RADIOACTIVE CONDITIONING
OF
DOMESTIC WATER SUPPLIES

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1960-61

CERTIFICATE

CERTIFIED that the dissertation entitled "Radioactive Conditioning of Domestic Water Supplies" which is being submitted by Sri A.R. Kumbhare in partial fulfilment for the award of the Degree of Master of Engineering in Public Health Engineering of University of Roorkee is a record of student's own work carried out by him under my supervision and guidance. The matter embodied in this dissertation has not been submitted for the award of any other Degree or Diploma.

This is further to certify that he has worked for a period of four and a half months from April 15th to August 31st 1961 for preparing dissertation for Master of Engineering Degree at the University.

Signature M Chandra

Designation of the Supervisor Reader in

Seal ......

Dated 28.8.61

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Arun Kumbhare

(ARUN KUMBHARE)

SYNOPSIS

This dissertation deals with radioactive conditioning of domestic water supplies. It is believed that in order that an engineer may be able to condition water radioactively, he must know at least four things. Firstly, he must know what is radioactivity and nature of radiations, secondly the grave health hazards caused by the presence of excess of radioactivity in drinking water, thirdly the instruments, appliances and techniques employed in detection and measurement of radioactivity, and lastly methods of removing radioactive contaminants from water by various water treatment processes.

An effort has been made to emphasise and discuss all these aspects, specially the last one. The data and results included are not claimed as the Author's own findings but are a systematic collection and compilation from various sources.

CONTENTS

INTRODUCTION

The topic of "Radioactive Conditioning of Domestic Water Supplies" has abruptly gained importance during the last few years. Owing to a number of nuclear tests and extensive use of radioactive isotopes in various fields such as scientific, industrial, research and medical etc., the sources of water supplies have recently been contaminated with radioactive materials. Extensive research has been undertaken in various countries to explore the harmful effect of radioactivity and soon it was established that for safety waters used for drinking purposes should contain the least possible amounts of radioactive elements. Consequently, engineers investigated methods for decontaminating waters which have been polluted by radioactive substances. Substantial work has been done at various research laboratories and an effort has been made to present the useful information obtained as a result of these investigations.

The treatment of water for radioactivity has already been started in various countries and it has earned the same importance and status as other conventional water treatment processes. The author feels the necessity of monitering radioactivity at least in waters that constitute the important sources of supply in India - for example Bombay and Tarapore where we propose to set up our nuclear power station, and suitable treatment processes adopted to condition water radioactively. This happens to be the source of inspiration for undertaking this study. An attempt has, therefore, been made to provide all that an engineer incharge of water-works requires in the form of background for the subject. It is pertinent to point out that this study is based on uptodate thinking and recent trends in radioactive conditioning of water and it is possible that they may be modified, ammended or radically changed by further investigation and lapse of time.

CHAPTER I

Elementary Concept of Radioactivity

1. Introduction
2. Historical
3. Atomic Structure
4. Nuclear Theory
5. Electron Orbits
6. Neutron-Proton Concept of Nuclear Structures
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9. Nature of Radiations- Alpha Rays; Beta Rays; Gamma Rays.
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12. Atomic Transmutations and Artificial Radioactivity.
13. Units of Radioactivity.

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ELEMENTARY CONCEPT OF RADIOACTIVITY

1. Introduction

Before going into the details of the detection and removal of radioactivity from drinking water and other allied problems it is important to know what exactly radioactivity is. In order to understand radioactivity one must have the basic knowledge of the structure of atom. There is a very close relation between the structure of atom and radioactivity. Here, therefore, an attempt has been made to include something about the structure of atom and laws governing the structural changes in atoms in a brief and comprehensive manner. Units of radioactivity and certain definitions which would be useful throughout have also been explained. It should be appreciated that radioactivity as such falls under the domain of Physics or Radio-Chemistry and much work has been done on this topic. We have to view the topic through an engineers eye. With this in mind enough background is being provided in this chapter to permit understanding of the terms, units, formulae and other statements related to radioactivity.

2. Historical

The science of radioactivity deals with atomic transformations and may be considered to date from 1895 when Roentgen discovered a new form of radiation from cathode-ray tubes. The rays caused certain salts to become luminescent and also affected photographic plates. These are called roentgen rays or X-rays.

In 1898, Pierre and Marie Curie concluded that the X-rays from uranium were an atomic phenomenon characteristic of the elements, and they introduced the name radioactivity. The Curies pursued their studies of radioactive materials with great vigour. They found that compounds of thorium emitted rays similar to those of uranium. They also noted that certain ores of uranium were more radioactive than uranium itself.(1) This led to a search for other materials in the residues remaining after uranium extraction. Two new radioactive elements were isolated, polonium and radium. Radium is several thousand times more radioactive than uranium.

3. Atomic Structure

Modern concepts of atomic structure are largely the result of knowledge gained from the behaviour of radioactive materials. It is difficult therefore to discuss one without considering the other. Prior to the discovery of radioactivity, atoms were considered to be indivisible.

