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Radiation Effects in the Environment

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PREFACE

In 1997 Dr. Thomas Atcity, Vice President, Navajo Nation, submitted a request to Dr. Fred Begay, Assistant for Science and Technology, Navajo Nation, and technical staff member, Los Alamos National Laboratory, to organize an education workshop on the radiation effects in the environment for the Navajo communities located at Church Rock, New Mexico and Crownpoint, New Mexico.

Since there exist plans for the uranium industry to mine uranium ores near these communities and since the history of uranium mining operations have had adverse human and natural environmental effects on several Navajo communities, the Navajo leadership has requested that an education program be developed and implemented for public understanding of nuclear science and technology and related issues.

In July 1998 Dr. Fred Begay conducted an education workshop on radiation effects in the environment where selected scientists from the nuclear science community were invited to participate in the workshop.

It is a pleasure to acknowledge scientists from the E.O. Lawrence Berkeley National Laboratory, the Los Alamos National Laboratory, and the University of New Mexico who made contributions to the education workshop. A highlight of the workshop was a video presentation by Dr. Glenn T. Seaborg on the history and his contributions to nuclear medicine. In 1951 Dr. Seaborg was awarded the Nobel Prize in Chemistry.

In addition, we acknowledge Mr. James Tutt, President, Crownpoint Institute of Technology, for the gracious use of his facilities for the workshop. Furthermore, we acknowledge the following sponsors; the Office of the President and Vice President, Navajo Nation, the Waste-Management Education & Research Consortium at New Mexico State University, the Seaborg Hall of Science, the E.O. Lawrence Berkeley National Laboratory, the Los Alamos National Laboratory, the LaFonda Hotel, and GreekTown Cafe.

In the preparation of the manuscript for the workshop, we acknowledge contributions made by Mike Kolb from the Los Alamos National Laboratory. In addition, we are grateful to Dr. Charles Doering, Senior Editor, and his staff at the American Institute of Physics for publishing the Proceedings of the workshop. The published Proceedings will be used as an education document to support future Navajo education programs in workshops and K-14 schools.

This work was done under the auspices of the U.S. Department of Energy.

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Introduction

Fred Begay, PhD

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Man has existed for about a million years. He has possessed writing for about 6,000 years, agriculture somewhat longer. Science, as a dominant factor in determining the beliefs of educated men, has existed for about 300 years; as a source of economic technique, for about 150 years. In this brief period it has proved itself an incredibly powerful revolutionary force. When we consider how recently it has risen to power, we find ourselves forced to believe that we are at the very beginning of its work in transforming human life. What its future effects will be is a matter of conjecture, but possibly a study of its effects hitherto may make the conjecture a little less hazardous.

The effects of science are of various very different kinds. There are direct intellectual effects: the dispelling of many traditional beliefs, and the adoption of others suggested by the success of the scientific method. Then there are effects on technique in industry and war. Then, chiefly as a consequence of new techniques, there are profound changes in social organization which are gradually bringing about corresponding political changes. Finally, as a result of the new control over the environment which scientific knowledge has conferred, a new philosophy is growing up, involving a changed conception of man's place in the universe.

The effects of science and technology on the Navajo communities have been limited. Due to lack of capital and quality human resources, the socio-economic indicators¹ of the Navajo show that the Navajo exist in a human and natural environment which is similar to that of a Third World country.

Although the Navajo possess substantial resource wealth-coal, gas, uranium, water-this potential wealth has been translated into limited permanent economic or political power. In fact, wealth or potential for wealth has often made the Navajo the victims of more powerful interests greedy for the assets under limited Navajo control. Without wealth or political power, the Navajo has had to rely upon the constitutional-legal system and the moral conscience of society for their survival.

Tribal survival was an issue at the time the United States was established, and it has continued to be an issue throughout our history. A key factor in this issue is the status of Navajo people as governmental entities, as a tribe with a land base and power over their land base. This factor distinguishes the Navajo from any other group in American

¹ *The Navajo Ten Year Plan*, Navajo Research and Statistics Center, The Navajo Tribe, June, 1972; *NN FAX 93*, Division of Economic Development, The Navajo Nation, March, 1994.

society. *Indian people relate to the Federal Government as nations and tribes and citizens of nations and tribes, and on that basis first and in other ways only incidentally.*²

Since World War II, the Navajo has had adverse experiences with the mining of uranium. For example, the early Navajo miners who were exposed to radon in uranium mines have died due to lung cancer problems.

The primary focus for this education workshop on the radiation effects in the environment is to provide a forum where scientists from the nuclear science and technology community can share their knowledge toward the advancement and diffusion of nuclear science and technology issues for the Navajo public.

The scientists will make an attempt to consider the following basic questions; what is science; what is mathematics; what is nuclear radiation? Isaac and Leavitt will discuss the question of nuclear radiation in these Proceedings.

Although, most western social scientists agree that Navajo thought has no structure of knowledge concerning concepts about science, mathematics, and radiation....Dr. Begay and Hataalii(Chanter) Yazzie will present a brief discussion in both the Navajo and English language on the Navajo view of radiation. The origin of Navajo concept of radiation is embedded in Navajo religion and medicine.

During the decade, 1972-1982, Fred Begay and the late Hataalii Fred Stevens, Jr. devoted hundreds of hours to investigating the parallels between Navajo and modern scientific thought. In order to understand these parallels, abstract levels of thought are required to understand the structure and meaning of the Navajo and modern perspectives of nature, and valid connections between the two structures of knowledge. Ancient Navajo Hataaliis devised an abstract language of nature to describe and explain the properties of connections between thought, language, and reality.³ On the other hand, the architects of modern scientific thought have devised a complex abstract mathematical language to reason about the structure and meaning of the laws of nature. Progress for the advancement and diffusion of modern⁴ and Navajo language of nature is based on the research and analysis of three kinds of objects, and valid connections between these objects. These objects include events, laws of nature, and symmetry or invariance principles. Laws of nature are determined by regularities in events, and the symmetry or invariance principles are determined by regularities among the laws of nature. This process in the analysis of valid connections between thought, language, and nature has occupied the attention of man since ancient times.

Results of the Begay-Stevens research has revealed the existence of strange and mysterious concepts in the Navajo language of nature. With these research results Begay and Stevens prepared a lecture entitled *The Physics of Laser Fusion*⁵ in the Navajo language. This lecture has been delivered in numerous Navajo schools, and in universities and colleges in the US, Canada, and Alaska.

² U.S.,Department of Interior, Bureau of Indian Affairs, *Report on the Implementation of the Helsinki Final Act as Applied to Native Americans* (1979), p. i.

³ JB Carroll, *Language, Thought, and Reality: Selected Writings of Benjamin Lee Whorf*, MIT Press and John Wiley & Sons, Inc, New York, 1956.

⁴ EP Wigner, *Symmetry in Nature*, Proceedings of The Robert A. Welch Foundation conferences on Chemical Research, XVI. Theoretical Chemistry, November, 1972.

⁵ J Emmett, J Nuckolls and L Wood, *Fusion Power by Laser Implosion*, Scientific American 230, p24, June 1974.

In 1941 Courant⁶ pointed out that *mathematics as an expression of the human mind reflects the active will, the contemplative reason, and the desire for aesthetic perfection. Its basic elements are logic and intuition, analysis and construction, generality and individuality. Though different traditions may emphasize different aspects, it is only the interplay of these antithetic forces and the struggle for their synthesis that constitute the life, usefulness, and supreme value of mathematical science.*

Without doubt, all mathematical development has its psychological roots in more or less practical requirements. But once started under the pressure of necessary applications, it inevitably gains momentum in itself and transcends the confines of immediate utility. This trend from applied to theoretical science appears in ancient history as well as in many contributions to modern mathematics by engineers and physicists. The value of mathematics to the physicist is that mathematics is used as a language to reason about physical processes.

In 1960 Wigner⁷ pointed out that *the language of mathematics reveals itself unreasonably effective in the natural sciences...., a wonderful gift which we neither understand nor deserve. We should be grateful for it and hope that it will remain valid in future research and that it will extend, for better or for worse, to our pleasure even though perhaps also to our bafflement, to wide branches of learning.*

In 1975 Mandelbrot⁸ reported on his research on fractal geometry where he discussed that fractal geometry can be regarded as a new language in mathematics. Furthermore, he showed that the properties of fractal geometry can be classified into the following categories; artistic, mathematical, historic, practical and scientific. In 1891 Hilbert⁹ demonstrated the first visualization of a fractal in his space-filling curve. In modern times, the impact of the use of computers have improved the visualization of fractal geometry images. In 1988 Barnsley¹⁰ showed that a self-similar fractal such as the Sierpinski gasket can be used to investigate fundamental properties on the deep-seated connections between probability and determinism.

Sagdeev¹¹ and his colleagues have reported on extensive investigations on the fractal properties of chaos. For example, in their study in the acceleration of relativistic particles, they can predict the acceleration mechanism which was suggested by Fermi¹² to explain the origin of cosmic rays.

Winfree¹³ and his colleagues have reported on their investigations on the application of fractals and chaos to describe mechanisms of cardiac fibrillation.

⁶ RC Courant and H Robbins, *What is Mathematics*, Oxford University Press, 1941.

⁷ EP Wigner, *The Unreasonable Effectiveness of Mathematics in the Natural Sciences*, Communications on Pure and Applied Mathematics, 13, pp 1-14, 1960.

⁸ BB Mandelbrot, *Les objets fractals: forme, hasard et dimension*, Paris, Flammarion, 1975.

⁹ D Hilbert, *Über die stetige Abbildung einer Linie auf ein Flächenstück*, Math. Ann. 38, pp 459-460, 1891.

¹⁰ MF Barnsley, *Fractals Everywhere*, Academic Press, 1988.

¹¹ RZ Sagdeev, DA Usikov, GM Zaslavsky, *Nonlinear Physics: From the Pendulum to Turbulence and Chaos*, Harwood Academic Publishers, 1988; and GM Zaslavsky, RZ Sagdeev, DA Usikov, AA Chernikov, *Weak Chaos and Quasi-Regular Patterns*, Cambridge University Press, 1991.

¹² E Fermi, *Phys Rev* 75, 1169(1949).

¹³ AT Winfree, *Rotating Chemical Reactions*, *Scientific American* 230(6), 82(1974); and RA Gray, J Jalife, AV Panfilov, WT Baxter, C Cabo, JM Davidenko, AM Pertsov, *Mechanisms of Cardiac Fibrillation*, *Science* 270, 1222(1995).

Ours is a time of contests over issues incompletely understood. To those with a sense of history there is no novelty in that, but what seems new in our age are the number and variety of issues of this sort, and also the fact that a few of them really have a visible connection to the ultimate question of human survival.

Policy toward nuclear energy is such an issue. Where optimists see a front door to unlimited low-cost energy, pessimists see a back door to worldwide nuclear calamity, and neither perception is wholly unfounded. For more than fifty years both promise and menace have been strongly debated, and in the early 1970s the urgency of the debate was intensified as a consequence of heightened concern for energy supply, environmental hazard, and nuclear proliferation.

Louis Rosen presented a discussion on the results of an analysis of world energy perspective which shows that world energy demand will increase faster than the population growth. This effect has the potential for adversely affecting our way of life, including national security and international stability. A new concept for an alternative energy source was presented which has the potential to alleviate the major problems confronting current nuclear energy sources.

Maria Isaac and Christopher Leavitt presented a lecture on the *A,B,C's of Nuclear Science* where the basic properties of nuclear structure and radioactivity were discussed. In addition, experimental methods to measure alpha, beta and gamma radiation from low energy radiation sources were demonstrated.

Glenn T Seaborg prepared a video to discuss the role of nuclear medicine in the treatment of a wide range of illnesses. Dr. Seaborg shared his personal experiences in the evolution of the field of nuclear medicine.

Donald F. Petersen presented a lecture on the *Radon in the Environment* where he discussed the origin of radon production in the natural radioactive elements, and its impact on lung cancer. Dr. Petersen devoted some discussion on the transport of radon in the environment. In addition, Dr. Petersen has presented explanatory notes on the radon in the environment for the non-specialists, which is an important exercise for public understanding of this topic.

Since the World War II, uranium mining operations have deposited several mountains of uranium mill tailings on the landscape of the Navajo Reservation. Caroline Mason presented a lecture on *Bicarbonate Leaching of Uranium* where Dr. Mason has proposed that this technique be employed for remediation of uranium-contaminated sites across the Navajo Reservation. Dr. Mason has demonstrated the cost/effective use of this technique at the Los Alamos National Laboratory. Since future uranium mining operations will apply the bicarbonate leaching technique, Dr. Mason's discussion of the principles of this uranium mining technique has provided a timely opportunity for the Navajo public to review the merits of this mining technology.

Preliminary analysis of the water aquifer-uranium ore system show that the uranium ore is deposited in the water aquifer that serves the Navajo communities in the ChurchRock-Crownpoint, NM region. Consequently, since it is crucial to understand water aquifer dynamics relative to the geology of the uranium ore, Bryan J Travis presented a lecture on *Computational Methods for Subsurface Flow and Transport* where a review of computational methods which can be applied to investigate the water aquifer-uranium ore dynamics was made. Reliable experimental data will be needed to calibrate the computational studies.

The Proceedings of this Workshop will be used as a valuable reference material in future workshops and K-14 classrooms in Navajo communities that need to improve basic understanding of nuclear science and technology issues.

Navajo View of Radiation

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Abstract. This will be the first attempt to describe the Navajo view of radiation in the English language by a Navajo natural scientist and a Hataalii. The concepts of science such as radiation is used extensively in Navajo religion and medicine. We will review the origin of the concept of radiation in Navajo thought and compare its meaning relative to the concept of radiation from the modern perspective. Our task is to demonstrate that the Navajo concept of radiation can be used to discuss in the Navajo language modern issues such as nuclear radiation and radioactivity which is vital for Navajo public understanding of the impact of nuclear science and technology on the quality of their life.

Ancient Navajo Hataaliis(Chanters) devised an abstract language of nature with which to reason about natural phenomena. The structure of the Navajo religion and medicine is strongly based on the laws of nature which govern the evolution of the natural and human environment. Bergman¹ points out that the Navajo language of nature which is used extensively by the Navajo Hataaliis blend healing and worship in a powerful vision of life. As is the case with any culture, the ancient Navajo Hataaliis have devised models concerning the origin of natural phenomena such as the universe and life. Other primary properties that characterize Navajo thought include the method of counting objects in the Navajo healing therapies , and religion is based on an arithmetic which is similar to modulo-8 arithmetic; the structure of Navajo grammar is similar to that of German grammar; and skills in abstract reasoning are required to reason about natural phenomena.

Let us first analyze a comparison of equivalent energy released from the following energy sources. 10^{14} joules of equivalent energy are released from the following sources; daily energy output of Hoover Dam, solar energy per day on two square miles, burning of 7000 tons of coal, energy released in complete fission of one kg of uranium-235, energy equivalent ($E = MC^2$) of one gram of matter.

Let us now discuss the properties of radiation(Tsa'jilgish) emission from the burning of coal(Leejin), uranium(Leetsoii), and heavy water(To' Niidaaz). Note that the Navajo terms will be expressed in parenthesis.

Today, coal is widely used as a fuel to heat homes and for cooking meals on the Navajo Reservation. Chemical energy in the coal is converted into heat which is used in the home. Let us turn our attention to the 'burning' of uranium. In this case, we convert nuclear energy into heating of water to produce electrical energy where neutron radiation

¹ RL Bergman, *Navajo Medicine and Psychoanalysis*, Human Behaviour, pp8-15, July, 1973.

is used to transport the nuclear energy into the generation of electrical energy. The application of nuclear fission energy include nuclear weapons, electrical power plants, and nuclear medicine.

Let us now consider the 'burning' of hydrogen isotopes such as deuterium and tritium to generate electrical energy. Although enormous investments in capital and human resources have been made to demonstrate a thermonuclear fusion reactor across the world over the past four decades, we have yet to develop a fusion reactor which can compete economically with other electrical power plants. Thermonuclear fusion reactions have been demonstrated with the (a) hydrogen bomb and (b) conversion of fusion energy into the generation of solar radiation which plays a essential role in supporting life processes on the earth.

There exist Navajo concepts to describe the propagation of solar radiation(Johonaa'ei bii' Tsa'jilgish bii' 'A'aad) and gravitational radiation (Johonaa'ei bii' Tsa'jilgish bii' Bikaa') between the sun(Johonaa'ei) and the earth(Nihosdzaan). In 1971 Yazzie² reported on her investigations into the history of the Navajo culture. A major portion of the Navajo's knowledge concerning radiation processes is embedded in a story about a supernatural family that includes the sun(father: Johonaa'ei), Changing Women(mother: Asdzaa Nadleehe), and their twin sons Monster Slayer(Naayee' Neezghani) and Child Born of Water(To' Bajish Chini).

Another example is the concept of LASER (light amplification by stimulated emission of radiation) radiation (Hatsoo'algha k'aa'). The origin of the Navajo concept of LASER radiation is associated with the occurrence of electrical storms in the earth's atmosphere(Nltch'i). The Navajo model predicts that during an electrical discharge (Atsiniltl'ish k'aa') in the atmosphere, the electrical discharge excites the molecules in the atmosphere and generates a different light form which can be described as LASER radiation. This model is similar in principle to the performance a modern gas discharge LASER³ in the laboratory.

A concept which is used extensively throughout the Navajo view of natural phenomena is the concept of duality. For example, the concept of duality is used to describe fundamental connections between the LASER and the elecrrical discharge, the gravitation and solar radiation, the twins Monster Slayer and Child Born of Water, and healing and worship. In quantum physics⁴, the concept of duality emerges where we attempt to analyze the dual behavior of an electron as a particle and as a wave.

