larger cities. To ensure a constant and reproducible volume of air in the lungs at the end of each breath, the dogs were anesthetized and kept on a mechanical respirator throughout the experiment.

One of the experimental constraints was the need to distinguish between annihilation counts from the two radioisotopes nitrogen-13 and oxygen-15. Both produce 0.511-MeV gamma rays and so affect the detector in the same way. This made it necessary to administer the two radioisotopes at different times so

that one would have decayed substantially before the other was introduced (Fig. 1).

Because the objective of these tests was to determine the changes in regional ventilation resulting from exposure to ozone, our first step was to establish a normal (pre-ozone) baseline for each subject. After the subject had breathed normal air for two hours, we changed the supply to air containing a small concentration of nitrogen-13 and monitored the counting rate in each of a hundred or more lung areas for roughly two minutes or until the nitrogen reached equilibrium in each of them, indicating that the particular portion of the lung had become ventilated. We recorded the measured decay rate and the number of breaths needed to reach this plateau for each separate lung area. Figure 2 is a typical plot of nitrogen-13 activity in one of these lung regions vs the number of breaths.

Exactly 30 minutes after the nitrogen-13 exposure, the subject took a deep breath of air containing 5% of carbon dioxide labeled with oxygen-15 and held it for 20 seconds. Our objective in this part of the experiment was to observe ozone-induced changes in the lung's gas-exchange membranes. During this time, we measured the clearance rate (wash-out) of the carbon dioxide from the lungs (entering the bloodstream and being carried away), which was much faster than the two-minute half life of oxygen-15 (Fig. 3). We repeated this process twice, to establish the consistency of the baseline measurement.

After establishing the baseline for an animal, we changed for a total of three hours to an air supply to which we could add ozone. Every hour during this interval, we repeated the nitrogen and carbon dioxide measurements described above. Comparison of the two sets of data, before and after the start of ozone exposure, reveals any changes in the ventilation characteristics of the dog's lungs. The nitrogen-13 data gave clear evidence of changes that correlated with ozone exposure (Fig. 4). The oxygen-15 data were considerably more erratic and are still being studied.

Three of the 12 animals used in the experiment served as a control group and

were not exposed to ozone. The remaining nine animals, grouped into three groups of three each, were exposed to ozone concentrations of 0.13, 0.25, or 0.45 parts per million (ppm). For comparison, ozone concentrations in the range of 0.5 to 1.0 ppm have been measured in the open air during rush hours near Los Angeles freeways.

Fig. 1

Experimental gas-supply schedule. The major air supply could be shunted from pure air to air containing several known levels of ozone concentration. At intervals, we administered nitrogen-13 or oxygen-15 (in the form of carbon dioxide) to measure changes in ventilation characteristics.

Fig. 2

A typical nitrogen-13 influx curve for a particular subregion of the dog's lung. The saturation time for the subregion is defined as the point where the count rate reaches 95% of the plateau value.

Fig. 3

The activity of carbon dioxide tagged with oxygen-15. The dog inhales 1.5 litres of air containing 5% carbon dioxide in 8 s, then holds it for 20 s. It can be seen that the carbon dioxide leaves the lungs (diffuses into the bloodstream) with a half time of about 10 seconds.

Analysis of the nitrogen-13 data indicated that central areas of the lungs were becoming fully ventilated early and those around the periphery much later. For simplicity in presenting the data, however, we simply plotted, as a function of the number of breaths taken, the

Fig. 4

The ventilation characteristics of three dogs, before and after ozone exposure. The consistent differences between the pre- and post-exposure curves clearly indicate that even very low ozone concentrations reduce lung ventilation. The effects generally appear within one hour of exposure and affect most strongly the outer regions of the lungs. For the ozone concentrations studied, there was a direct relation between the amount of ventilation impairment and the concentration.

percentage of lung regions that had reached saturation. Figure 4 presents typical results of these measurements.

Each post-exposure curve must be compared only with its corresponding baseline curve because the baseline curves for individual subjects differ greatly. Comparison of the corresponding baseline and ozone-exposure curves shows a definite trend. The changes in ventilation (longer time to reach saturation) appear with exposures of an hour or less and increase with increasing ozone concentration.

Figure 5 shows images of nitrogen-13 distribution obtained before and after a heavy exposure to ozone (5 ppm). The height above the base plane indicates the number of annihilation events detected from different parts of the field. In the baseline image (Fig. 5a), the major lobes of the lungs stand out clearly. In the ozone-exposure image (Fig. 5b), the right lung has essentially ceased to function. Because the respirator supplied a constant volume of air, the activity in the left lung increased correspondingly.

The mechanisms by which ozone produces these changes are poorly understood. Our findings in dogs suggest that the changes appear within one hour of the onset of ozone exposure, and, in some cases, at 0.13 ppm (the National Ambient Air Quality Standard for one hour of exposure is 0.12 ppm).