With the discovery that radioactive materials emitted positively and negatively charged particles, the foundation was laid for a new concept.(2)

4. Nuclear Theory

By 1900 it was realised that atoms are not indivisible. However, it was not until 1911 that Rutherford proposed the nuclear concept of the atom. This theory held that atoms were composed of a small positively charged nucleus, containing most of the mass of the atom, with a cloud of negatively charged electrons surrounding it. (2)

5. Electron Orbits

Bohr was the first to propose that the electrons about the nucleus of an atom are arranged in a methodical manner and revolve in orbits about the nucleus. (3) Although, his theory propounded in 1913, has undergone some refinements, it remains the basis of our modern day knowledge. The present tendency is to think of the electrons as being arranged in shells about the nucleus. (4) A major contribution was made to the Bohr theory by Sommerfield who has shown that the electrons within a given shell occur in several energy levels. (1) Other contributions particularly with respect to chemical properties, were made by Langmuir (Octet theory), Mosely, G.N. Lewis and W.Kossel.

The simplest atoms, hydrogen and helium, have one shell of electrons and the most complex have seven. The

TABLE 1

ARRANGEMENT OF ELECTRONS FOR SOME COMMON ELEMENTS

SYMBOL	ATOMIC NUMBER	NO. OF ELECTRONS IN SHELLS						
		K	L	M	N	O	P	Q
H	1	1						
He	2	2						
N	7	2	5					
Ne	10	2	8					
Na	11	2	8	1				
Cl	17	2	8	7				
Ar	18	2	8	8				
Ca	20	2	8	8	2			
[illegible]	31	2	8	18	2			
Cu	29	2	8	18	1			
Kr	36	2	8	18	8			
Ag	47	2	8	18	18	1		
Xe	54	2	8	18	18	8		
Ba	56	2	8	18	18	8	2	
Hg	80	2	8	18	32	18	2	
Pb	82	2	8	18	32	18	4	
Rn	86	2	8	18	32	18	8	
Ra	88	2	8	18	32	18	8	2

shells or rings are designated as K, L, M, N, O, P and Q in the order of their increasing remoteness from the nucleus. The arrangements of electrons for a number of elements is given in Table 1. Over the course of ensuing years, the positively charged nucleus was considered to consist of protons and electrons, except in the case of hydrogen atoms ($_1H^1$). The protons were always in excess of the electrons in the nucleus, and this excess was equal to the planetary electrons; thus the net charge on an atom was zero.

6. Neutron - Proton Concept of Nuclear Structure

In 1930, Bothe and Becker discovered a very penetrating secondary radiation when light elements such as beryllium and lithium, were subjected to bombardment by alpha particles from polonium. (2) The new rays were first thought to be X-rays of very short wavelength. In 1932, Chadwick showed this secondary radiation to be made up of neutral particles having a mass comparable to that of proton. The new particles were given the name neutrons, and since their source was obviously the nucleus of the bombarded atoms, a new concept of nuclear structure evolved.

According to present day knowledge, the nucleus of all atoms, except the simple hydrogen atoms, consists of both neutrons and protons. The number of protons corresponds to the atomic number and is equal to the number of electrons about the nucleus. (3) The number of neutrons

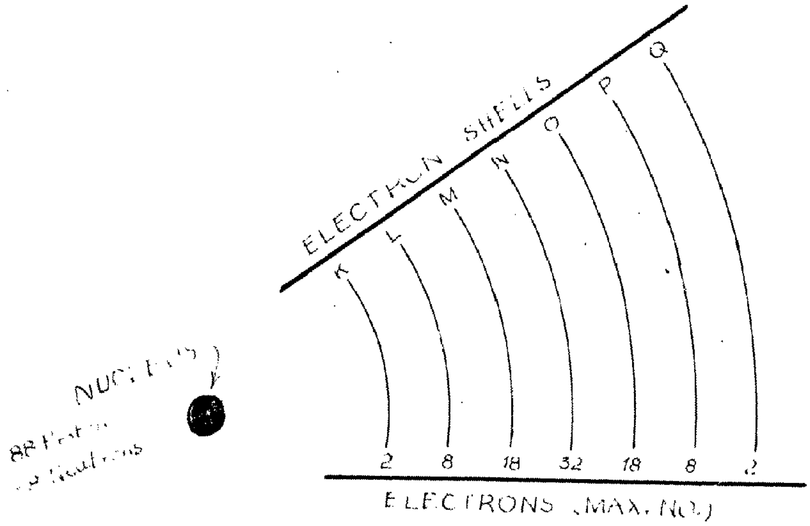

FIG 1

STRUCTURE OF RADIUM ATOM ($_{88}Ra^{226}$).

is equal to the atomic weight (expressed as the nearest whole number) less the number of protons. The structure of the atom may be represented as shown in Figure 1. The nucleus has the diameter of the order of 10^{-12} to 10^{-13} cm and the atom a diameter of about 10^{-8} cm. The density of nuclear matter is tremendous. It is estimated that one cubic centimeter would weigh 10^{8} tons.(5)

7. Nomenculture of Isotopes

All isotopes of the same element have the same number of electrons, and of course, the same number of protons. Since the masses of the isotopes vary, the number of neutrous must vary. In order to differentiate between isotopes a new system of symbol writing had to be developed. The system is vouge in the United States includes the atomic number as a subscript just before the symbol and the atomic weight, or mass number as a subscript after the symbol. For example, $_{82}Pb^{204}$, $_{82}Pb^{206}$, $_{82}Pb^{207}$, and $_{82}Pb^{208}$ represent ($_{82}Pb^{204}$) four isotopic forms of lead all of which have 82 protons and 82 electrons. Since the atomic number of a given element is always the same, it is frequently eliminated when discussing isotopes.