During the decade, 1972-1982, Fred Begay and the late Hataalii Fred Stevens, Jr. devoted hundreds of hours to investigating the parallels between Navajo and modern scientific thought. In order to understand these parallels, abstract levels of thought are required to understand the structure and meaning of the Navajo and modern perspectives of nature, and valid connections between the two structures of knowledge. Results of the Begay-Stevens research has revealed the existence of strange and mysterious concepts in the Navajo language of nature. Subsequently, with this knowledge, Begay and Stevens prepared a lecture entitled *The Physics of Laser Fusion*⁵ in the Navajo language. This

² E Yazzie, *Navajo History: Volume 1*, Navajo Community College Press, 1971.

³ AL Shawlow, *Lasers and Light*, Readings from Scientific American, 1969.

⁴ AP French and EF Taylor, *An Introduction to Quantum Physics*, WW Norton & Co, Inc, NY, 1978.

⁵ J Emmett, J Nuckolls and L Wood, *Fusion Power by Laser Implosion*, Scientific American 230, p24, June 1974.

lecture has been delivered in both the Navajo and English language in numerous Navajo schools, and in universities and colleges in the US, Canada, and Alaska.

Although traditional Navajos understand these abstract concepts about natural phenomena in the Navajo language, they are not familiar with modern nuclear science and technology issues such as the impact of uranium mining operations on the their environment. There exist adequate Navajo knowledge among the Navajo Hataaliis and elders to understand radiation processes in natural phenomena. Modern man-made radiation processes can be analyzed using the ancient Navajo language of nature.

This Workshop has provided a unique opportunity for the Navajo public to improve their literacy on modern nuclear science and technology issues which are associated with uranium mining programs.

Nuclear Energy, National Security and International Stability

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Abstract. World energy resources are on a course which will cause them to fall behind requirements by the middle of the next century. This has the potential for adversely affecting our way of life, including national security and international ability. A new concept has the potential of alleviating the major problems confronting nuclear energy.

INTRODUCTION

In attempting to construct a reasonable talk for this occasion, I asked myself what will be the problems to be faced by my great grand-children during their adult years? I see five major ones: (a) world population pressures, (b) environmental degradation, (c) world energy resources, (d) international instability, and (e) national security. Guess what! They are all interrelated. Failure to solve one will affect all the others. To be in some sort of harmony with the subject matter of this workshop, I shall attempt to summarize some thoughts on nuclear energy, national security, and international stability. They are, indeed, very closely coupled.

AN UNFORESEEN LEGACY OF WORLD WAR II

It has been known, for about 70 years, that, aside from gravitational energy, all sources of energy on earth and in the heavens above, are nuclear in origin. It was 53 years ago that scientists, at Los Alamos, demonstrated that nuclear energy which powers the sun and the stars can be released on earth on a very large scale. Immediately thereafter nuclear weapons brought a swift end to World War II. Since then they have helped to resolve the Cold War by deterring hostilities involving the major world powers. These 54 years without conflict among the major world powers in the longest such period in modern history-250 years. With the peaceful end of the Cold War there is realistic hope that war involving the major powers can be avoided indefinitely. There are however major concerns. One is state-sponsored terrorism. Another is that population pressures and legitimate desires for improved living standards can culminate in energy shortages which can lead to food and water shortages. Such shortages can, in turn, lead to irrational behavior, including major conflicts, and even World War III.

NUCLEAR ENERGY

The sun and the stars are huge thermonuclear reactors. Radiation from the sun heats the earth, grows our food and produces every facet of our weather. The development of nuclear energy on earth is inevitable. I regret that the first practical use was military. However, I believe it is extraordinarily fortunate that the first realization of that application was in the United States than in Hitler's Germany or Stalin's Soviet Union. A major world concern now is to fashion an international order which strongly mitigates against the use of weapons of mass destruction so that they do not threaten our survival or our freely chosen social structures. Non-proliferation and counter-proliferation of nuclear, chemical, and biological weapons are but one aspect of a much larger problem. Today, the world comprises islands of affluence in a sea of poverty. In our age of high technology and high mobility, such a situation can not be peacefully sustained. A major key to the solutions to the problems of poverty, hunger, and disease can, in my opinion, be found in the peaceful applications of nuclear energy, and that can contribute enormously to national security and international stability.

Current estimates project that world population will double during the next 40 years as will the requirements for energy. Energy is basic to every aspect of life from food production to telecommunications.

Where will this energy come from? Today, most of our energy derives from fossil fuels. There are two major problems with these sources. By the middle of next century the availability of oil and gas will be on a steep decline. Coal will be the choice of necessity. But all fossil fuels, and especially coal, are hazardous to our health. The recent world conference on the environment, the Kyoto conference, reached the consensus that it is imperative to limit the CO₂ influx into the atmosphere, in order to reduce the probability of disastrous climate change and also to halt the frightening increase of pulmonary problems such as asthma and emphysema, especially among children.

At a recent miniconference, sponsored by New Mexico Senator Pete Domenici, I had the occasion to say the following; *the recently concluded Kyoto Conference on Global Climate Change focused on the deleterious effects of burning fossil fuels. Computer modeling of the environment predicts increasing levels of CO₂, leading to serious global warming by way of the Greenhouse Effect. Whether or not we are actually seeing global warming, and if we are, whether it does more harm than good is arguable. The degree of economic damage and habitat dislocation that is likely to occur with increasing CO₂ levels are in even greater dispute. However, not arguable is the fact that environmental pollution, such as results from the production of nitrogen and sulfur compounds, are producing serious health effects for millions of people, especially children, and reducing the quality of life for many more. Also, not arguable is that US production of oil is declining year after year and that world reserves of oil and gas are being transparently depleted to the extent that, by the middle of the next century, their use will start to decline even as increasing population and pressure for improved living standards accelerate demand for more energy. To*

this scenario must be added the fact that the forests of the world are disappearing. Underdeveloped energy sources such as solar, wind and ocean currents may be helpful but cannot begin to compensate for our enormous dependence on fossil fuels.

So, what are our options for avoiding catastrophic energy shortages while protecting the environment? Hydroelectric power is almost fully utilized, already. Ocean tides and thermal ocean gradients have very limited potential. Geothermal energy, solar thermal and solar photovoltaic will become important contributions as will wind power. But all of these, together, can probably account for not more than 25% of total requirements. The only practical source of increased large-scale energy production, which is environmentally friendly, is nuclear energy by way of the fission process. Fusion energy is very far in the future.

Some months ago I presented a talk at the Shiprock, NM campus of the Navajo Community College where I summarized the science that underlies nuclear energy. One must realize that nuclear energy differs from fossil fuel energy in that it is one million times more efficient in the use of fossil fuel. There are, however, three main barriers which must be surmounted. They are not technical-they are social and political. In the US fission reactors, to be acceptable, must be transparently safe, waste products must be dealt with in a socially acceptable way and the by-products must be such that they are not readily available for producing nuclear weapons. I will briefly address each of these issues.

The world-wide nuclear energy enterprise, which now provides almost 20% of electrical energy, has been responsible for fewer fatalities and injuries than any combination of industries of comparable size, notwithstanding the early deaths and injuries caused by uranium mining and including the effects of the Chernobyl disaster. The Three-Mile Island accident destroyed a reactor but was not harmful to the people or the environment. Modern nuclear reactors have superb safety records and release less radioactivity to the environment than coal-fired plants of equal electrical output. Nuclear reactors are, today, fueled by uranium. They, therefore, produce plutonium which can be chemically separated from fuel rods and be used to make bombs. For nations that have signed the nuclear non-proliferation treaty there are international controls that deal with this problem, but not all nations have thus far signed that treaty. There is also the consideration that, in a uranium-based nuclear energy economy, uranium supplies will be exhausted in well under 100 years, unless breeder reactors are implemented. This, however, pose additional proliferation problems.

But now there is some good news. It was Niels Bohr who suggested that every problem bears within it the seeds for its solution. And so it appears with nuclear energy. Some decades ago, at the Chalk River Laboratory in Canada and, more recently, at the Los Alamos National Laboratory, there have been proposed ways to realize a type of nuclear reactor which operates below criticality and thereby avoids being subject to the most serious kind of reactor accident such as occurred at Three-Mile Island and Chernobyl, no matter what human or mechanical failures might occur. In addition this reactor can burn up plutonium and dramatically reduce the half-lives of other reactor by-products. The fuel rods utilized in this new reactor concept contain plutonium, but not in sufficient amount to permit the reactor to operate on its own.

They cannot sustain a chain reaction. The reactor comes to life when external neutrons flood the fuel rods. These external neutrons are produced by a high intensity proton beam which strikes a heavy element target at the center of the fuel rod assembly. Figure 1 indicates how the system works. The protons are accelerated to approximately 1 GeV by a linear accelerator much like the one Professor Seaborg approved for construction at Los Alamos when he was Chairman of the Atomic Energy Commission. The new concept has been given the name *Accelerator Transmutation of Nuclear Waste (ATW)*¹.

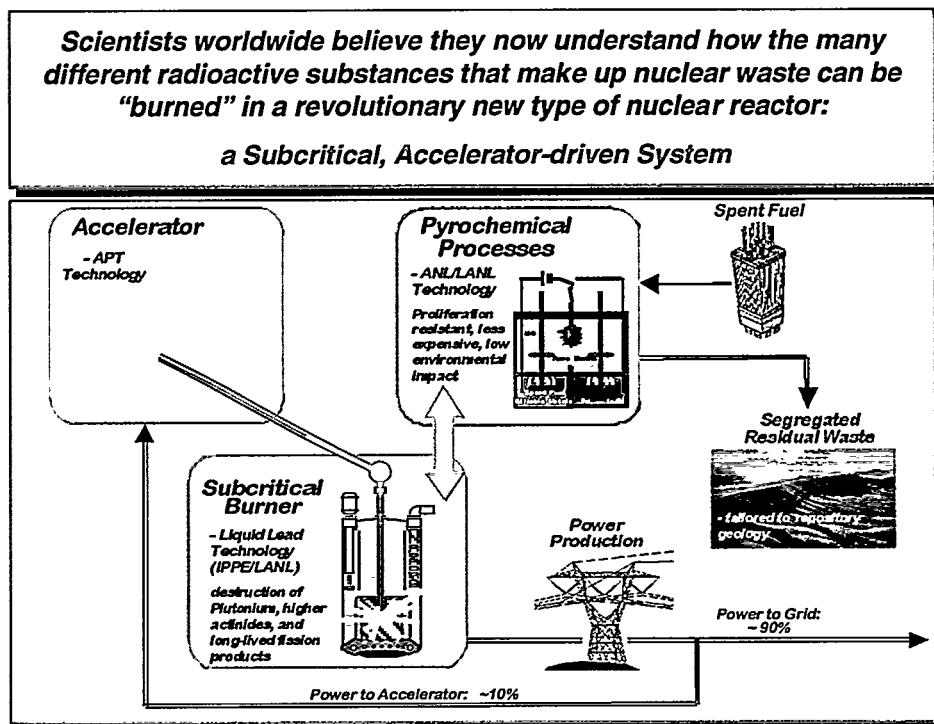


Figure 1. An ATW facility consists of three major elements: (1) a high-power proton linear accelerator; (2) a pyrochemical spent fuel treatment / waste cleanup system; (3) a liquid lead-bismuth cooled burner that produces and utilizes an intense source-driven neutron flux for transmutation in a heterogeneous (solid fuel) core.

¹ F. Venneri, M. Williamson, N. Li, M. Houts, R. Morley, D. Beller, W. Sailor, G. Lawrence, *Disposition of Nuclear Waste Using Subcritical Accelerator-Driven Systems*, Los Alamos National Laboratory Report LA-UR-98-985; and <http://www-adtt.lanl.gov>.

What you should remember about ATW is that (a) it is a *nuclear waste management enhancing* technology, (b) addresses issues of repository storage capacity and the longevity of radiological hazards, (b(1)) destruction of all transuranics, (b(2)) transmutation of Tc-99 and I-129, other isotopes if required, (b(3)) partition of all other fission products for optimal disposal, (b(4)) radiotoxicity of ATW waste after 300 years is lower than direct-disposal waste after 100,000 years, (c) it is reactor-like in scale and function; produces usable energy by destroying nuclear waste, (d) its components are based on proven technology, can be directly transferred to nuclear power production, (e) although spent fuel is processed, weapons-usable material is not made available, (f) it is adaptable to different fuel cycles, and (g) in the mission of waste destruction, it is significantly faster and more complete than other schemes. An additional bonus is that with the ATW concept one can use thorium, which is much more plentiful than uranium, as a fertile fuel to breed U^{233} which fissions like U^{235} but, absent U^{238} , so very little plutonium is produced.

But now human inertia and political correctness come into the picture. It has been 25 years since a nuclear reactor was licensed in the US. We now derive 17% of our electricity from nuclear power plants, but in about 20 years their licenses will expire and we will be in danger of losing the only energy source which can see us through the next century without serious damage to the environment and the health of our progeny. The abandonment of nuclear energy in the US will undoubtedly cost us world leadership and world influence, especially in matters such as the safety of nuclear reactors and other environmental concerns.

The biggest fly in the ointment appears to be radioactivity which fills the uninformed with fear and suspicion. So, the most important antidote is literacy in science, generally, and in nuclear science and biology specifically. That I believe, is a major purpose of this workshop. So what about the demon called radioactivity? I am 80 years old and have been working in radiation environments most of my life. I have suffered no ill effects even after being subjected to accidental fall out while within a few miles of a large nuclear detonation. As an experimental nuclear physicist, I have worked with radiation at levels well above natural background day after day and year after year. So what is the fuss about? At Los Alamos and also here at Crownpoint, New Mexico we live in a natural radiation environment of approximately 300 MR per year, about 1000 times less than what is required to produce serious harm. And people have been living in such environments since the beginning of time. Current government regulations, which to me defy common sense, dictate that man-made radiation should not expose the general public to more than 1% of natural radiation. What sense does this make in the light of research which has found no indication that low level radiation is harmful and in fact that such radiation may reduce illness and prolong life?

I leave this puzzle as an exercise for the student.

A, B, C's of Nuclear Science

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Abstract. Basic properties on the structure of nuclear science will be presented. Natural and man-made radiation and radioactivity processes will be described.

SCIENTIFIC BACKGROUND

All matter in the world, living or non-living, is made of about 90 basic substances called elements. Elements are substances which cannot be broken down into simpler substances. Examples are iron, oxygen, iodine, gold, etc. In Fig. 1 the

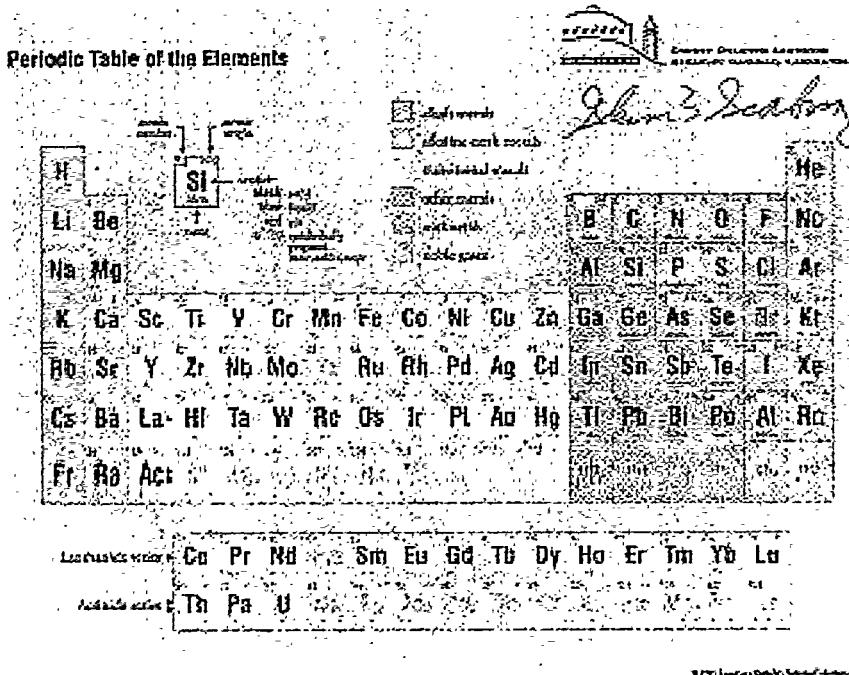
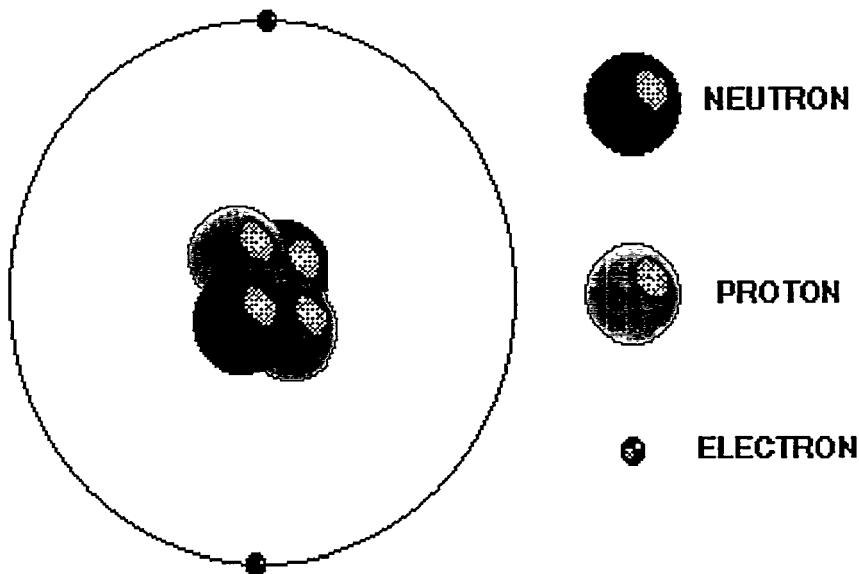


Figure 1. Periodic Table of the Elements.

periodic table of the elements is shown.