Similar effects have been found in healthy human subjects. Persons with respiratory disease can experience magnified symptoms. Our human-subject experiments included exercise periods on a bicycle, during which the ozone effects were also enhanced. Healthy lungs appear to recover from these effects within a day or two, but the long-term effects on humans are unknown and will require further study.

Decompression Sickness

In cooperation with the Naval Medical Research Institute, we have conducted similar experiments to study the transport of nitrogen in human tissues. Such measurements are important for the better understanding of decompression-sickness phenomena, commonly known as "the bends." Ultimately, this research

Fig. 5

Positron camera images of the active lungs of a dog, in which the height above the plane is a measure of the nitrogen-13 activity. (a) Normal ventilation before administration of ozone-containing air. The major lobes of the

lung are clearly distinguishable. (b) Impaired ventilation after breathing air containing 5 ppm of ozone for a few minutes. The right lung appears to have stopped ventilating, and the left lung is receiving most of the nitrogen-13.

Radioisotope Production

The first step in the production of oxygen or nitrogen tracers with our linear accelerator is the generation of energetic x rays (bremsstrahlung) by stopping an 80-MeV electron beam in a target made of tungsten or tantalum. The x rays pass through a high-pressure stainless steel bottle containing either pure nitrogen or pure oxygen. In the resulting photonuclear reactions, a high-energy photon is absorbed and a neutron is ejected, converting nitrogen-14 into nitrogen-13 or oxygen-16 into oxygen-15.

Both of these products have in their nuclei one too many protons for stability and decay by emitting a positron. When these positrons interact with electrons in the surrounding matter, they mutually annihilate one another and create pairs of 0.511-MeV gamma rays emitted in opposite directions.

Because both oxygen-15 and nitrogen-13 emit positrons, they are indistinguishable to the camera and must be administered separately. Hence, it is important to use only pure tagged gases as tracers.

To avoid contaminating the experiment with radiation from other elements, also made active by photonuclear reactions, we fill the target bottle only with highly purified nitrogen or oxygen gas. Oxygen-free nitrogen forms no significant by-products under x-ray irradiation at the energies used here.

When oxygen is bombarded with x rays, however, the production of contaminants cannot be avoided. These include ozone (which might or might not be tagged with

oxygen-15) and a radioisotope of carbon (carbon-11, with a 20-minute half life) formed by ejecting both a neutron and an alpha particle from the oxygen-16 nucleus. The carbon-11 promptly reacts with the oxygen to form carbon dioxide.

Before this oxygen is inhaled, we must remove both the ozone and the radioactive carbon, trapping the ozone with activated charcoal and the carbon dioxide with soda lime. Then, we convert the radioactive oxygen into carbon dioxide by passing it through a charcoal oven and a bed of cupric oxide heated to 850°C. For administration, we mix five parts of this pure carbon dioxide gas with 95 parts of clean air. The entire process must be done rapidly because of the two-minute half life of oxygen-15.

Although the 20-minute half life of carbon-11 makes it unsuitable for use in our respiration experiments, we can produce this isotope whenever we need it. In this case, we might irradiate pure methane to eliminate the formation of contaminating radioisotopes and burn the resulting mixture of hydrocarbon gases in pure oxygen to produce pure carbon dioxide. We are investigating potential applications for this capability.

Our irradiations convert about one nitrogen atom in every trillion from nitrogen-14 to nitrogen-13. This is equivalent to an initial decay activity of about 1.7 mCi/l, or about 60 million positron annihilation events per second per litre of nitrogen gas. The oxygen-15 activity obtained is just slightly larger.

The cause of decompression-sickness symptoms when a diver's ascent is too rapid is unknown, but it is presumed that bubbles of gas form in the tissues and blood stream as the external pressure is reduced.

could influence the revision of diving decompression tables and the treatment of stricken deep-sea divers.

In underwater diving, the external pressure on a diver increases by about one atmosphere for every 10 m of descent. As long as the pressure in a diver's sinuses and lungs exactly balances the external pressure, there is no danger of a catastrophic compression or over-expansion during descent and ascent, respectively. The problem of maintaining this balance is greatly complicated, however, by the solubility of nitrogen in tissues. Nitrogen is only poorly soluble in the blood at normal atmospheric pressure. As the pressure in a diver's lungs increases, however, more and more nitrogen is forced into blood and tissue.

Nitrogen is more soluble in fatty tissue, which has a meager blood supply. As a diver ascends, nitrogen is removed from fatty tissue very slowly, by gas exchange with the blood. In general, nitrogen diffusion through tissue is also quite slow. Thus, it takes a long time to establish a satisfactory equilibrium between the nitrogen dissolved in the tissues and that in the surrounding air.