However, in radioactive changes involving transmutation of one element into another, such as in the conversion of U^{238} to Pb^{206}, the change is best shown by $_{92}U^{238}$ $_{82}Pb^{206}$. Any one familiar with radiochemistry

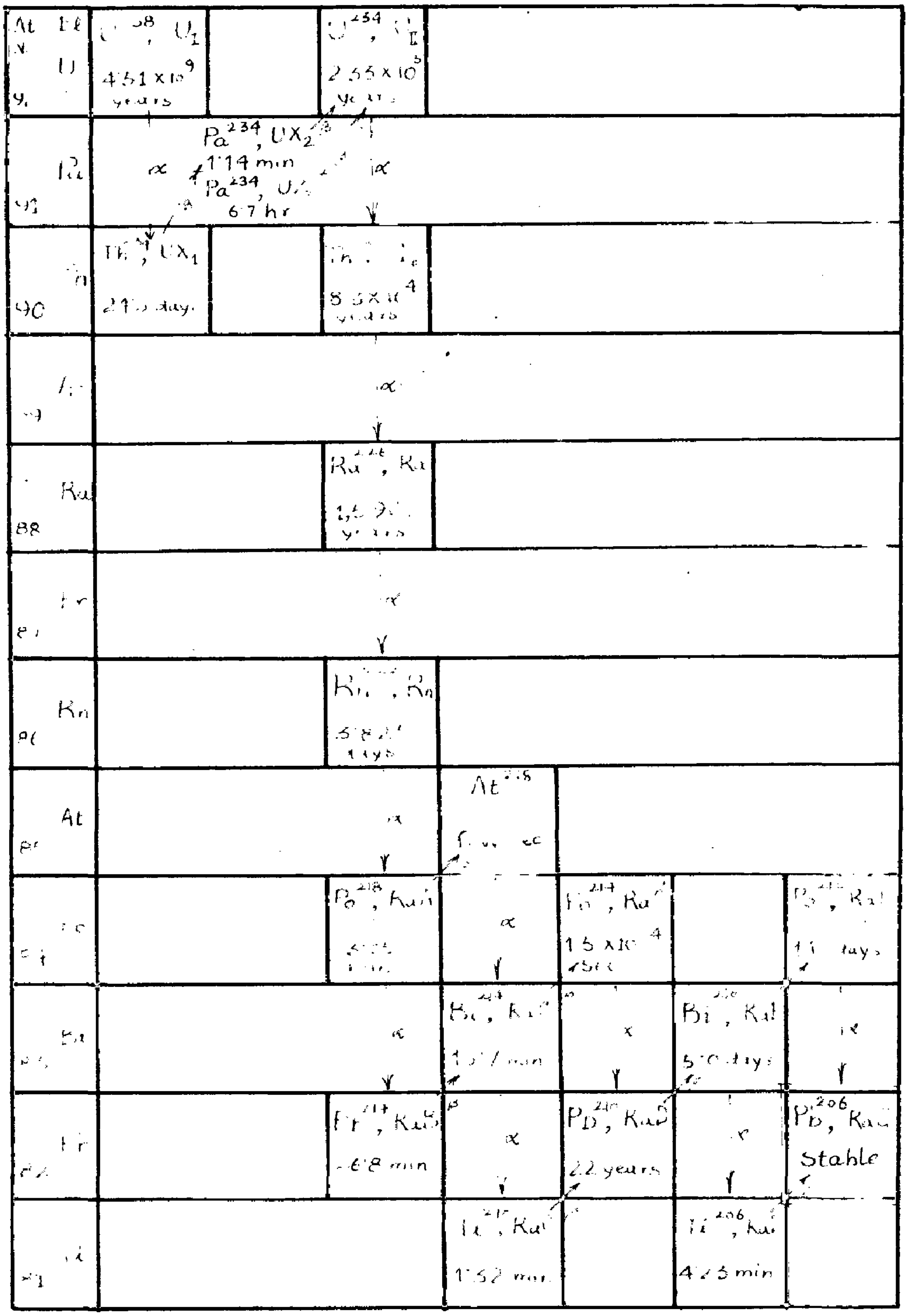

FIG. 2

STEPS IN RADIOACTIVE DECAY OF U^{238} TO STABLE Pb^{206}.

knows that such a change cannot occur in one step and that several intermediate steps are involved. See Figure 2.