An atom is the smallest bit of an element that can exist and still have the properties of that element. Atoms with the same number of protons, but different numbers of neutrons are called isotopes of that same element.

Schematic diagram of a helium atom



MODEL OF A SIMPLE ATOM

Figure 2. Schematic Diagram of a Helium atom.

In Fig. 2 the schematic diagram of a helium atom is shown. Some of these atoms are stable, which means that they do not change with time; whereas others are unstable and undergo changes during their lifetimes. These unstable atoms actually change from one kind of atom to some other kind. Usually the new atom is also unstable and at some time will, in turn, change into yet another kind of atom. Hence, these unstable atoms are classified as radioactive atoms because while changing from one element into another, they release energy.

Each element has its own assortment of isotopes. In Fig. 3 the three different isotopes of lithium is shown. All of them have three protons (lithium always has three protons, otherwise, it is not lithium!) but they each have different numbers of neutrons.

Three isotopes of lithium

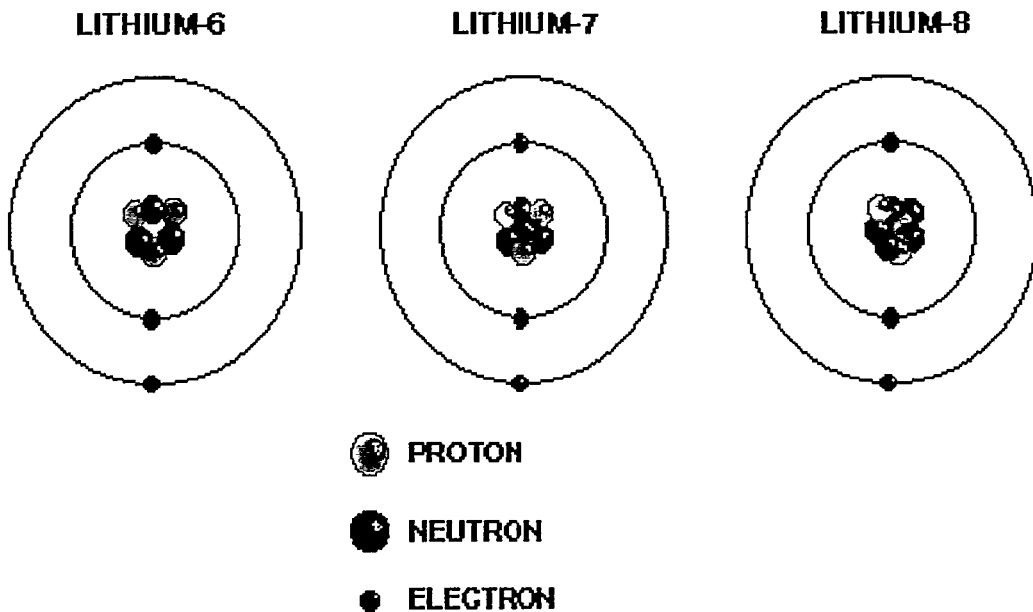


Figure 3. Three Isotopes of Lithium.

To distinguish the different isotopes of the same element, we add a number to their names. This number is equal to the total number of protons and neutrons in the nucleus and is called the atomic weight or mass number. The three kinds of lithium are thus called lithium-6, lithium-7 and lithium-8. If the isotopes give off ionizing radiation they are called radioisotopes. Lithium-6 and Lithium-7 are stable while Lithium-8 is unstable and decays with a lifetime of 0.8 seconds into Beryllium 8.

Radiation emission cannot be turned off or destroyed. It has to self-destruct. Different radioactive materials self-destruct or lose their strength over different periods of time. These periods are measured in half-lives: A half-life is the time it takes for the material to lose one-half of its radioactivity. For example, the half-life of radium-226 is 1622 years, which means that a given quantity of radium will lose half of its strength in 1622 years, and half of its remaining strength in another 1622 years, and so on. In Fig. 4 the beta- decay of Americium-242 is shown.

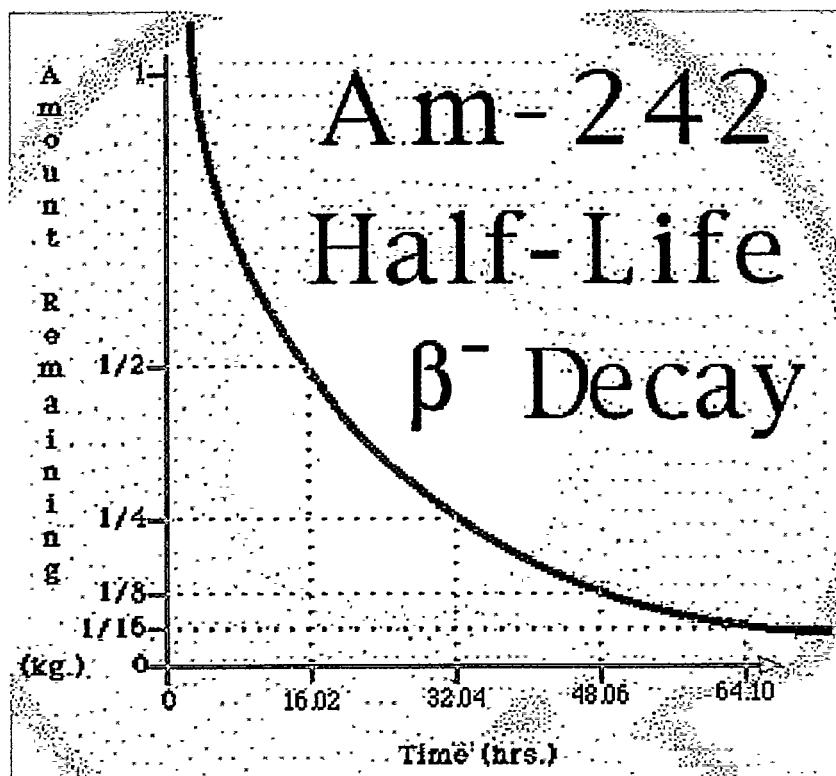


FIGURE 4. Beta- radioactive decay of Americium-242.

Uranium-238 has a half life of 4,500,000,000 years. Uranium is a naturally occurring radioactive isotope. It was there at the same time the Earth was created, some 5 billion years ago. The process of atoms changing is frequently referred to as radioactive decay. This is the process by which the energy stored in the nuclei is released.

FREQUENTLY ASKED QUESTIONS

Q:What is Radiation?

A: Radiation is energy traveling through space. Sunshine is one of the most familiar forms of radiation. It delivers light and heat. We enjoy the benefits of the sun every day of our lives, but as we have become increasingly more aware of some of the sun's harmful effects, we've begun to protect ourselves with sunscreens, sun hats, clothing and shade. As we learn about other sources of radiation, we can appreciate their benefits, understand how they work, and protect ourselves from any harms they might threaten. According to the dictionary, there are two physics definitions of the term radiation:

1. The emission and propagation of waves or particles (carrying energy).
2. The propagating waves or particles, such as light, sound, radiant heat, or particles emitted by radioactivity.

In other words, radiation is the emission of waves or particles -energy- in general. Here we are specifically discussing the energy emitted in radioactive processes.

Q: What else is also Radiation?

A: One particular form of radiation is what is called electromagnetic radiation. Electromagnetic radiation are waves from different sizes or wavelengths.

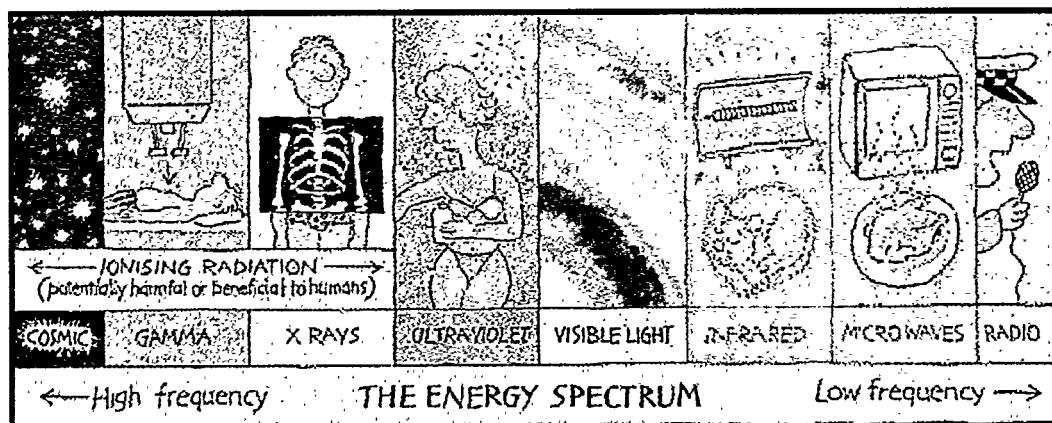


Figure 5. The Energy Spectrum.

Radiation that does not have the amount of energy needed to ionize an atom is called non-ionizing radiation. Examples of non-ionizing radiation are radar waves, microwaves and visible light.

Q: How do you measure amounts of radioactivity?

A: There are different measures for determining the quantity of radioactivity of something. Normally one measures the rate at which the radioactive isotopes decays in disintegration or decays per second (dps = international unit). 1 decay per second (dps) is also called 1 Bq (one Becquerel) in the honor of Henry Becquerel. The Curie (Ci) is the first unit which was used to measure amounts of radiation.

1 Ci = 2,200,000,000,000 dpm (decays per minute)

1 Ci = 370,000,000,000 dps (decays per second)

Since these numbers are so large, the usual way to write them is by multiples or submultiples of 1000. For example:

1 kilogram(1 kg) = 1000 gram

1 milligram(1 mg) = 0.001 gram

1 nanogram(1 ng) = 0.000000001 gram

1 picogram(1 pg) = 0.000000000001 gram

Thus, 1 pico Curie (1pCi) = 2.2 decays per minute (2.2 dpm)

Q: What are the units used to measure radiation dose?

A: There are many different units used to measure radiation dose, or the amount of energy deposited somewhere by radiation. Some of these units are international, i.e., it was internationally agreed that these units would be used all over the world to measure radiation and the energy deposited - the dose.

Roentgen: is the unit used for measuring ionization in air. It is defined as the quantity of X-rays producing an ionization of 1 esu per cubic centimeter in air. esu stands for electrostatic unit , i.e., the electric charge of one electron. 1R (Roentgen) produces 2.58 Coulombs in 1 kg of air at STP. The way to write it is: 2.58 C/kg.

$$1\text{mR} = 0.00258 \text{ C/kg}$$

RAD (Radiation Absorbed Dose) : is the unit for measuring absorbed dose in any material. The dose results from energy deposited by radiation. It applies to all types of radiation, but does not take into account the biological effect on the human body. Dose measures energy per mass. 1 rad = 100 erg/g. This means that 1 rad is going to deposited 100 ergs of energy in 1 gram of material.

REM (Roentgen Equivalent Man) : is the unit for measuring dose equivalent pertaining to humans. REM's take into account the energy absorbed and the biological effect of due to different types of radiation. The REM is gradually being replaced by the term Sievert (Sv): 1 Sievert = 100 rem. REM takes into account how ionizing is the radiation, how much energy it can deposit.

THE NATURE OF RADIATION

Radiation can be *natural* or *man-made*. They differ in their origin, but their composition is the same, i.e., they are made of the same stuff: particles or waves.

Natural

Cosmic Radiation

The cosmic radiation we receive on Earth has its origins in the interaction of primary cosmic rays with the Earth's upper atmosphere. Primary cosmic rays permeate all of space and are composed of highly energized, positively charged particles (nuclei) as well as high energy photons and electrons. The origins of the so-called primary cosmic rays is not known for sure, although we know they can be produced in stars, starts explosions or other astrophysical events.

As they approach the Earth, primary cosmic rays interact with the Earth's protective atmosphere and magnetic field. As a by-product of these interactions, many radioactive isotopes and other particles are formed, the secondary cosmic rays. Not all secondary cosmic rays are prevented from reaching the earth's surface. The higher you are in altitude, the more you are exposed to cosmic radiation. In fact, the average amount of exposure to cosmic radiation that a person gets in the roughly doubles for every 6,000 foot increase in elevation.

EAS Development in the Atmosphere

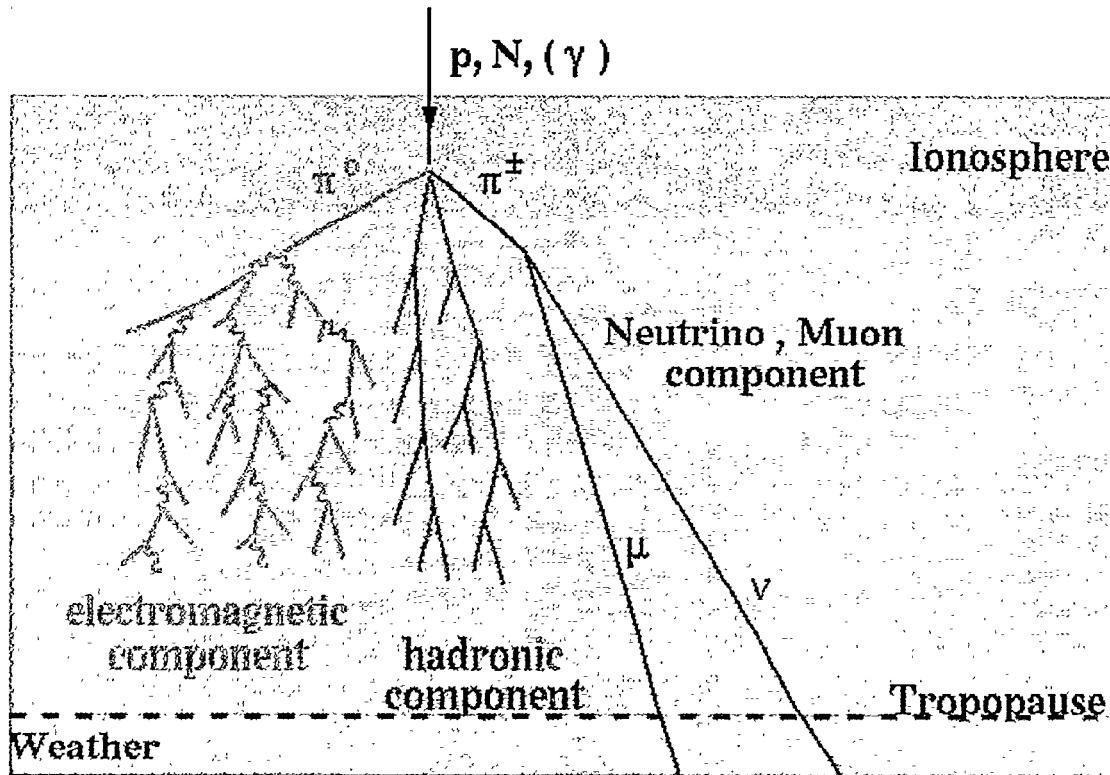


Figure 6. Extensive Air Shower(EAS) Development in the Atmosphere.

Terrestrial

There are natural sources of radiation in the ground, rocks, building material, and drinking water supplies. The origin of this radiation is what we call *naturally occurring radioactive isotopes*. They are naturally occurring because they were incorporated in the Earth at the same time the Earth was created, some 450,000,000 years ago... Since their half lives are very long (some 100,000,000 or 4,000,000,000 years...) they are still around. They are the origin of natural radioactive chains, and produce other radioactive isotopes. Examples are the 232Th (Thorium) and 238 U(Uranium) chains. Like all of the elements heavier than iron, uranium and thorium are believed to be created in star explosions, called Supernovae.

The food we eat and the water we drink all contain some trace amount of natural radioactive material. This naturally occurring radioactive material can get incorporated in our tissue or be eliminated. Some naturally occurring radioactive isotopes commonly found in food and water are C-14, Ar-41, and K-40.

K-40, is a radioactive isotope of potassium. 0.0117% of all potassium is K-40. This means that if we drink, for example apple juice which contains 0.260 g of potassium, 0.000030 g will be K-40. If you do the correct math, you are drinking about 8 Bq or dps (decays per second) of K-40 with your apple juice.

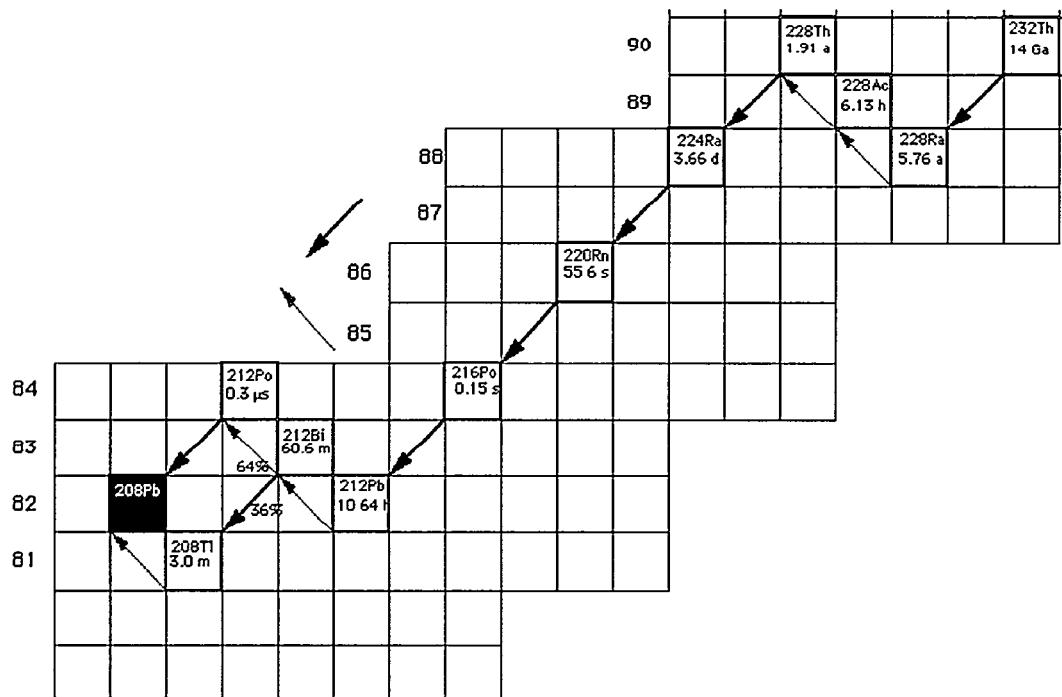


Figure 7. The thorium radioactive series.