The cause of decompression-sickness symptoms when a diver's ascent is too rapid is unknown, but it is presumed that bubbles of gas form in the tissues and blood stream as the external pressure is reduced. The diver's body might tolerate a few small bubbles, but large numbers could cause excruciating pain, particularly near joints. Gas bubbles reaching the brain or the central nervous system could obstruct the flow of blood, causing deafness, blindness, paralysis, or even death. All of these outcomes have been observed in decompression sickness, although it has never been proved that nitrogen bubbles are the cause.

Temporary decompression-sickness symptoms can be dramatically relieved by immediate recompression underwater or in a hyperbaric decompression chamber. Such symptoms usually can be prevented by gradual decompression, regulated according to schedules that specify how fast a person should be able to surface with little risk of decompression sickness.

The Positron Camera

Our positron camera⁵⁻⁷ consists of two identical scintillation-detector heads, each equipped with 37 photomultiplier tubes, mounted on a 1.25-cm thick, 40-cm-diam scintillator crystal of sodium iodide. The two detector heads are arranged in a coincidence circuit to detect the two oppositely directed gamma rays generated during each positron-electron annihilation event. Such "coincidence detectors" register two events that occur simultaneously (or within a very short time of one another) and reject all others. The counting speed of our system is on the order of 50 000 events per second for each head.

A precision resistor-capacitor network coupled to the outputs of the photomultipliers makes it possible to analyze the signals and determine where each gamma ray hit the crystal. This information is digitized and stored by an on-line computer, which then fits an imaginary straight line (along which the annihilation event must have occurred) between each pair of scintillation points. The intersections of many of these lines determine the location of the radioactive material between the detector heads (the lung is shallow in comparison with the distance between scintillation detectors).

We divide the lung into 90 to 110 separate regions and keep separate counting records for each region. This enables us to determine the location of the radioactivity as a function of time.

The point-source resolution of the camera, in the plane of focus, is about 1.5 cm. In the case of lung tissue, which has an average density of only 0.23 g/cm^3 , the positrons can travel several centimetres from the emitting nucleus before the annihilation takes place. This blurs the image somewhat, further degrading the camera's resolution. Even so, the information obtained is far superior to that obtainable from previous nonnuclear measurements.

With our techniques, we can observe the nitrogen-13 activity in specific body areas such as shoulders, neck, ears, elbows, and knees.

At present, such decompression-table schedules are based on presumed rather than directly measured rates of inert-gas exchange in specific tissues. In previous measurements of nitrogen kinetics,⁸ the nitrogen content was determined by subtracting measured quantities of oxygen and carbon dioxide from the known volume inhaled or exhaled. Then, comparison of inhaled and exhaled air provided an estimate of the average value for nitrogen movement in the body.

With our techniques, we can observe the nitrogen-13 activity in specific body areas such as shoulders, neck, ears, elbows, and knees. Nitrogen diffuses through all human tissue, at different rates in different tissues. Since these diffusion rates are generally low (it has been estimated that it takes about 72 hours to replace all the dissolved nitrogen in the body's tissues), we must count the nitrogen-13 activity for as long as it is practical to do so.

This time is determined by two factors: the initial source strength (the amount of nitrogen-13 available initially for uptake, which is limited by the allowable total dose rate for the individual) and the half life of nitrogen-13, about 10 minutes. The sensitivity of our positron camera makes it possible to detect nitrogen-13 activity over an interval of about two hours, i.e., 12 half lives, by which time the original counting rate has been reduced by a factor of more than 4000.

Our nitrogen-exchange measurements in human tissue appear to confirm the existence of sharply different exchange rates in different tissues.⁹ In particular, data from the knee-joint region (Fig. 6) indicate that nitrogen accumulated there for a considerable time after we stopped administering air that contained nitrogen-13. Thus, the knee appears to be a "sink" for nitrogen, concentrating it as it is released from other parts of the body into the blood stream. There is at this time no known physiological model for this striking affinity, but our initial findings correlate with the observation that one of the major symptoms of decompression sickness is pain in the knees.

Summary

The radioisotope technique described here holds great promise for adding significantly to our understanding of inert-gas exchange in body tissues. We have demonstrated regional impairment of ventilation on exposure to low concentrations of ozone and an unexpectedly high affinity for nitrogen in the human knee. The Navy is considering hyperbaric (high-pressure) experiments to extend our decompression-sickness studies with nitrogen and a series of argon-gas tracer experiments to compare the action of an inert gas in the body with that of nitrogen. This research may make it possible to revise the standard diving tables for decompression in nitrogen-oxygen or helium-oxygen atmospheres.

Key Words: linear accelerator; ozone; photonuclear reactions; positrons; radio isotopes; smog; respiration.

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Fig. 6

This graph, which is corrected for the effect of radioactive decay on the counting rate, shows the nitrogen-13 activity (as a function of time) in the knee of a Navy diver. The activity continued to increase dramatically, long after we stopped administering nitrogen-13. The physiological reason for this striking affinity is unknown, but one of the major symptoms of decompression sickness is pain in the knees.

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