8. Natural Radioactivity

Over 40 kinds of atoms are known that display the property of natural radioactivity. Most of these have atomic weights greater than 200.(4) The heavy-metal radioactive elements fall into three series: uranium, thorium and actinium. The uranium series has U^{238} as its parent substance and after fourteen successive transformations have occurred, the end product is Pb^{206}. Thorium (Th^{232}) is the parent substance of the thorium series. After ten transformations it remains as Pb^{208}. The parent element of the actinium series is U^{235} and after eleven transformations, it remains as Pb^{207}. This series takes its name from the fact that Pa^{231}(preactinium) and Ac^{227} (actinium) are long lived elements formed as steps in the transformation. The uranium series is sometimes called the radium series for the same reason. The steps in the radioactive decay are shown in Fig. 2. Th^{232} and U^{235} decay through similar steps.

9. Nature of Radiations

Early workers with radioactive materials were cognizant of the presence of only one form of radiation, and its properties were similar to those of X-rays. Later investigations established the presence of three kinds of rays designated as alpha, beta and gamma rays. Separation and identification were accomplished by directing the rays through a magnetic field, as shown in Fig.3. Under the influence of magnetic field certain rays were bent slightly toward the negative pole. This phenomenon indicated that they had a positive charge and, probably, were of considerable mass. These are called alpha rays. Other rays were bent radically toward the positive pole, showing them to be negatively charged, and,probably of smaller mass. These are called beta rays. A third group of rays were unaffected by the magnetic field indicating an absence of charge and are called gamma rays.

Alpha Rays

Alpha rays are not true rays as are light and X-rays. They consist of particles of matter and, therefore, should be referred to as alpha particles (2). Alpha particles are actually doubly charged ions of helium with a mass of 4. (3) Although these are propelled from the nucleus of atom at velocities ranging from 1.4 to 2 x 10^9 cm/sec.(about 10 per cent of the velocity of light), they do not travel much more than a few cm. in air at room temperature. They may be stopped by an ordinary sheet of paper. The alpha particles emitted by a particular element are all released at the same velocity. The velocity may vary, however, from element to element. The alpha particles have extremely high ionizing action within their range (2).

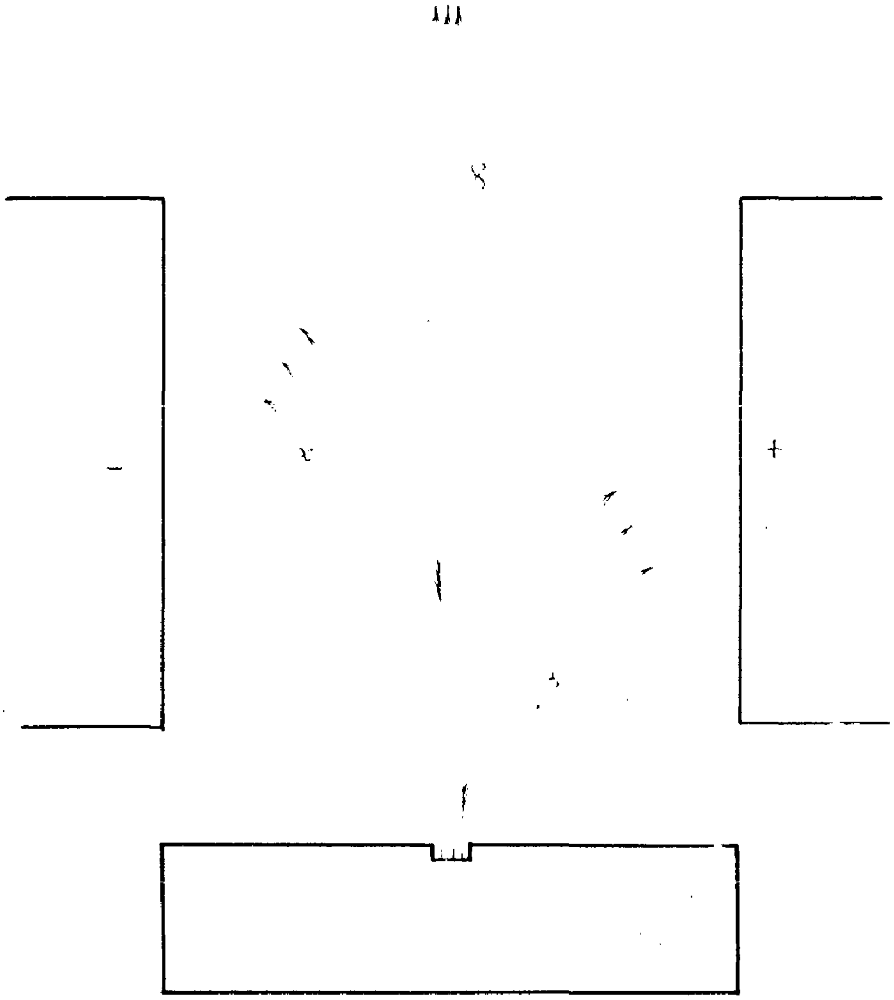

FIG. 5.