Man-Made

Man-made radiation found in the environment is originated from medical and dental diagnostic procedures, atmospheric tests of atomic bombs, emissions from nuclear plants, certain occupational activities, and some consumer products.

Man-made isotopes have medical or industrial applications. A good example is 131-I. Because of the chemical affinity of iodine for the thyroid gland and the relatively short half-life of Iodine-131, that radioisotope is used as a drug to treat certain diseases involving the thyroid. Its radioactivity drops to a negligible value in a length of time that is reasonable for such medical treatments. These isotopes are used to:

Generate Power

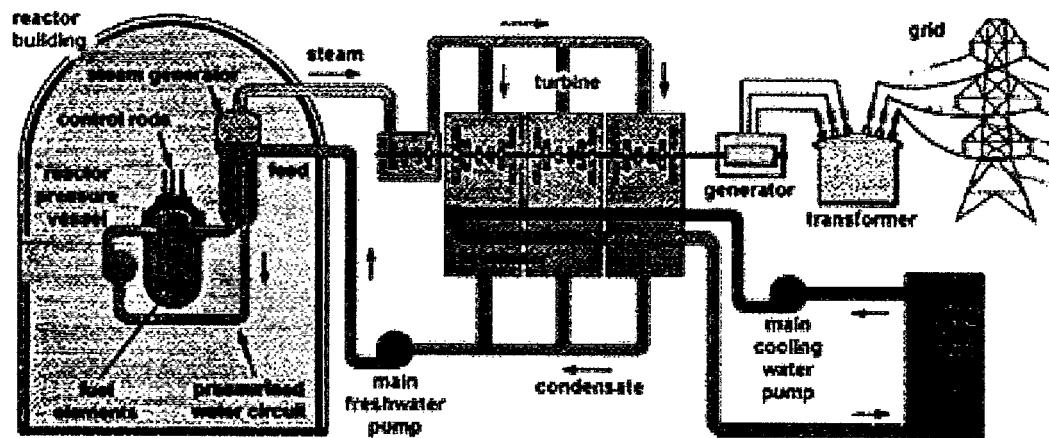


Figure 8. Nuclear Reactor Power Plant.

Medical Diagnoses

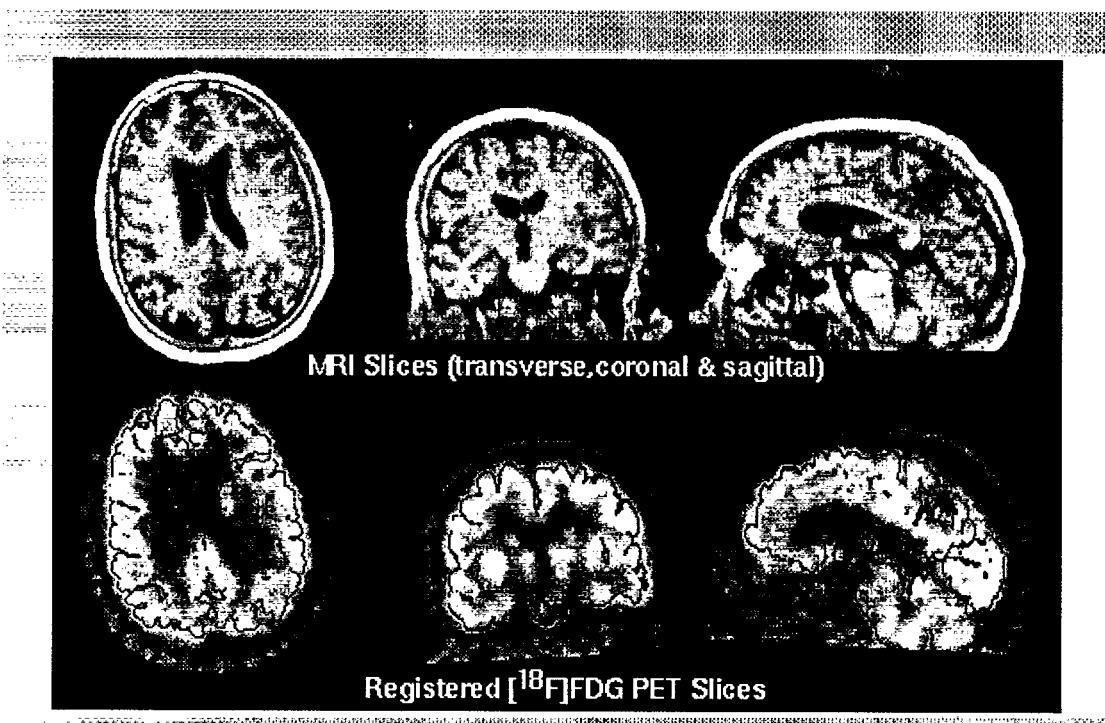


Figure 9. MRI Images of Brain.

Household Items (smoke detector)

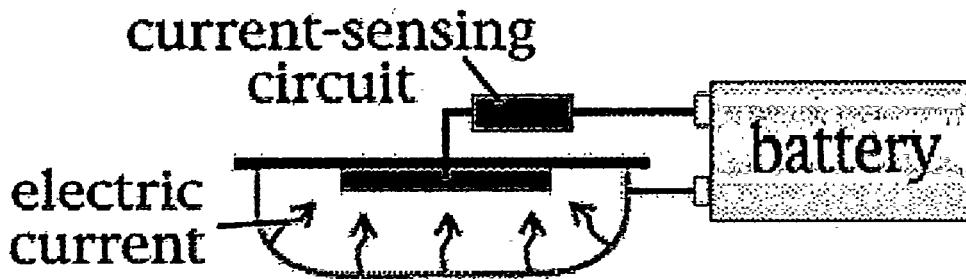


Figure 10. Smoke Detector System.

Industrial Applications

- Radioactive tracers (AKA: Tribology) can be injected into a mechanical system to find any problems and discover how machinery wears. Much like scientific and medical tracers, industry uses tracers to ensure that machinery is running well.
- Sensitive gauges and dials utilize radioactive materials. These can be used in various areas of industry as well as in defense, for example in anti-aircraft missiles.
- Equipment can be sterilized by irradiation to ensure that it is sanitary for any of a variety of uses.
- Machinery X-rays function much like X-rays we receive at the dentist's office. They can illustrate a weakness in a given mechanical system. Machinery can get stress fractures just like humans can, and machinery X-rays save time and money, by diagnosing the exact cause and location of a problem.

IONIZING RADIATION

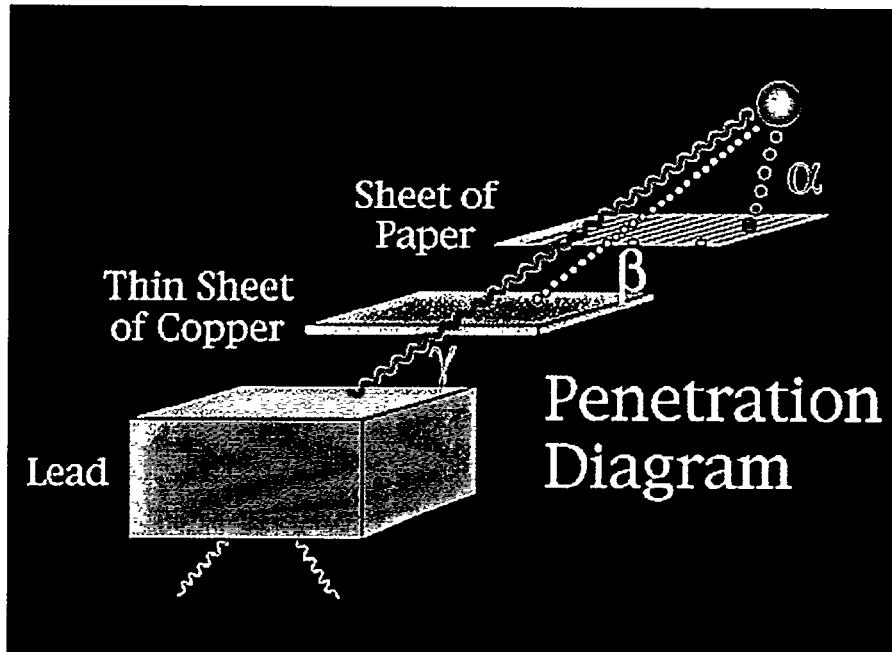


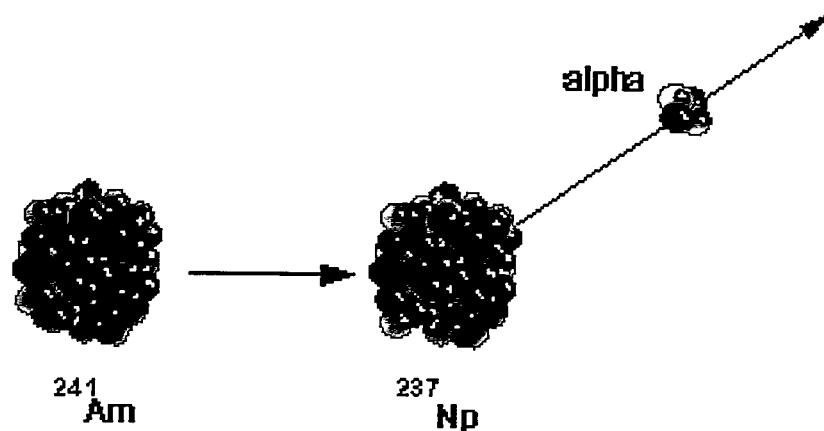
Figure 11. Stopping Power of Ionizing Radiation.

Ionizing radiation is potentially harmful to living cells. As it penetrates the material, it kicks electrons from their orbits, producing electrically-charged nuclei, or ions in the materials it strikes. This process is called ionization. Ionizing radiation affects the large chemical molecules of which all living things are made and might cause changes which are biologically important.

There exist three different types of ionizing radiation: Alpha, Beta and Gamma. In Fig. 11 we show that a sheet of paper can stop alpha particles, a thin sheet of copper can stop beta particles, and several inches of lead can stop the gamma rays.

Alpha Radiation

An example of alpha radioactivity



ALPHA DECAY OF AMERICIUM-241

Figure 12. Alpha Decay of Americium-241.

Alpha particles have a large mass and consists of two protons and two neutrons. They are Helium (He) nuclei and have a positive charge of plus two. This positive charge causes the alpha particle to strip electrons from nearby atoms (ionizes them). In Fig. 12 we show the alpha decay of Americium-241.

Alpha radiation can penetrate only two or three inches of air, or can be stopped completely (absorbed) by a single sheet of paper or human tissue of equal thickness. The very outer, non-living layer of human skin is thick enough to protect us from external alpha radiation.

Beta Radiation

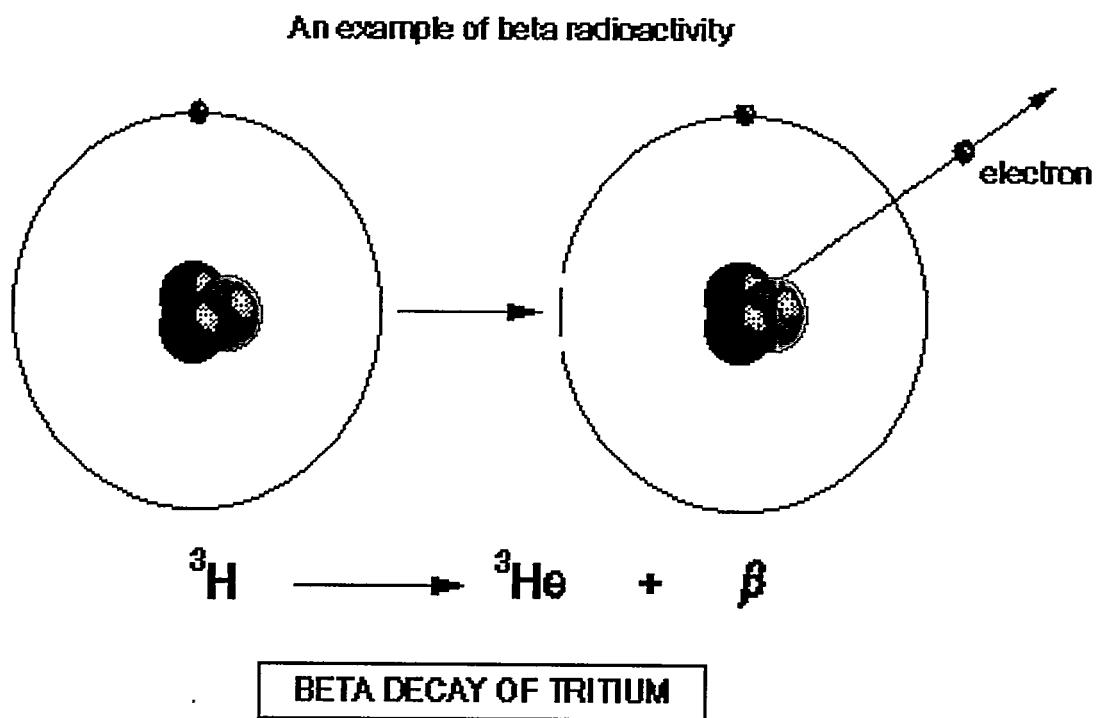


Figure 13. Beta Decay of Tritium.

Beta particles have a small mass and an electrical charge of minus one. They are electrons. Beta radiation, like alpha, is released in some decay processes, but has more penetrative power than alpha radiation. It can penetrate 1/2 to 3/4 inch of water or flesh, the thickness of a human hand. An eighth of an inch of aluminum, however, will stop or absorb most beta rays. In Fig. 13 we show the beta decay of tritium.

Beta rays are emitted by some naturally-occurring radioactive materials and by some of the waste products of nuclear reactors. Beta rays are very useful in the treatment of skin diseases.

Gamma-Rays and X-rays

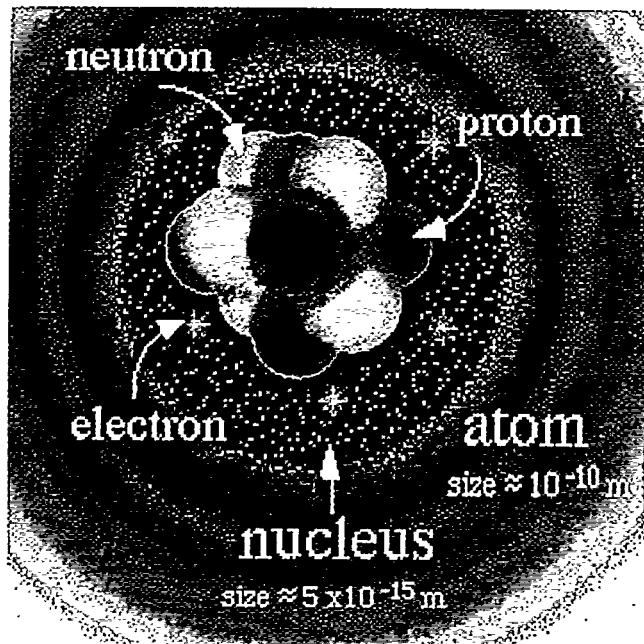


Figure 14. Gamma-ray emission originates from the nucleus and x-ray emission originates from the electrons.

Gamma rays and X-rays are the same type of radiation, although the customary origin of the radiation is different. Gamma rays originate in the nucleus and X-rays are atomic. Typically, gamma rays are more energetic than X-rays. In Fig. 14 we show the relative sizes of the atom and the nucleus.

Gamma/X-ray radiation is electromagnetic, like visible light. It has no electrical charge. It can ionize matter as a result of direct interactions with orbital electrons.

Gamma rays are produced in the radioactive decay process and occur with a wide range of energies, dependent upon the radioactive material that they come from. They can penetrate the human body, which absorbs only a small part of them. Three to four feet of concrete or several inches of lead are needed to completely stop or absorb gamma rays.

Also considered radiation but **not** ionizing is neutral particles such as neutrons. Neutrons collide with the nucleus of atoms and are not able to interact directly with electrons due to their lack of charge. Neutrons can stop in materials that have lots of hydrogen where their energy is dissipated through elastic neutron-proton collisions.

Neutron radiation is generally of little concern to the public as it exists primarily inside nuclear reactors where the neutrons can be adequately contained. Some neutrons are produced naturally at very high altitudes but do not reach the earth. Neutron therapy is extremely useful for treatment of some diseases.

WHAT RADIATION EXPOSURES OR DOSES PEOPLE GET?

Different kinds of radiation and their sources have been mentioned. Let's take a closer look on how they contribute to the dose people normally get.

Most of the radiation doses we discuss in reference to exposures of the public are less than one rem, so we often speak in terms of 1/1000th of a rem -- a millirem. The pie chart below taken from NCRP Report No. 93, illustrates the sources of radiation exposure to people in the US. It indicates that the average exposure of persons in this country is about 360 millirems in a year.