EFFECT OF MAGNETIC FIELD UPON ALPHA, BETA, GAMMA RAYS

Beta rays

Beta rays, like alpha rays are not true rays since these consist of negatively charged particles moving at speed ranging from 30 to 99 per cent of the speed of light. Beta particles are actually electrons and the velocity of flight of individual electrons varies considerably for a given element as well as for different elements. The penetrating power of beta particles varies with their speed. They normally travel several hundred feet in air. Shielding with aluminium sheet a few millimeters thick will stop the particles. The ionizing power of beta rays is much weeker than that of alpha rays. (5)

Gamma Rays

Gamma rays are true electromagnetic radiations which travel with the speed of light. They are similar to X-rays but have shorter wave-lengths and, therefore, greater penetrating power which increases as the wave-length decreases. Proper shielding from gamma rays requires several centimeters of lead or several feet of concrete. The unit of gamma radiation is the photon (the light-particle).

10. Atomic Changes Resulting from Release of Radiations.

The change that atoms undergo when releasing alpha particles is considerably different from the change when beta particles are released. These changes are illustrated

HALF LIVES OF COMMON RADIOACTIVE ELEMENTS.

ATOMIC NUMBER	ELEMENT	HALF LIFE	NATURE OF RADIATION
1	H^3	12 years	β
6	C^{14}	5,570 years	β
11	Na^{24}	14.8 hours	β, γ
15	P^{32}	14.3 days	β
16	S^{35}	87.1 days	β
27	Co^{60}	5.3 years	β, γ
35	Br^{78}	6.4 min	β, γ
38	Sr^{90}	25 years	β
88	Ra^{226}	1,590 years	α, γ
92	U^{238}	4.51×10^9 years	α

in Fig. 2 and were formulated into so called displacement laws by Fajans, Rutherford, and Sodd as follows -
Alpha particle Release - "When an element emits an alpha particle, the product has the properties of an element two places to the left of the parent in the periodic table". In other words emission of an alpha particle decreases the mass number by four units and the nuclear charge, or atomic number by two units.

Beta-Particle Release - "When an element emits a beta-particle, the product has the properties of an element one place to the right of the parent in the periodic table. In this change the mass remains the same and the atomic number increases one unit.

Gamma Radiations - Gamma radiation may accompany the release of alpha or beta particles and is a result of energy released by nuclear transformations. (Ref.(3))

11. Half Lives

Radioactive decomposition is a true unimolecular reaction. The rate is constant over a wide variety of environmental conditions. Half-lives of the radioactive elements vary from a very small fraction of a second to about 10^{12} years (2). The half-lives of a number of elements are given in Table 2.

12. Atomic Transmutations and Artificial Radioactivity

The experimental conversion of one element into another was accomplished by Rutherford in 1919. When alpha-particles derived from Radium C were passed through nitrogen gas, protons were detected. The collisions between alpha particles and nitrogen nuclei resulted in the formation of an isotope of oxygen and a proton as follows -

$$ {}_7N^{14} + {}_2He^4 \longrightarrow {}_8O^{17} + {}_1H^1 \quad \dots\dots\dots (1) $$

By 1922, Rutherford and chadwick had shown that all elements in the periodic table between boron and potassium, except carbon and oxygen, underwent similar transmutations when submitted to bombardment by alpha particles.(5)

It was not until 1930 that radiation other than protons was detected when elements were subjected to alpha-particle bombardment. In that year, Bothe and Becker discovered a very penetrating, neutral, secondary radiation when beryllium or lithium was subjected to alpha particles from polonium. In 1932, Chadwick showed the particles to be neutrons, and the changes were described as follows:

$$ {}_3Li^7 + {}_2He^4 \longrightarrow {}_5B^{10} + {}_0n^1 \quad \dots\dots\dots (2) $$

$$ {}_4Be^9 + {}_2He^4 \longrightarrow {}_6C^{12} + {}_0n^1 \quad \dots\dots\dots (3) $$

Here neutron is represented by ${}_0n^1$

The third important step in transmutation of elements involved the discovery that a third particle was found in certain cases. In 1934, I Curie and Jabot noted that, when either boron, magnesium, or aluminium was bombarded with alpha particles, the expected transmutation with neutron release occurred and that positrons (positive electrons) were also produced. In addition, they found that positron emission continued after alpha bombardment was discontinued. The emission of positrons was shown to decrease in accordance with the decay law for radioactive materials. Through careful analysis of materials produced, they were able to show that alpha bombardment of these elements had produced an atom with an unstable nucleus that underwent radioactive positron decay; thus the production of artificial radioactive materials by alpha bombardment was established. (6)

$$ {}_{5}Ba^{10} + {}_{2}He^{4} \longrightarrow {}_{7}N^{13} + {}_{0}n^{1} \quad \cdots\cdots\cdots (4)$$

$$ \quad \hookrightarrow {}_{6}C^{13} + \beta^{+} \qquad (t_{\frac{1}{2}} = 9{\cdot}9 \text{ min}) $$

$$ {}_{13}Al^{27} + {}_{2}He^{4} \longrightarrow {}_{15}P^{30} + {}_{0}n^{1} \quad \cdots\cdots\cdots (5)$$

$$ \quad \hookrightarrow {}_{14}Si^{30} + \beta^{+} $$

It soon became apparent that there is no real distinction between a nuclear reaction leading to stable products and one leading to unstable products. According to the Bohr concept, all bombardments result in an absorption of the bombarding particle by the nucleus to produce and unstable compound nucleus.(3) The life of the compound nucleus is extremely short (10^{-12} to 10^{-14}sec), and decomposition occurs to a set of products. These products may be stable or they may be unstable.