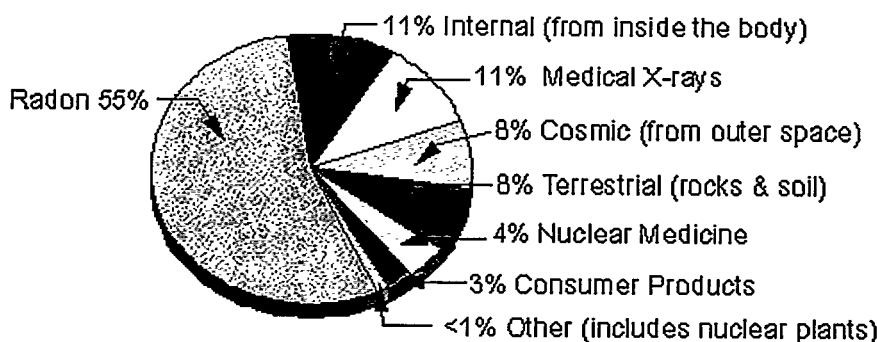


Figure 15. Sources of Radiation.

About 82% of our exposure to ionizing radiation is from natural sources in the environment. The largest contributor to the total, about 55%, is radon, a radioactive gas which originates in natural radioactive isotopes, present in practically all types of soil and rock.

The other exposures from natural radiation are those from cosmic radiation from outer space (about 8%), terrestrial sources (about 8%), internal radiation, primarily from radioactive potassium in our bodies (about 11%). The sum of all of these is an average exposure of almost 300 millirem in a year.

Medical x-rays account for an average of only about 11%, and nuclear medicine, another 4%, of the average person's radiation exposure, for a total of about 54 millirem. These medically related exposures generally have clear-cut benefits to our health.

Radiation exposure from commercial applications of ionizing radiation contributes only about 3%, or 11 millirem, to our total exposure. Among the most important of these sources of radiation are our domestic water supplies, building materials, mining and agricultural products, and fuels, particularly coal. Each of these applications is generally associated with some net health benefit. The average exposures from these sources are small and are often expressed in microrem (millionths of a rem) rather than millirem.

Further information on the basic properties of nuclear science can be viewed at the WEB site: http://user88.lbl.gov/NSD_docs/abc/.

Nuclear Medicine

100 Years in the Making

1896-1996¹

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INTRODUCTION²

You're about to start an exercise program after years of sedentary living. Is your heart up to it?

To find out, your doctor might order a nuclear stress test—one of the most common procedures in nuclear medicine today.

Your daughter hurt her leg running around. Is it a stress fracture? A bone scan will let you know.

Or perhaps someone in your family suffered from an overactive thyroid gland, a condition known as Graves' disease. This ailment is easily treated with nuclear medicine.

Nuclear medicine uses tracers—tiny amounts of radiative substances—to diagnose or treat disease.

These substances pass harmlessly through the body. But because they are radioactive, their movement can be detected with special devices, yielding a wealth of information about bodily processes.

The essence of nuclear medicine is that it can “visualize” changes in the function and biochemistry of body organs and tissues. Measurements of such changes offer unique information for diagnosis and therapy that can't be obtained through other tests.

One of the great benefits of nuclear medicine is that regional abnormalities in the body often can be detected before an abnormality is noted in over-all organ function—before the patient “feels something is wrong.”

For example, problems with lung function can be detected before measurements of overall lung ventilation fall outside the normal range. Smokers often

¹ GT Seaborg and HN Wagner, Jr., *Nuclear Medicine: 100 Years in the Making: 1896-1996*, Society of Nuclear Medicine, 1996.

² HN Wagner, Jr., MD, Director, Division of Radiation Health Sciences, John Hopkins University, Baltimore, Maryland.

do not detect that their lungs are deteriorating until more than 70 percent of their lungs is impaired. Similarly, regional chemical changes can be measured before abnormalities can be found in the concentration of chemical constituents in blood or urine.

Early detection allows a disease to be treated before it becomes advanced, when there is generally a better outcome.

Radiation measurement devices are so sensitive that they detect the radiation from a single atom. Every time you hear a radiation detector go "click," you are hearing the disintegration of a single atom. This sensitivity makes it possible to administer very small amounts of radiation to an individual to "image" the patient in a nuclear medicine procedure.

The radiation dose to the patient from a diagnostic nuclear medicine scan is very low-about the same as the radiation dose the patient receives from the environment, or "background radiation." Background radiation comes from the sun, the ground, and even elements normally found in the human body.

At high doses, radiation can be used to destroy tissues, sometimes curing the patient.

The therapeutic uses of radioactive isotopes such as phosphorus-32 (to treat leukemia and polycythemia vera, an excess of red blood cells and platelets in the blood) and iodine-131 (to treat thyroid disease) truly were revolutionary.

Currently, radionuclide therapies are being tested for treatment of many types of cancers, as well as arthritic joints, and are very promising. Radionuclide therapy is being used to relieve pain associated with cancer that has spread to the bone.

Nuclear medicine has dramatically changed the way medicine is practiced in hospitals all over the world. In the future, it promises still more wonders-more precise diagnoses, better treatment of diseases, and a deeper understanding of the functioning of the human mind.

"The history of the living world can be summarized as the elaboration of ever more perfect eyes in a cosmos in which there is always something more to be seen," wrote French philosopher Teilhard de Chardin.

Nuclear medicine provides "more perfect eyes," allowing us to "see" inside the processes of life.

It has been a century since Henri Becquerel discovered the mysterious "rays" from uranium, which Marie Curie later named "radioactivity." In the following pages we celebrate the first 100 years of nuclear medicine.

THE FIRST 50 YEARS³

The early researchers in what is now called "nuclear medicine" had no idea that their work would revolutionize the diagnosis and treatment of disease and injury.

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Like their counterparts in other fields, they built their discoveries, driven forward by their fascination with an exciting new frontier of science. They gave little thought to the possibility that an object of their search might have practical value.

But as we often see in science explorations, they were in for some surprises.

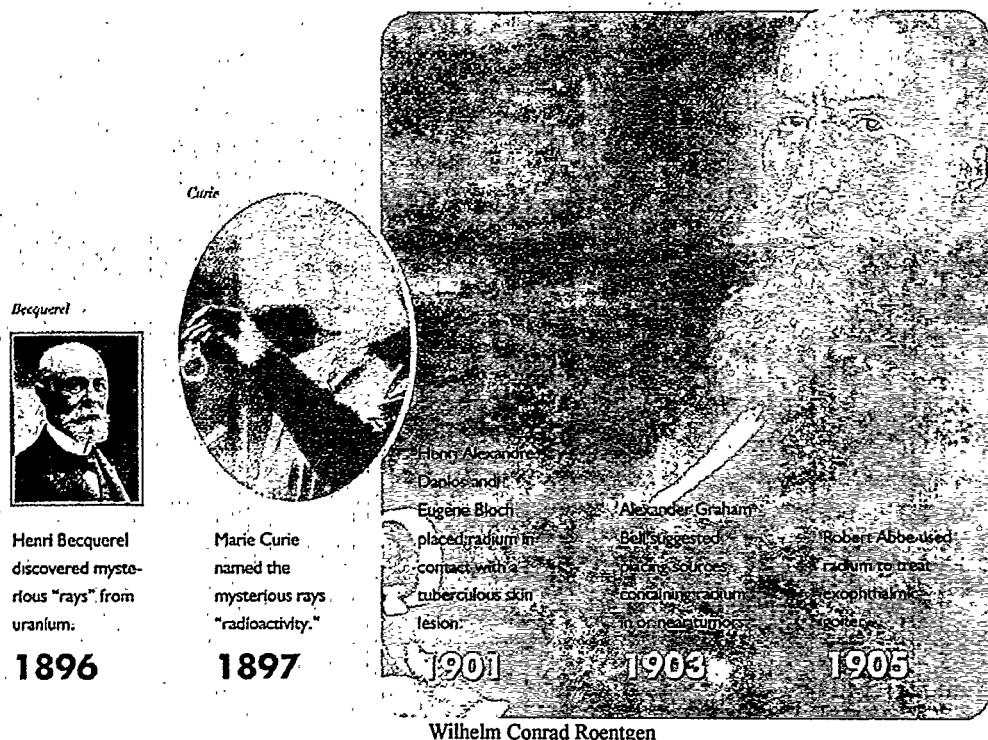
I speak from personal experience. For example, medical researchers on two occasions expressed hope that radionuclides of specific elements with particular half-lives would be found, and we were fortunate enough to hit the jackpot. As the years passed, we witnessed the appearance of many a delayed jackpot, some of which had a positive impact on my own health and that of my immediate family.

At the time of our discoveries, we would have scoffed at the idea that many radionuclides would have any practical value-yet some of the most scorned have turned out to be among the most valuable.

There is a lesson here: Very often, we cannot predict the practical applications of basic science discoveries, but we can predict that some applications will be found later, to the enduring benefit of humankind.

As discoveries typically are built on previous findings, the discovery of nuclear radiation resulted from the invention of the photographic plate.

In 1895, physicists throughout the world were excited by Wilhelm Conrad Roentgen's discovery of a ray that could penetrate the human body and reveal broken bones. Henry Becquerel, newly appointed to the chair of physics at the Ecole Polytechnique in Paris, had the idea that X-rays and visible fluorescent light might be



produced by the same mechanism. In early 1896, Becquerel tested this hypothesis. He exposed various crystals to bright sunlight and then placed each of them in a black paper envelope with an unexposed photographic plate, which would record any fluorescence that might be produced.

To his delight, he indeed observed this effect when he used a phosphorescent material, uranium potassium sulfate.

Then he made a confusing discovery.

One day, Becquerel happened to develop a photographic plate that had been stored in a dark drawer next to some uranium salt that had never been exposed to sunlight. To his surprise, the plate was darkened where the uranium salt had been in contact with it—even though the sun could not possibly have produced fluorescence. He concluded that uranium in its normal state gave off X-rays or something similar.

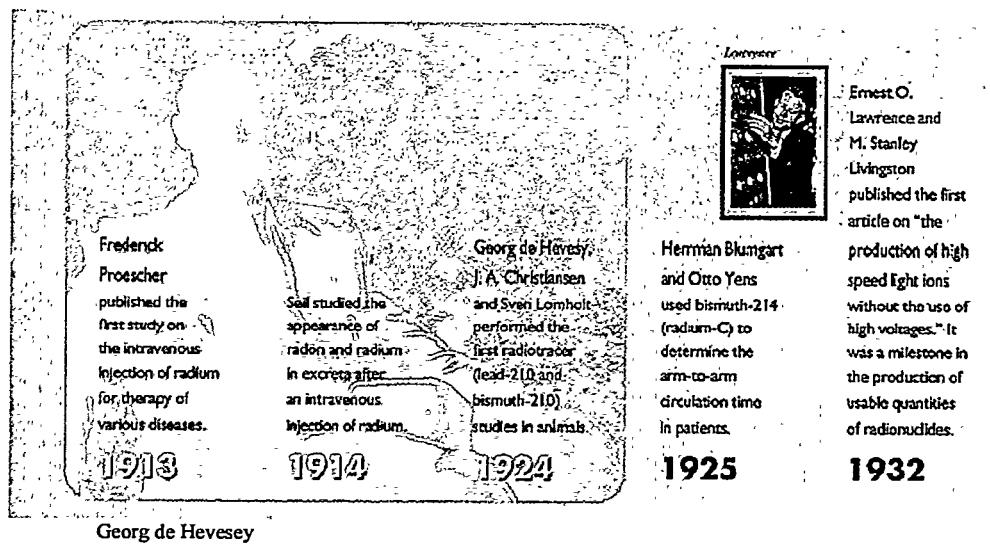
As often happens in scientific research, the wrong hypothesis led to one of the most important discoveries of all time: radioactivity.

In 1897, Becquerel's friend Pierre Curie, also a professor of physics in Paris, suggested to his young bride, Marie (Marja Skłodowska-Curie), that she study the phenomenon discovered by Becquerel for her doctoral thesis. (some references indicate that Becquerel suggested her doctoral thesis. As with any good idea, everyone would like to take credit, but the credit really goes to the person who does the work and makes the discovery.)

In her research, Marie Curie found that both uranium and thorium possessed mysterious properties and—what was more surprising—that some of the components of uranium minerals were more radioactive than purified uranium itself.

“We shall call the mysterious rays *radioactivity*,” she told Pierre, and the substances that produce the rays “radioelements.”

In 1898, Pierre abandoned his study of crystals to help Marie search for the mysterious rays from uranium.



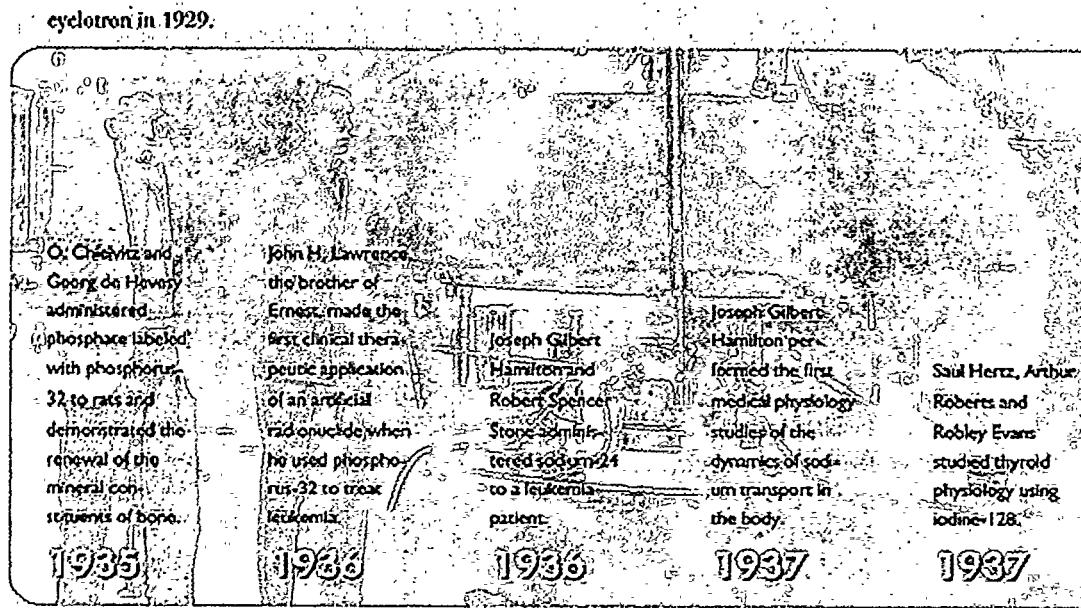
That July, the Curies reported the discovery of polonium (Po-210) named in honor of Poland, Marie's birthplace. In December 1898, they announced the discovery of yet another element that emitted the mysterious rays, and they named this element radium (Ra-226). For their work, Becquerel and the Curies received the Nobel Prize in physics in 1903.

LAWRENCE INVENTS THE CYCLOTRON

For more than 30 years, X-rays, radium and a few anecdotal uses of artificially produced radionuclides were the only sources of radiation used in medical diagnosis and therapy. During this period, an understanding of radiation and radionuclides was enhanced by the work of many great scientists-Joseph Thomson, Ernest Rutherford, Frederick Soddy, Otto Hahn, Lise Meitner, John Cockcroft and Harold Walton, the Joliot-Curies and others.

Their work increased our understanding of the atom and laid the foundation of the nuclear age, but it did not have any direct and broad application in medicine. A great step in this direction-and it was a giant one indeed-was Ernest O. Lawrence's invention of the cyclotron in 1929.

The cyclotron rapidly developed in the 1930s and allowed us to create a number of new radionuclides, many of which proved useful in biological and medical work.



M. Sherry Springer and Ernest O. Lawrence stand in front of the 28-inch cyclotron

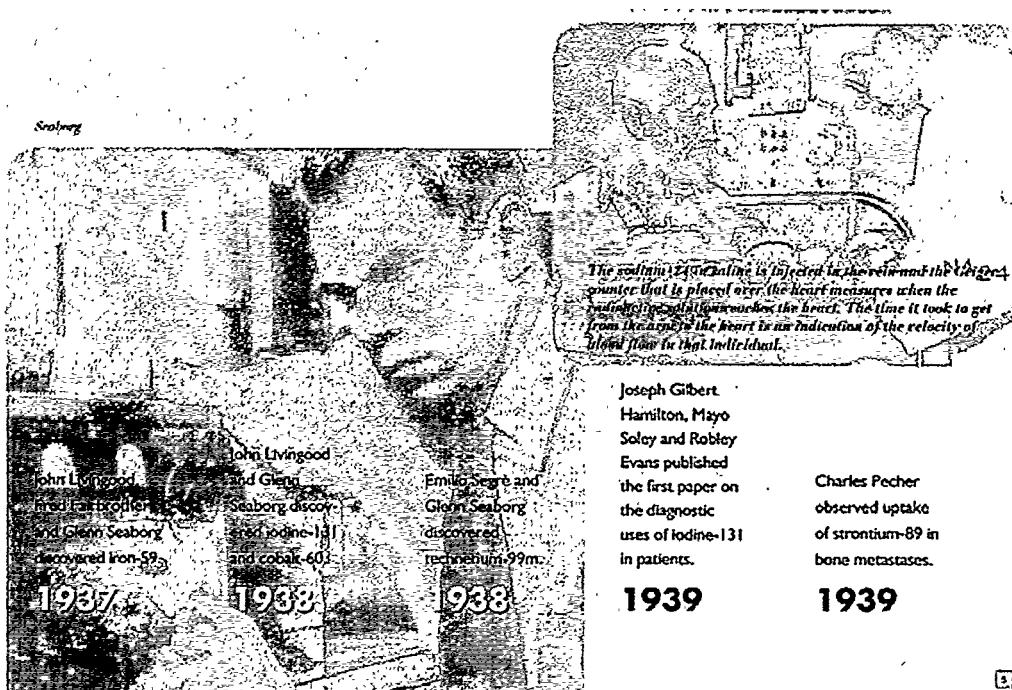
I said "us" in mentioning the production of these new isotopes because I had the extreme good fortune to be part of a group of young scientists at the University of California at Berkeley at the time of Lawrence's work, and was able to use his new cyclotron in my work in nuclear chemistry. I was most concerned with learning about the nucleus of the atom, but we were kept aware of the utilitarian aspect of our work by requests from our biological and medical colleagues for new radionuclides.

My contributions to nuclear medicine resulted from an accidental encounter.

In spring 1936, I was a second-year graduate student in the Department of Chemistry at the University of California at Berkeley. One day, I ran into a young physicist, John J. Livingood, somewhere between the chemistry building and the physics building. He said he had a tin target that was being bombarded with 5 million electron-volt (MeV) deuterons in the 27-inch cyclotron at that very moment, and he asked if I would like to perform the chemical separations needed to identify the transmutation products. The chemical operations would have to be performed immediately after the bombardment was completed, which was within the next hour.

Naturally, I jumped at the chance. Within an hour, I literally was handed a "hot" target and given some work space in the physics building—tap water, a sink, a primitive hood and a small workbench.

So began my career.



As the scientific community became knowledgeable about the ability of the 27-inch Lawrence cyclotron (later replaced with a 37-inch cyclotron) to produce "artificial" radionuclides, it started asking for radionuclides of certain elements.

Professor George H. Whipple of the University of Rochester (N.Y.) was hoping for a reasonably long-lived radioisotope of iron for use in his studies of hemoglobin in human blood.

We bombarded an iron target with 5.5 MeV deuterons and discovered iron-59 (Fe-59), which has a half-life of 44 days. Shortly thereafter, we sent Dr. Whipple the Fe-59 made from the first production run. Whipple's experiments using Fe-59 opened a new era in our understanding of iron metabolism and blood production.

DISCOVERY LEADS TO NEW THYROID TREATMENT

Perhaps the most interesting of all my collaborations with Livingood, and one that has special meaning to me, was the discovery of iodine-131.

One day in spring 1938, Joseph G. Hamilton mentioned the limitations on his studies of thyroid metabolism imposed by the short half-life (25 minutes) of the radioactive iodine (I-128) then available to him. When he inquired about the possibility of finding an iodine radionuclide with a longer half-life, I asked him what half-life would be best for his work. "Oh, about a week," he replied.

Shortly thereafter, Livingood prepared tellurium targets that we bombarded in the 37-inch cyclotron, some with 8 MeV deuterons and some with neutrons. I put the targets through chemical separations and identification procedures, wearing a mask and obstetrical gloves to avoid acquiring the dreaded "tellurium breath." The presence of tellurium in the body can be detected by a strong garlic-like odor on the breath.

Soon we were able to identify iodine-131 (I-131), and luckily enough, its half-life turned out to be eight days. Today, this isotope is used worldwide to treat thyroid disease. My mother was one of the first to benefit from the use of this radionuclide. She suffered from hyperthyroidism, a condition related to the one from which her sister died. But diagnosis and treatment with I-131 led to my mother's complete recovery. She lived to be 81.

My role in the discovery of technetium-99m (Tc-99m) and cesium-137 (Cs-137) involved collaboration with other colleagues, Emilio Segre (Tc-99m) and Margaret Melhase (Cs-137). At the time, we had no expectations concerning the work's ultimately beneficial applications to medicine. But today, Tc-99m is the most widely used radioisotope in vivo diagnostic medicine and has become one of the mainstays of nuclear medicine. My wife, Helen, and I have benefited from diagnosis with Tc-99m. We were accorded the peace of mind that comes with the determination of a clean bill of health.

The 60-inch cyclotron at Berkeley was the original source of phosphorous-32 (P-32) for medical uses. But during World War II, the cyclotron and most of the associated personnel were diverted to the top secret Manhattan Project, which was developing an atomic bomb.

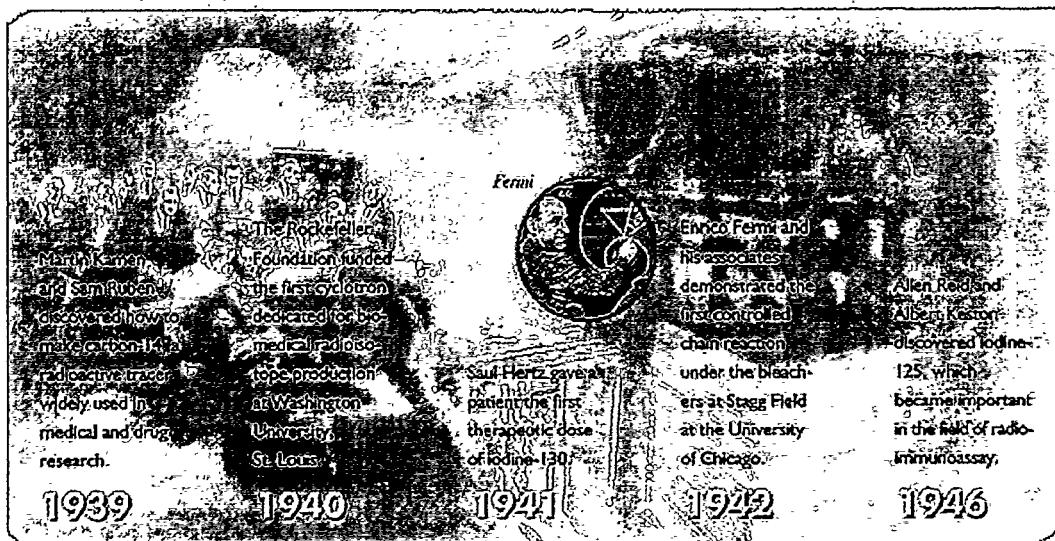
Meanwhile, the cyclotron at the Massachusetts Institute of Technology, which went on line in 1940, provided radionuclides exclusively for research and medical uses.

OAK RIDGE REACTOR STARTS OPERATING

In November 1943, a new source for medical radioisotopes became available: the nuclear reactor at Oak Ridge, Tenn.

The reactor's existence had to remain a secret because of its involvement in the Manhattan Project. Reactor-produced P-32 was shipped to Berkeley and, from there, shipped to medical centers—under the pretense of being cyclotron-produced.

To provide additional cover for the Manhattan Project, Berkeley's John Lawrence—who had been treating leukemia patients with radiophosphorus for many years—was instructed to continue his P-32 work with many publications and lectures.



Stagg Field reactor

Radon in the Environment

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INTRODUCTION

The introductory paragraphs for this workshop written by Fred Begay(1) describe the historical impact of uranium mining on the Navajo Reservation. The lack of understanding by health officials and the urgency of acquiring large amounts of uranium isotopes in a short time for a crucial wartime effort 50 years ago has resulted in many lung cancer deaths among Navajo miners from radon inhalation and environmental degradation because of the mine tailings. The central role played by the Navajo in these historic events is the reason for the unusually heavy price paid in suffering and anxiety about the consequences of exposure to uranium and other naturally radioactive elements in the earth's crust.

Although the dangers of radiation are generally accepted, the amount of radioactivity that is harmful is a subject of disagreement and the protection standards we observe and enforce are more conservation than the evidence than the evidence gathered from controlled animal experiments and clinical observation justifies. This paper attempts to identify the features of man's contact with naturally radioactive materials that are truly dangerous. It also describes interactions that are not especially hazardous and, finally, suggests steps that can be taken to minimize or eliminate exposure to potentially harmful radionuclides that have always existed in our environment.

THE NATURAL RADIOACTIVE ELEMENTS

The naturally radioactive elements are widely distributed in rocks and soils. The energy release can be totally accounted for by the decay of potassium-40, uranium-238, uranium-235, thorium-232 and their decay products. Even though radioactivity can be detected from each decay in an unstable element, most of the events occur in locations or under conditions where they pose no threat to man.

Nevertheless, others result in exposure and are the root cause of the cancers and the environmental degradation we have seen as a result of early uranium mining.

Potassium-40 is uniformly distributed throughout all the potassium that occurs on earth and is absolutely essential to the survival of all plants and animals. Although most of the radiation dose man receives from internal radionuclides comes from potassium-40, there is no choice about the presence of this rare potassium isotope in our bodies and the significant radiation dose we receive from potassium-40 decay over our lifetime. There is also no evidence that the radiation dose we receive is harmful. We irradiate both ourselves and those close to us with gamma rays from potassium-40 every sleeping and waking moment and there is no escape. Because the radiation dose from potassium-40 is simply unavoidable and the elimination of potassium from our bodies would be fatal, we accept that we cannot completely prevent radiation exposure and turn our attention to other natural radiation sources we can control.

Thorium-232 is abundant in the earth's crust, long-lived and contributes somewhat more environmental radioactivity than the uranium isotopes simply because there is so much of it. Although colloidal thorium dioxide was used clinically for almost 30 years as an x-ray contrast medium, it is no longer given to patients and the only people at risk received the material, called Thorotrast, as intravenous injections over 40 years ago. There is no known environmental radiation hazard from thorium decay in the population at large and even the Thorotrast-injected patients have low risks for the development of bone and liver cancers and leukemia(2).

The uranium isotopes account for the remainder of the natural radiation in the form of decay products of the abundant(over 99 %) uranium-238 and the rare uranium-235 and uranium-234 isotopes. Because of their shorter half-lives, the rare isotopes contribute about half of the natural uranium radioactivity. These isotopes have complex decay schemes shown in Fig. 1. However, the formidable detail in these schemes tends to obscure the real problem, the effects of certain of these isotopes on human health and the steps that can be taken to minimize or remove the human health hazard. Uranium isotopes have two potential health effects arising from chemical toxicity and alpha radiation. The chemical toxicity is specifically kidney damage and no other organs have been shown to be involved. The alpha radiation from massive doses of uranium oxide deposited by complex techniques in the lungs of experimental animals has caused lung cancer(3,4). However, environmental human exposures have not been high enough to cause kidney damage or lung cancer. The hazard from uranium exposure is very low to negligible.

Radium-226, a decay product of uranium-238 has caused bone cancers in radium dial painters and radium chemists exposed during the first few decades after its discovery. The dimensions of the hazard from radium-226 are well understood in quantitative terms and natural radium exposure does not appear to pose a significant risk in the general population(5). The risk is low because the concentration of radium-226 is not high enough to cause appreciable bone cancer. In cases where intense effort was spent to concentrate and isolate radium for medical and industrial applications, occupational exposures occurred in earlier times but negligible with modern

understanding and safeguards. For example, radium needles are very useful in the modern treatment of prostate cancer but radium injections are never given.

The serious cancer problem stems from human exposure to further decay products of the abundant isotope, uranium-238. Radon-222 is a noble gas that can diffuse great distances and because it is mobile, it can concentrate in confined spaces.

U 92	^{232}U (U.I) $4.51 \times 10^9 \text{ y}$		^{234}U (U.III) $2.33 \times 10^7 \text{ y}$				
Pa 91		^{234}Pa (UX ₂) α 1.14 m	89.85% β 11.15% α				
Th 90	^{234}Th (UX ₃) 24.5 d		^{229}Th (Io: ionium) $8.3 \times 10^4 \text{ y}$				
Ac 89			α				
Ra 88			^{226}Ra ("radium") 1622 y				
Fr 87			α				
Em 86			^{222}Rn (Rn: radon) 3.825 d				
At 85		α	^{218}At 1.3 s				
Po 84		α	^{218}Po (Ra A) 3.05 m	0.03% α	^{214}Po (Ra C) 150 μs	^{210}Po (Ra F) 140 d	
Bi 83		α 99.97%	^{214}Bi (Ra C) 19.7 m	99.95% α	^{210}Bi (Ra E) 5.0 d	β 100% α	
Pb 82		β	^{214}Pb (Ra B) 26.8 m	α .04%	^{210}Pb (Ra D) 22 γ	β $5 \times 10^{-6} \%$ α	^{208}Pb (Ra G) stable
Tl 81				^{210}Tl (Ra C) 13.2 m	β	^{208}Tl (Ra E) 4.23 m	β

FIGURE 1. The uranium radioactive series.

Deep, poorly ventilated mines are good examples of spaces where radon can concentrate and where miners working daily shifts can suffer devastating effects of a very specific kind. Radon can be inhaled and if decays occurs while the radon is in the lung, cells in the lung will be exposed to alpha radiation from the decay. However, because radon is a gas, it is blown back out again in the expired air and hence is not dangerous as a radionuclide that can be effectively retained. A more efficient way to hold dangerous material in the lung long enough to ensure that decay will occur happens when radon-222 decays in sequence to four short-lived, alpha and beta-emitting daughters; polonium-218, lead-214, bismuth-214, and subsequently to polonium-214. The radon daughters are charged solids that can attach to dust particles of the proper size to be inhaled and deposited in the upper passages of the lung. Unattached daughters also can interact with the cells of the air passages. The time the dust particles remain in the air passages is long(hours to days) compared to the half-lives of the radon daughters(minutes) so the cells lining the upper air passages are efficiently irradiated by alpha particles and after long exposures in the mines, lung cancers develop. *Most of the radiation dose to cells lining the upper air passages is received as a result of radon daughter decay.*

In Fig. 1 the uranium radioactive series is shown. This mechanism is extremely important to understanding the development of lung cancers in uranium miners. If there is a high concentration of radon and a large amount of dust, the conditions provide the highest risk for developing lung cancers. As mentioned earlier, the lung cancers resulting from radon exposure are rare except in miners where the high lung cancer incidence is recognized world-wide(6). Mines in North America, Europe, and Asia have caused increased lung cancer in miners.

For the general population, the source of radon is diffusion from soil. The diffusion of radon from soil is not a health hazard except when radon diffuses into a home from the soil beneath. In most homes, ventilation is sufficient to prevent radon from building to dangerous levels. However, heat conservation through insulation and sealing of cracks to ensure minimum heat loss from infiltration has the effect of concentrating radon and increasing radon and increasing the inhalation exposure. Recommendations for restricting radon exposure are conservative but steps taken to remove or substantially dilute radon accumulation in areas where people spend large amounts of time are probably worthwhile.

The lung cancer caused by radon and radon daughters appears to require lengthy exposures to fairly high concentrations of radon. A complicating factor is the observation that cigarette smoking sharply increases the number of miners who develop lung cancer(7). We know that cigarette smoking causes lung cancer. We also know that working in deep mines with inadequate ventilation causes lung cancer. However, miners who smoke are much more likely to develop lung cancer than miners who do not smoke. The number also exceeds the number of lung cancer victims who smoke but do not work in the mines. In other words, the number of cancer victims is larger than the number obtained by adding the smoking and the mining-induced cancers. The reason for the increase is not understood but clearly is

related to the susceptibility of normal cells lining the upper air passages to become cancerous.

REMEDIAL ACTION

The fact that cannot be overemphasized is the importance of understanding how the hazardous radionuclides move in the environment and the conditions that prevail when dangerous concentrations accumulate. In the present discussion, the most important fact to remember is that the only nuclide capable of moving without help is radon. If radon buildup is prevented, the radon decay daughters will not concentrate in a confined space, cannot enter the lung and cannot cause lung cancer. Thus, ventilation is the best method for ensuring the prevention of inhalation exposure to dangerous radon concentrations. Remediation of surface sources of radon such as tailings is also desirable but the return in safety for the amount of effort expended is not as high as the use of measures to dilute dangerous nuclides. Simple avoidance is not easy unless the extent of the hazard is known. The ability to obtain reliable estimates involves knowing how to obtain analytical services and advice. State health agencies provide these services and work closely with the Indian Health Service.

The other important feature in the effort to minimize radon exposure is to know or be able to measure radon concentrations indoors. Whatever the confined space, the air can be evaluated and protective steps can be taken. Finally, for a variety of reasons, it is probably wise to stop smoking cigarettes.

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EXPLANATORY NOTES FOR NON-SPECIALISTS

The natural background provides about 80 percent of our radiation dose from all sources. A principal contributor is uranium-238, the most common of the uranium isotopes in the earth's crust and the source of radon-222. By far the largest fraction of the natural radiation dose to man comes from radon-222 and its decay daughters polonium-218, lead-214, bismuth-214 and polonium-214. These radioactive atoms move freely in the environment for two reasons. Radon-222 is a gas that can diffuse and concentrate in confined spaces while its daughters, though solids, are charged and can attach to dust particles. The decay daughters move with the dust and deposit in the upper air passages of the lung. Both radon and its daughters can be inhaled as gas or as particles from the environment and held in the large air passages of the lungs long enough for decay to occur and alpha particles to strike the cells that line the airways. The energy is largely in the form of alpha particles. Although there are other mechanisms for getting radiation into the body from the environment such as swallowing radioactive material dissolved in drinking water or contaminated food, or through cuts, only breathing provides enough radioactivity to be dangerous. The most common danger is the development of lung cancer. In the world's population these cancers are rare. However, certain groups, all underground miners, have suffered a large number of these cancers and have been extensively studied. From the rarity of these cancers, except in underground miners, it is possible to conclude that the environmental conditions resulting in radon-induced cancers depend on significant concentration of radon in areas where people spend a third to half of every day. Wind and artificial ventilation both are capable of eliminating the surface hazard by dilution even over such rich radon sources as tailings piles. Cigarette smoking increases the risk of lung cancer in radon-exposed populations beyond the risks of smoking or radon exposure alone but the details affecting the increased risk are complex and poorly understood.

Bicarbonate Leaching of Uranium

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Abstract. The alkaline leach process for extracting uranium from uranium ores is reviewed. Uranium minerals dissolve in oxygen rich carbonate leach solutions. This extractive mechanism may be used in any type of mining process such as conventional, heap, and in-situ. Particular reference is made to the geochemical conditions at Crownpoint. Some supporting data from studies using alkaline leach for remediation of uranium-contaminated sites is presented.

INTRODUCTION

Just eight years after the planet Uranus was detected in 1781, a new element was discovered. This element was named uranium in honor of the planet. There was little commercial interest, except as a source for radium, until, 1939, when fission was discovered. Uranium increased rapidly in importance due to its value in nuclear weapons and nuclear reactors. Production peaked in 1962 reaching values of 17,000 tons per annum in the US. Although production has declined, uranium is still an important raw material for nuclear power.