The discovery of neutrons and the fact that radioactive elements could be produced artificially set the stage for the tremendous developments in the area of nuclear energy that have occurred in the past few years.

13. Units of Radioactivity

The unit of radioactivity is the "Curie". Formerly, it was considered to be the number of disintegrations occurring per second in one gram of pure radium. Since the constants for radium are subject to revision from time to time, the Inter-national Radium Standard Commission has recommended the use of a fixed value, 3.7×10^{10} disintegrations per second, as the "standard curie" (c).

The curie is used mainly to define quantities of radioactive materials. A curie of an alpha emitter is that quantity which releases 3.7×10^{10} alpha particles per second. A curie of a beta-emitter is that quantity of material which releases 3.7×10^{10} beta-particles per second, and a curie of gamma emitter is that quantity of material which releases 3.7×10^{10} photons per second. The curie represents such a large number of disintegrations per second that the millicurie (mc) and microcurie (c), corresponding to 10^{-3} curie and 10^{-6} curie respectively, are more commonly used.

The "roentgen"(r) is a unit of gamma or X-ray radiation intensity. It is of value in the study of the biologie effects of radiation that result from ionization induced within cells by the radiations. The roentgen is defined as the amount of gamma or X-radiation that will produce in one cubic centimeter of dry air, at 0°C and 760 mm pressure, one electrostatic unit of electricity(esu).

This is equivalent to 1.61 x 10^{12} ion pairs per gram of air and corresponds to the absorption of 83.8 ergs of energy.

The roentgen is a unit of the total quantity of ionization produced by gamma or X-rays, and dosage rates of these radiations are expressed in terms of roentgens per unit time. In the United States, the maximum allowable continuous daily dose for humans is considered to be 0.06 .

With the advent of atomic energy involving exposure to neutrons, protons, alpha and beta-particles which also have effects on living tissue, it has become necessary to have other means of expressing ionization produced in cells. Three methods of expression have been used.

The "roentgen-equivalent-physical" (rep) is defined as that quantity of radiation (other than X-rays or gamma-radiation) which produces in one gram of human tissue ionization equivalent to the quantity produced in air by one roentgen of gamma-radiation or X-rays (equivalent to 83.8 ergs of energy). The rep has been replaced largely by the term "rad" which has wider application.

The "roentgen-absorption-dose"(rad) is a unit of radiation corresponding to an energy absorption of 100 ergs per gram of any medium. It can be applied to any type and energy of radiation that leads to the production of ionization. Studies on the radiation of biological materials have shown that the roentgen is approximately equivalent to 100 ergs/g of tissue, depending on the energy of the X and gamma-radiation and type of tissue. The rad, therefore, is more closely related to the roentgen than is the rep, in terms of radiation effects on living tissues, and is the term preferred by biologists.

The rad represents such a tremendous radiation dose, in terms of permissible amount for human beings,that another unit has been developed specifically for man. The term "roentgon-equivalent-man"(rem) is used. It corresponds to the amount of radiation that will produce an energy dissipation in the human body that is biologically equivalent to one roentgen of gamma-radiation or X-rays or approximately 100 ergs.

(For units of radioactivity, refer 8 & 1.)

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CHAPTER 2.

RADIOACTIVE CONDITIONING IN RELATION TO PUBLIC HEALTH

RADIOACTIVE CONDITIONING IN RELATION TO PUBLIC HEALTH

2. 1. Introductory.

Consideration of radioactive contamination of domestic waters and the health hazards due to these, are of recent origin. The health hazards involved due to these sources are sometimes latent and fatal. Water is a universal solvent and in this case also it carries the radioactive substances in solution or suspension. Due to great advancement of science and large scale use of atomic energy for peaceful and other purposes in countries like U.S.A., U.S.S.R., U.K. etc., the problems of studying the subject from the Public Health point of view was also initiated in those countries. It has now been decided beyond doubt that the drinking waters should only have a limited concentration of a particular radioactive isotope or else the health hazards involved are probably so large as may be difficult to control otherwise (by treatment). Also "Prevention is better than cure".

There are countries (U.S.A. for example) where the maximum permissible standards (MPC) of radioactive isotopes in drinking water have been established (for some of the isotopes) and measures taken to control the contamination.