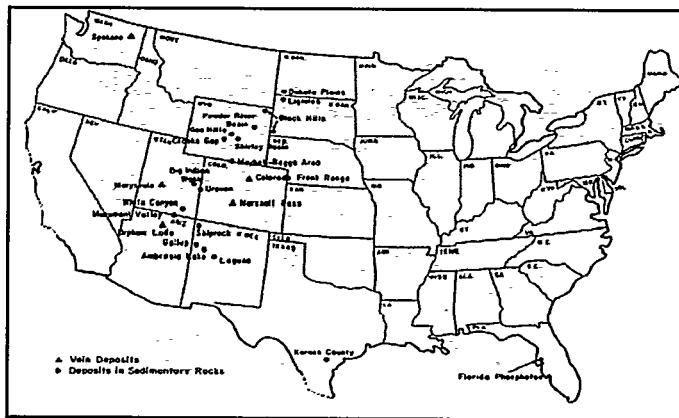


Figure 1. Location of uranium ore deposits in the U.S.

In the United States uranium ores are found in three major districts: the Wyoming basins, the Colorado Plateau and the Texas Gulf Coast as shown in Fig. 1. The Colorado Plateau extends from Colorado into Utah, Arizona and New Mexico.

Most uranium deposits are less than 800 ft deep, are horizontally bedded in sandstones, mudstones and limestones, the minerals occurring as impregnations and pore fillings in the host rocks.

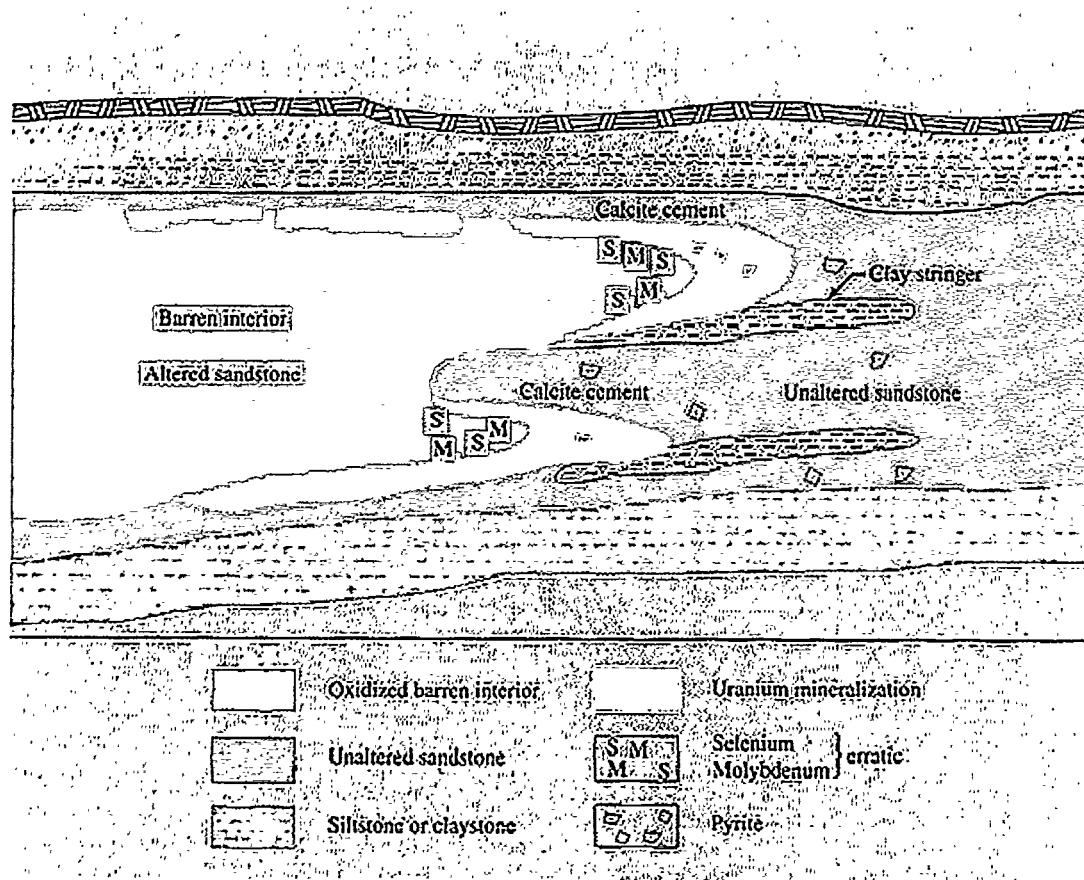


Figure 2. Schematic cross-section of an idealized roll-front orebody showing the zonation of elements and primary hydrologic and geochemical features. Oxidized groundwaters flow from left to right. The roll front and associated redox interface moves in the same direction.

Typically uranium ores are found in roll front formations as shown in Fig. 2.

Uranium is leached from its original formation by alkaline carbonate-rich ground water under oxidizing conditions. It remains in the groundwater until encountering reducing conditions where it is precipitated as insoluble uranite and coffinite. Such conditions exist at Crownpoint, NM.

MINING

In conventional mining uranium ore is brought to the surface and crushed. The uranium is extracted into solution using a leach reagent, followed by concentration,

often on ion exchange resins. Two variants on this process are the heap leach process and in-situ mining. These are shown schematically in Figs. 3. and 4.

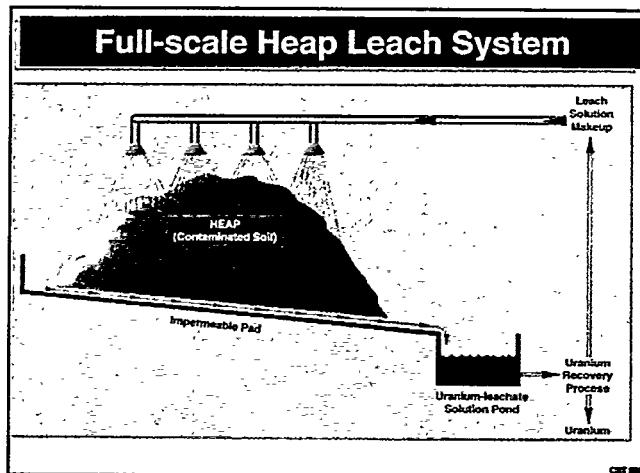


Figure 3. Schematic of heap leach process(3).

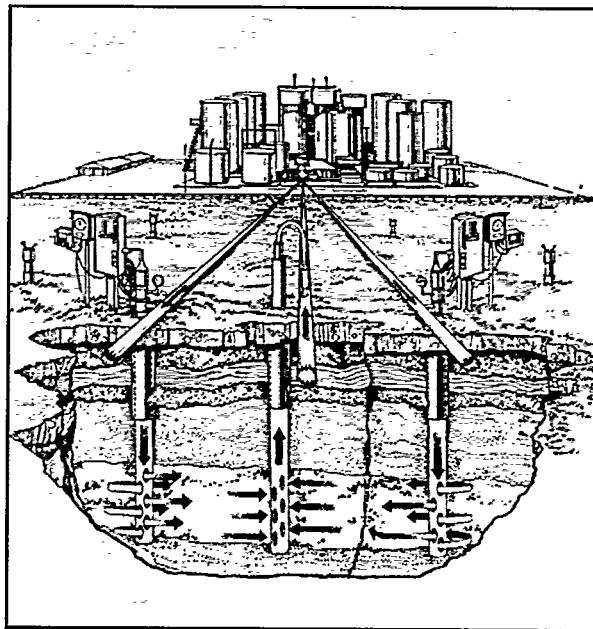


Figure 4. In-situ leach process(4).

Both methods have been used for uranium mining; the heap leach process has also been used for remediating uranium-contaminated sites. In the heap leach process, ore is brought to the surface, crushed and placed on an impermeable membrane. The leach reagent is sprinkled onto the heap, trickles through it, dissolving uranium as it passes. In in-situ mining, the leach reagent is injected through injection wells surrounding a pumping well. The reagent is pumped to the surface after it has

dissolved uranium. In both cases (heap and in-situ) the leached uranium solution is passed through an ion exchange resin which collects the uranium.

Both acid and alkali (bicarbonate) have been used as leach agents. Acid is often used but dissolves other metals as well as uranium. In regions with carbonate bearing rocks, such as limestone, acid is unsuitable and bicarbonate is used. At Crownpoint, concentrations of carbonate rocks are sufficiently plentiful that carbonate is present in the ground water making natural alkaline leach the leach of choice. In order to be dissolved uranium must be in the oxidized form.

CHEMISTRY OF EXTRACTION PROCESS

The mineral forms of uranium found at Crownpoint are listed in Table I. The major minerals are in the reduced state (+4) which accounts for their presence, since if in oxidized conditions the uranium would have been transported away through ground water movement. Those minerals that are oxidized (+6) (such as carnotite) owe their

Table 1. Mineral Forms of Uranium at Crownpoint, NM(4).

Name	Formula
Oxidation State +4	
Coffinite major phase	$U(SiO_4)_{1-x}(OH)_{4x}$
Uraninite minor	UO_2
Oxidation State +6 (all minor)	
Bayleyite	$Mg_2(UO_2)_3CO_3)_318H_2O$
Uranophane	$Ca(UO_2)_2(SiO_3)_2(OH)_25H_2O$
Tyuyamunite	$Ca(UO_2)_2(VO_4)_25-8H_2O$
Carnolite	$K_2(UO_2)_2(VO_4)_23H_2O$

presence to the insolubility of the specific salt. The effect of redox potential (Eh) and acidity (pH) is shown in Fig. 5.

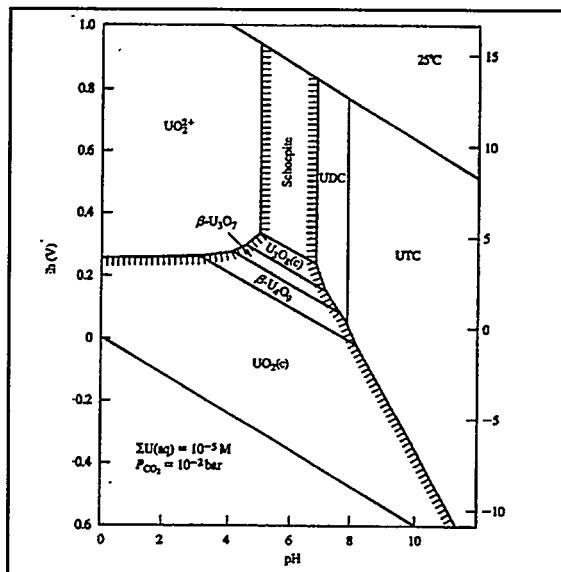
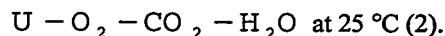
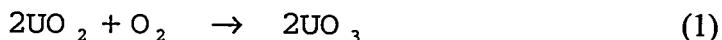


Figure 5. Eh-ph diagram for aqueous species and solids in the system

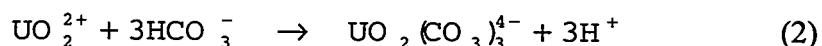


At low pH and low Eh the stable form is UO_2 . At low pH and oxidizing conditions (positive Eh), uranyl ions are the stable cations. At high pH, uranyl carbonate species are stable over a wide range of Eh values.

In order to leach the minerals, they first must all be in the oxidized state. Different oxidation agents have been used, including potassium permanganate and hydrogen peroxide. Dissolved oxygen itself is the agent of choice at Crownpoint. Regardless of the agent used, the defining chemistry is the same:



This is followed by the formation of soluble uranium carbonate species:



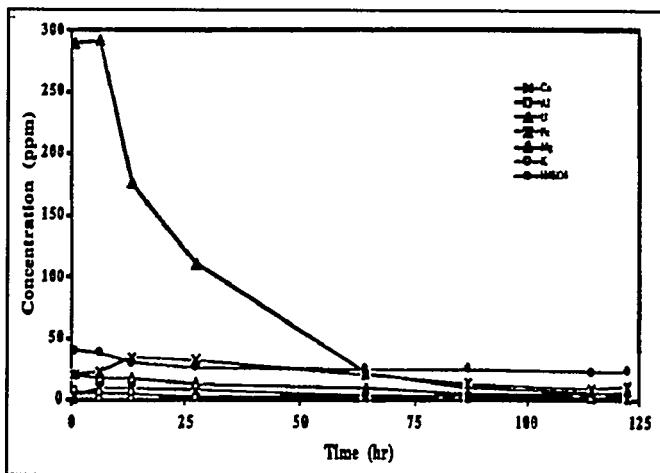


Figure 6. Concentration of metals in leach solution as a function of time(5).

No other commonly occurring metals form soluble complexes with carbonate. This is illustrated in Fig. 6. These results are taken from laboratory studies in support of environmental remediation activities at the Fernald site in Ohio. Although the details may change, the essential results are the same. Other redox systems may affect the solubility of different species.

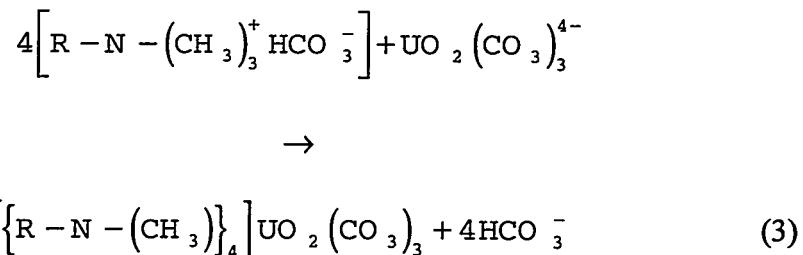
Table 2 shows the concentrations of chemical species present in the groundwater at Crownpoint. The high natural concentration of carbonate is sufficient to mobilize uranium. Clearly, for maximum removal of uranium, strict controls of pH and Eh must be maintained. The multiplicity of other ions in the groundwater make the possibility of subsidiary reactions likely.

TABLE 2. Concentrations of major ions in groundwater at Crownpoint, NM(4).

Chemical Species	Concentration(mg/L)
Calcium	100-350
Magnesium	10-50
Sodium	500-1600
Potassium	25-250
Carbonate	0-500
Bicarbonate	800-1500
Sulfate	100-1200
Chloride	50-1800
Silica	25-50
Uranium	50-250

CONCENTRATION OF SOLUBLE URANIUM SPECIES ON ION EXCHANGE RESINS

Ion exchange resins are insoluble polymer beads with easily exchangeable ions. The following reaction illustrates their use for removing uranium from solution:



Once the uranium is immobilized it can be easily stripped off the resin and concentrated using the reverse process and further converted to yellow cake, U_3O_8 , for processing. Fig. 7 shows comparison of two resins used for uranyl uptake in laboratory studies in support of an environmental remediation site.

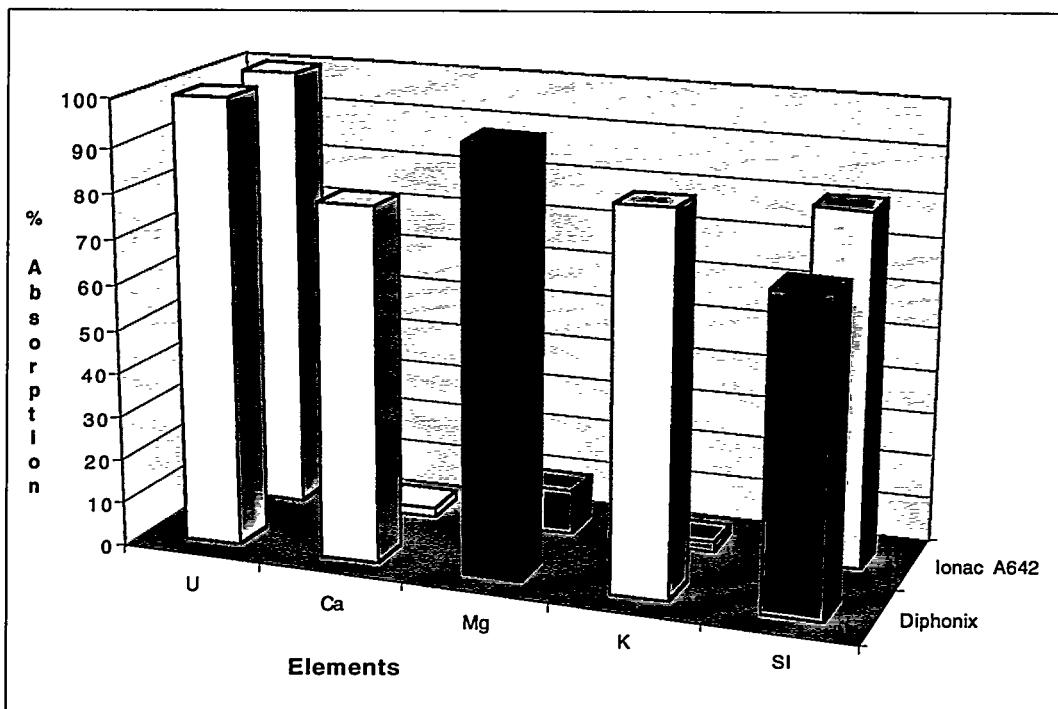


FIGURE 7. Comparison of two resins Diphonix and Ionac A641(3).

The first, Ionac A641, is a simple anion exchange resin and shows how the anion, silicate (SiO_4^{4-}), competes successfully for exchange sites. The second, Diphonix, is both an anion and cation exchanger resulting in uptake of calcium, magnesium and potassium. Since the ground water at Crownpoint has multiple ions, there will be competition for exchange sites on the resin. Fig. 8 shows the ion exchange columns used at a field remediation site for uranium contamination at Los Alamos where we successfully removed 99% of the uranyl ions from solution.

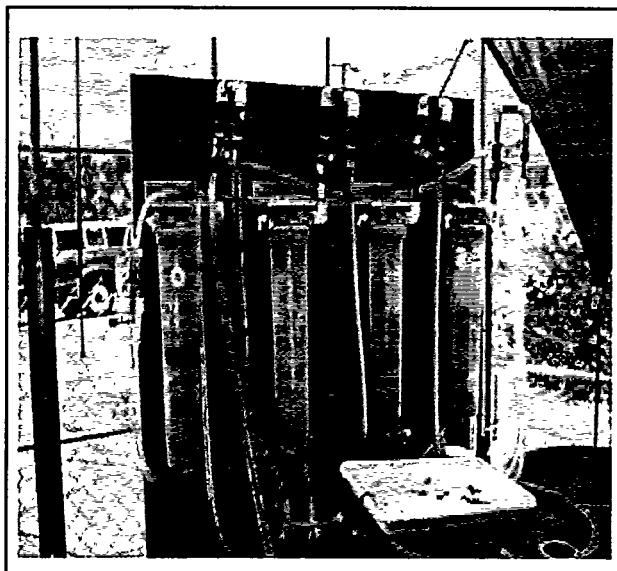


FIGURE 8. Ion exchange resin columns at remediation site for uranium-contaminated soil(3).

SUMMARY

Bicarbonate leach systems may be used to dissolve uranium regardless of the physical system involved. Each mining (or environmental) site is defined by its own local geology and geochemistry and must be considered on its own merits. Some chemical information needs further study, such as the mechanism of oxidation and the partitioning of the radiochemical daughters of uranium, including thorium and radium. Knowledge of these variables will help optimize the leach process.

ACKNOWLEDGMENTS

This summary is based on results from several uranium-contaminated sites as well as data from the Crownpoint mining project. Many scientists have contributed including Fred Begay, John Kaszuba, Ningping Lu, Bryan Travis, Ines Triay, William Turney, Don York, Bruce Thomson and Frank Lichnovsky.

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Computational Methods for Subsurface Flow and Transport

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Abstract. The design of in situ uranium leaching and prediction of its affect on local water quality rely heavily on computer simulation models of flow and transport through porous soil and rock. In order to evaluate the worth of computer models, it is important to understand the modeling process, what models are and what they can do, and what new features may be needed to improve them. In this paper, a brief description of models and some examples are given. In addition, recently identified transport mechanisms and processes are discussed. These include: colloids, which can rapidly transport low levels of sorbed chemicals; microbes, which can affect the rate of mineral dissolution; chemical reactions, which control the efficiency of in situ leaching; and spatially correlated soil heterogeneity, which can lead to channeling and loss of flow control. Incorporation of these features into computational flow and transport models will enhance the ability to predict the progress of an in situ leaching operation and its impact on water quality.

KINDS OF MODELS

Many experiments over the past century have established that water, air, oil and various chemical species move through porous media, such as soils and rocks, according to well-defined "laws" and principles. This experimental base of knowledge has led to the development of hydrological models. Hydrological models (and all models for that matter) fall into several categories. There is always a conceptual hydrological model, whether explicitly stated or only implicitly understood, when hydrologists work on a site. The conceptual hydrological model is simply the mental picture that the hydrologist has of how the various subsurface geological units are arranged, how water flows through the system, and how contaminants are transported. It may be correct or it may not be, but it is what the hydrologist thinks is happening, based on a variety of kinds of data from the site, as well as the hydrologist's experience. It is frequently useful to build physical models, that is, small representations built to scale, in which the field operations can be mimicked, in

miniature. These are usually of fish tank size, with glass or plastic sides so that the progress of flow and transport can be seen and recorded. Physical models are very useful for educational purposes, as well. Another very useful type of model is the mathematical model, in which the conceptual model is translated into mathematical equations that describe the assumptions, processes and field operations. If the equations of the model can be solved, then the outcome of field operations can be predicted at every stage. In every model, some simplifications are made. It is the hope of the modeler that the simplifications will not seriously affect the usefulness of the model and the accuracy of its predictions. That is why model developers must work closely with geologists, hydrologists, chemists and microbiologists to ensure that the model is as realistic as necessary. Often, the equations of the mathematical model cannot be solved directly. In this case, numerical solutions, which are implemented on computers, are substituted. These are frequently called computational or simulation models. Numerical solutions, if done properly, will very closely approximate the solution of the original mathematical model equations. Finally, the model predictions can be visualized through the use of modern computer graphics.

Computational Models

Computational models provide a mechanism for collecting what we know about a site and the processes involved into a single dynamically consistent system, whose interactions are constrained by fundamental laws of physics and chemistry. Models can be used in various ways: (a) to provide sensitivities of a process to the various parameters, data, forces and constraints; (b) to aid in interpretation of laboratory experiments, field tests and field data; and (c) for design and forecasting of in situ operations and post-closure remediation. A calibrated model can provide estimates of the temporal and spatial distribution of concentrations, pressures and water saturations and water velocities everywhere in a subsurface region.

Models of flow and transport are easy to understand in principle, even if the mathematics appears daunting. Mathematical models are based first on conservation laws; conservation of mass, energy and momentum are fundamental. These can be stated in simple English. First, imagine a box, or better yet, a box-shaped, fine wire-mesh cage, so that the sides are "leaky". Then the conservation laws can be stated simply: the total amount of energy presently inside the box cage equals the amount of energy present at some past time plus the energy that crossed the sides of the cage between the previous time and the present, plus the energy gained or lost within the box cage due to chemical reactions. Likewise, the total amount of mass or matter inside the box cage equals what was there at some past time plus the amount that passed through the sides of the cage between that previous time and the present. Even the most sophisticated subsurface flow and transport models are equivalent to these conservation statements. In addition to these conservation laws, some additional

relationships are needed, the most important being the equation of state. Once again, although the mathematical formulation may look impressive, the underlying idea is simple. An equation of state simply indicates how a fluid or a solid responds under different conditions. To be more specific, an equation of state for water, for example, simply tells us what the density of water is at different pressures and temperatures. These additional relationships are often called constitutive relationships, or constitutive equations. One more step is needed to make the connection between the wire-mesh box cage example above and a real site, such as an oil field, or a uranium ore deposit. Simply think of the field as being divided into thousands or even millions of little box cages. Then the conservation laws apply to each box, and what goes out the side of one box is what enters the side of the neighboring box. The numerical/computational models do exactly this – divide the volume of interest (the oil field or uranium deposit) into many small box cages, and keep track of how much mass and energy pass from one box to the next, and use the equation of state to determine the pressure and temperature in each box. Then the flow rates between boxes can be computed.

Examples

In the spirit of modern computer and communications technology, the reader is referred to the internet address: <http://ees-www.lanl.gov/EES5/capa.html> to see a wealth of examples of porous flow and transport models. Descriptions, plus color images showing results of simulations can be found there. These represent most of the capabilities that have been developed at Los Alamos National Laboratory, with a variety of applications. These capabilities are representative of the state of the art in subsurface modeling. It will be apparent, after visiting the website, that models come in a great variety of packages, depending on the intended use. In some cases, the target application is the understanding of small scale processes, that is, a highly detailed simulation of what could occur within the pores of a small sample of soil or rock. Experimentally, using x-ray tomography and mri (magnetic resonance imaging), the pore structure of samples of rock can be mapped to a resolution of a few microns. This enormous amount of information is digitized, providing an electronic picture of the interior structure of a rock sample (e.g., an oil-bearing sandstone). This digitized image is read directly into our Lattice Boltzmann computer code, which can then predict how water and oil and solutes will move through the pores of the rock. This kind of simulation is extremely useful because it allows the user to create constitutive equations that can be used in larger-scale simulations (e.g., on the scale of an oil field). Another example describes simulation of acid mine drainage from a uranium mill tailings site in Canada. This involves strong chemical reactions coupled with vertical water movement. Model results are compared to experimental measurements and field observations, to see if they are accurate. When model results compare closely to

observed behavior, we have confidence that the model includes the important features for a site, and we can then use the model for additional site-related studies. The reader will also find at the website given above, examples of field-scale simulation studies, such as Yucca Mountain, Nevada. Radioactive waste may be stored within Yucca Mountain in the near future. Numerical models are being applied to understand how radionuclides might move through the fractured rock layers found there. This requires our most advanced modeling capability, because the geology is three-dimensional and complex, fractures are present, and chemical and even biological reactions will occur along the flowpaths over long periods of time.

RECENT DEVELOPMENTS REGARDING TRANSPORT PROCESSES

Traditionally, transport models in soil and rock have considered only these mechanisms for moving contaminants: molecular diffusion, dispersion (a diffusion-like process caused by the pore structure of soils and rock) and advection (that is, movement with the groundwater). Some mechanisms that retard transport, such as chemical reaction between water-borne contaminants and the soil grains, are also included, but have been simple in concept, not always providing a good match to observations. Recently, as a result of much experimental work, a number of new transport mechanisms have been recognized as potentially very important. Model developers are enhancing their existing mathematical/computational models to include these processes. These mechanisms are described briefly below.

Colloids

Many studies suggest that colloids (particulate matter less than 10 microns in size) are found in all groundwaters. Natural colloids can consist of a variety of substances, including clay particles, iron oxyhydroxides, and even bacteria (1). Theoretical studies indicate that when solutes such as U(VI) sorb to colloidal material (and they do readily), their transport behavior can differ markedly from that obtained with traditional advective-diffuse-dispersive modeling (2). Field studies indicate that smaller-sized colloids are transported kilometers or more through groundwater flow. This transport mechanism may alter conclusions regarding local water quality. Recent experiments at LANL indicate that the traditional filtration model of colloidal movement is inadequate; rather than being permanently captured, small to mid-size colloids will attach and detach from soil grains with a distribution of residence times. At Los Alamos, we are incorporating colloid transport with residence time "filtration" into our field-scale flow and transport models and testing against data from experiments.

Microbes

Microbes are found in virtually every environment on the earth, from the arctic ice to boiling springs. Microbes have evolved to live on a variety of energy sources. They are not just passive guests living in soil; they are responsible for many chemical and mineralogical changes that occur in the subsurface (3). In the past decade, many laboratory and field experiments have demonstrated that microbes can be used to help remediate sites that have been contaminated with chemicals containing carbon, which includes a vast number of compounds and most of the common organic contaminants (4). Frequently, the action of microbes can be greatly enhanced by injecting other benign chemicals, such as oxygen and methane, that will stimulate growth of bacteria (5). Further recent data shows that under the right conditions, microbes can even be used to attack toxic metals in groundwater, such as arsenic, cadmium and other heavy metals (6). In this case, the metals are not destroyed, but are either made immobile through precipitation reactions, or mobilized through dissolution reactions, depending on the chemical state of the pore water. Mathematical models are available now which approximate the action of microbes on subsurface contaminants and can predict reasonably well the rate at which microbes will degrade contaminants under differing conditions (7).

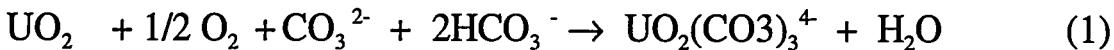
Microbial species are also known to have a significant effect on uranium leaching reactions, acting as a catalyst, under certain conditions, to accelerate the oxidation/ reduction reactions (8). Bicarbonate leaching stimulates classes of microbes. Their metabolic activity can be critical in controlling the dissolution or precipitation of uraninite. The kinetics of these microbial species have been determined by different research groups. Scientists at the University of New Mexico's Center for Radioactive Waste Management (CeRaM), e.g., are an in-state research group that has determined relevant data on microbial activity at uranium sites. Microbes are sometimes mobile, and can therefore also act as a class of colloidal material, carrying contaminants along with them that stick to the microbe cell wall.

Chemical Reactions

As water flows through rock, a great variety of chemical reactions can occur between chemical constituents in the water and the host rock (9). In some cases, these reactions are slow and mineral changes occur over long periods of time. In other cases, mineralogical changes can be rapid. Chemical reactions can lead to changes in the pore structure of the host rock, causing changes in porosity and permeability, which lead to changes in the groundwater flow field. In situ leaching of uranium is an example of a situation in which chemicals are injected into a groundwater system to deliberately speed up chemical reactions that will liberate soluble forms of uranium. It is critical that computer models be able to capture the principle chemical reactions and the

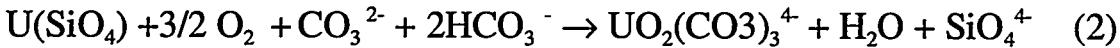
changes they produce in the flow field. This is a major focus of research today in subsurface water flow and contaminant transport.

Most naturally occurring uranium minerals are in a reduced chemical state. Under reducing conditions (low pH and no oxygen), a common form of uranium mineral is uraninite (UO_2). In situ leaching targets uraninite; the goal is to oxidize it to a water- soluble form. Oxygenated water passing over deposits of uraninite will cause uranyl carbonate to form, which is soluble. This then moves with the groundwater flow to withdrawal pumps where it can be brought to the surface and separated from the water. Previously, acid was injected into subsurface deposits to release uranium. However, this was expensive, and caused considerable changes in the rock matrix structure, leading to permeability changes and loss of control of the groundwater flow. Most recently, as in the HydroResources Inc., leaching procedure, a carbonate/bicarbonate solution is added to the groundwater passing through uranium ore deposits. This is significantly less expensive, and produces chemical reactions that are much less destructive to the host rock, and results in only minor impact on water quality, at most. This bicarbonate treatment can be summarized in the following chemical balance equation:



Another important uranium ore is coffinite. This form is a major mineral, e.g., at Crownpoint, New Mexico. It is a silica bearing mineral. It can be solubilized in a manner

very similar to that for uraninite. The chemical balance equation for this reaction is:



These chemical balance equations can be coupled to flow and transport models, providing a prediction of how much of and how fast the uranium ores will be oxidized and what pumping schedule will give the optimal recovery of uranium. This requires that models track each of the chemical species present in the stoichiometric equations above.

Heterogeneity

Probably the most distinctive feature of the subsurface environment is that it is very heterogeneous; soil and rock properties vary over a wide range of length scales. Even soils that are usually considered uniform, such as sand, will exhibit some variation in properties that are important for flow of water and transport of contaminants. Frequently, computer models ignore this heterogeneity, using an average value of some important property such as permeability. In some situations,

this is acceptable; however, in other situations, it can lead to serious errors in predicting where contaminants will go. A weakness in subsurface flow and transport simulation models is that they do not properly account for the variability seen in soil properties, especially at scales smaller than the computational grid size.

Computer simulations are used extensively now to estimate a variety of subsurface processes involving fluid flow, such as oil production, aquifer management, in situ leaching of minerals, and contaminant remediation. These all depend critically on the accuracy of subsurface flow models. However, the rate and direction of subsurface flow is governed by heterogeneities in geologic formations over many scales, from gross structural features to fine-scale details. Because of the limitations of our computers, we are forced to solve the governing equations on a coarse scale, missing the details that are finer than the size of computational mesh cells. Unfortunately, it has been shown that ignoring the subgridscale structure can (but not always) lead to serious errors in estimation of flow direction and speed.

Recent developments in modeling techniques attempt to incorporate fine-scale detail into a coarser-scale representation of subsurface flow by scaling up soil variability data and homogenizing the governing partial differential equations. Various attempts to capture subgridscale dynamics, e.g., homogenization, two-timing (two-scale perturbation analysis), renormalization group theory, fractal analysis, and stochastic models, show great promise (10).

Scale analysis in hydrology has focused on stochastic methods. More recently, that is narrowing to fractals. Field data from many sites world-wide show a fractal character to soil heterogeneity (11). (Fractals exhibit power law scaling, have structure at all scales, and are self-affine at all scales. Soils do not exhibit self-affine structure over all scales, but over a large range – many decades.) Recently, DiFederico and Neuman (12) have proposed a new model which accounts for this multiscale property. An important feature of their analysis is that the fractal structure is truncated, i.e., the range of scales is limited, in accord with observations. Their algorithm is simple in principle and will be relatively easy to implement. The first large scale application of their model for porous flow and reactive transport will occur very soon.

Elegant mathematical methods exist for creating structures having fractal character (13). We can represent fractal objects in very compact ways, e.g., using iterated function systems or fractal interpolating functions. The use of fractal functions in dynamics (i.e., differential or integral equations) is very recent. We can integrate fractal functions directly and exactly, e.g., to get average properties over some spatial domain. The attractive feature of this approach is that integrals of fractal functions can be related in a feasible manner to the corresponding classical linear interpolation functions. This is very useful, for example, for capturing subgridscale variability in a finite difference numerical simulation. Further there are analytic solutions available for "simple" problems, such as diffusion on canonical fractals (e.g., on Sierpinski triangles). These are convenient for testing numerical solutions. Merging of existing fractal

mathematics with the stochastic analysis of porous media will result in much more powerful modeling capability and is an exciting area of porous flow and transport research.

SUMMARY

We can use our computer models to address two principal questions related to the Crownpoint, NM and Shiprock, NM in situ leaching operations: (a) Will the operation be self-contained, as advertised, or will it leak into the nearby drinking water supply? Although the in situ leaching process appears to be well-controlled, a recent NRC study raised some questions. In situ operations are designed on the assumption that the soil/rock formations are homogeneous in nature and that the process zone is isolated by impermeable material above and below. However, the over/underlying units are not perfect barriers and colloid-bearing fluid may leak through. Further, no soil or rock formation is truly homogeneous in its properties. Spatial correlation in soil heterogeneity can lead to formation of permeability channels which may defeat the designed controls, allowing some process chemicals to migrate outside. We can estimate the likelihood of this happening. Further, the impact of microbial activity, which may be beneficial, has not been accounted for. (b) Can the leaching operation be improved? Optimization of the in situ leaching operation can be attempted by coupling our reactive transport model with optimization algorithms (14,15). Novel aspects of this optimization approach are inclusion of microbial reactions, and optimization in the presence of heterogeneity/uncertainty. Addition of microbial stimulants, such as lactate or acetate, to the in situ leaching injected fluids, could significantly accelerate the oxidation rates of U(IV), although excessive growth of bacteria would be counterproductive. These ideas can be tested through computer simulation, in collaboration with experimentalists and field tests.

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