There are legislation for controlling this and hence safety is assured. In this chapter effort has been made to present the nature of the ill effects due to radiation exposure and also to explain some of the elementary terminology of common occurance like "Critical Organ" etc. The MPCs of the radioactive isotopes in drinking water have also been included. The idea is to stress the importance of the health hazards due to radioactively contaminated water to a water-works engineer, to explain to him the various mechanisms of contamination and to provide him with Maximum Permissible Standards of the radioisotopes in drinking water so that he may be able to control the same within limits. It is important to bear in mind that Standards are subject to change from time to time and one should be in touch with the current research to know the uptodate information.

2.2. Sources of Radioactive Contamination of Water

(a) Natural.

Radioactivity is a pollutant only in a relative sense, because it exists everywhere in the environment. It is found naturally in soils. All natural waters contain some dissolved radioactive materials that are leached from uranium or thorium-bearing areas present in varying amounts throughout the earth's crust. On the average a volume of earth one square mile in area and one foot deep contains about 18 tons of thorium and 6 tons of uranium. Such a volume of earth will contain approximately 50 curies of radioactivity. (1) Thorium and uranium are radioactive and yield such daughter products as radium and radon. There are more than 50 naturally occurring radioisotopes including such ones as K^{40} and C^{14}. Ground and surface waters leach these radioactive materials from the earth, and they appear in water as "background radioactivity". (1) Although little information is available on natural backgrounds in past years, it may be assumed that the background at any given site was essentially constant prior to 1945. Today, background measurements are subject to wide variations, resulting principally from nuclear weapons and other tests of the U.S., the U.K., the U.S.S.R. Governments etc.

(b) Man-made.

(b1) Testing of Nuclear Weapons.

Since the development of atomic fission and the testing of atom and hydrogen bombs increasing amounts of radioactive dusts have been dispersed into the earths atmosphere. Following the detonation of a nuclear device a large mass of hot gas and pulvarised debris is carried upward sometimes to more than 80,000 feet. When the detonation takes place near the ground a large amount of soil and vapourised building material may be included in the rising mass. Vapourised radioactive material released by the explosion condenses on small particulate material present and ultimately falls back to earth as dust.

Radioactive fallout has a physical appearance similar to ordinary dust of the same particle size. Where and when it falls depends upon the prevailing air mass movement and precipitation. It is this radioactivity that, during the past few years has raised very significantly the levels of radioactivity in waters.

(b2) Other Uses of Nuclear Energy.

With the availability of a great variety of radioisotopes at low cost, many applications have been found for their use in medicine, industry, and research. Naturally, this increased demand for radioactive materials has resulted in increased discharge of these materials into the environment. One of the most obvious and convenient method of disposal is through discharge into sewerage system as indicated by the survey of the disposal particles of 1027 of users of radioisotopes. Forty one percent of the users disposed of their spent radioactive materials by dilution and discharge into the sewer. (9)

Following release into the sewerage system, the radioactive wastes may discharge directly into a stream or other body of water, or they may pass through a sewage treatment plant before entering a stream. Some radioactive materials will be taken out by the physical, chemical and biological processes employed in the treatment of domestic sewage and industrial wastes. Many water courses in addition to providing for disposal and dilution

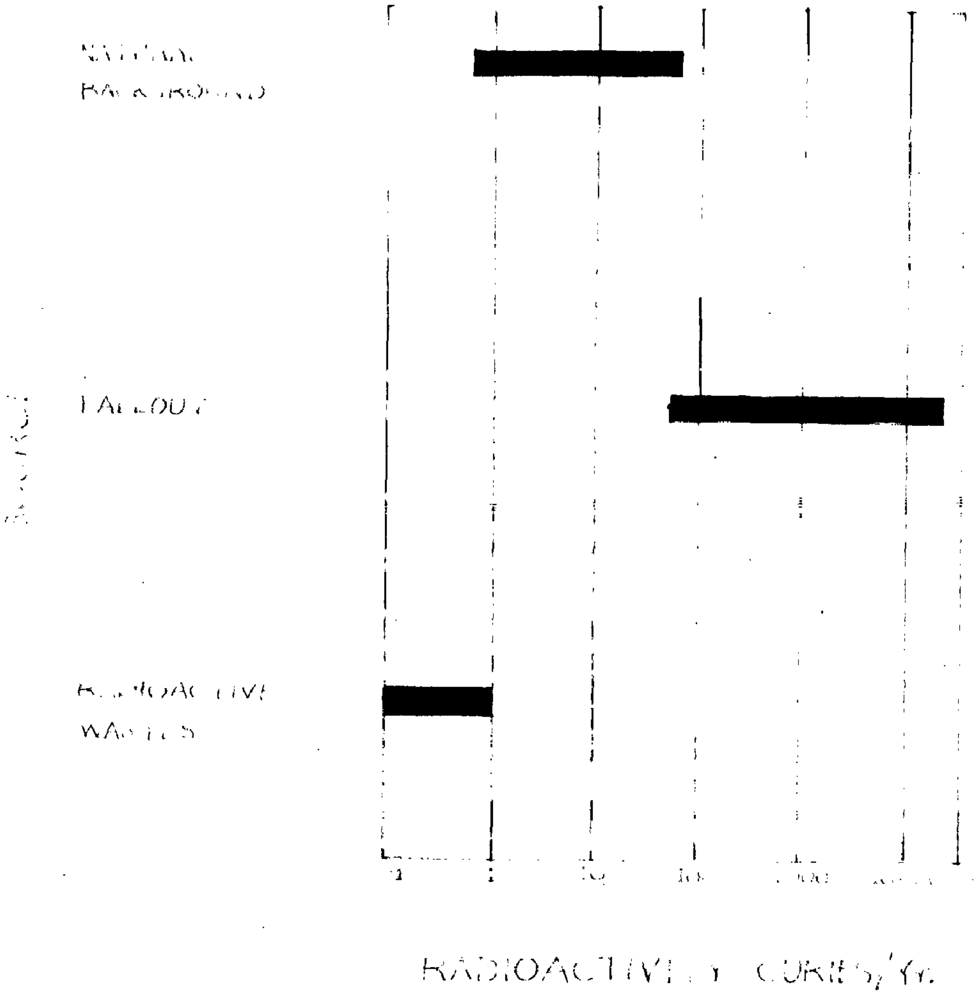

FIG. 1.

SOURCES OF RADIOACTIVITY IN PENNSYLVANIA WATERS of sewage, serve as sources of public water supplies, and the release of radioactive materials into such streams increase the radioactivity of such waters above maximum permissible concentration in water. Investigations were carried out in Pennsylvania, U.S.A., and Figure 1 shows the estimated ranges of radioactivity from various sources found in Pennsylvania streams during 1958-60.(9) The greatest amount of radioactivity found in the streams during the past few years was a result of fallout from nuclear tests.

2.3. Biological Effects of Radiation.

Man has always been exposed to radiation from cosmic and other sources. Present developments in nuclear energy are such that, with the passage of time, the radiation background, will be raised significantly by radiation sources of man's own making, if these are not controlled.

A nuclear fission is accompanied by high levels of immediate radiation and also by the production of a great number of radioactive isotopes, each with its characteristic radiation energy and radioactive life. The radiation emitted are of several kinds, differing both in character and energy.(3) The health concern arises from the fact that energy in these forms may be delivered from a distance, travelling a vacuum or penetrating materials ordinarily considered opaque to toxic substances. The nature of the biologic effects of these radiations and the means of controlling exposure become of primary concern to the public health engineer for the decades that lie ahed.

How does nuclear radiation exert its effect on living cells? While there are several different physical forms of nuclear radiation, they all have similar biologic effects, so that we may speak of various aspects of health with respect to radiation without becoming involved too deeply in the physical description of the radiations themselves. It appears that the chief mechanism of injury to living cells is through the ionization of water, of which all living tissues are largely composed. In a very real sense, man is an aquatic animal. He not only is mostly composed of water, but he lives and continues to exist in an aqueous atmosphere, being dependent on food stuffs which are themselves mostly water. Radiation appears, therefore to strike at life through the medium of its most basic substance - water (2).

From what is already known about the biologic effects of radiation, an intensification of the radiation background is likely to lead to what are known as "Somatic" and "Genetic" effects in man. The first of these has to do with the effects on the individual himself. The latter are manifest only in children or grandchildren of the individual exposed. The genetic effect results from the action of radiation on the germ cells of either male or

[illegible]
(Reference [illegible])

1895	DISCOVERY OF X-RAYS	ROENTGEN
1896	DERMATITIS OF HANDS (JANUARY)	GRUBBE
	SMARTING OF EYES (MARCH)	EDISON
	EPILATION (APRIL)	DANIEL
1897	CONSTITUTIONAL SYMPTOMS	WALSH
[illegible]	PAINFUL AND SPASMODIC MUSCULAR CONTRACTION	[illegible]
[illegible]	[illegible] VASCULAR ENDOTHELIUM	GASSMANN
1902	CANCER IN X-RAY ULCER	FRIEBEN
[illegible]	[illegible]	PERTHES
	STERILIZATION PRODUCED	ALBERS-SCHÖNBERG
1904	BLOOD CHANGES PRODUCED	MILCHNER & MOSSE
	[illegible]	HEINEKE
[illegible]	[illegible]	[illegible]
[illegible]	[illegible] X-RAY WORKERS	BÉCLÈRE

female. These matters have been most thoroughly reviewed by the comprehensive study on radiation injury conducted by the National Academy of Sciences.

(a) Somatic Effects.

Somatically, certain type of disease tend to be associated with excessive radiation exposure. Among these are specially to be noted, leukemia - which is a cancer of the bone marrow or the blood forming tissues - and increased frequency of carcinoma or sarcoma, the latter specially arising in the skeleton of those who have aquired excessive amounts of radioactive materials with special affinity for bone. A very important general manifestation with no specifi pattern of disease is decreased life-span, which is found to be quantitatively related to the total amount of radiation exposure. The measure of this effect is an increased rate of general aging. The exposed population simply grows older at a rate greater than that of the normal for that population. Evidence is accumulating that this is probably the most sensitive indication of population injury that is available and it is probable that studies of whole population will demonstrate changes which would be imperceptible on the basis of individual observation. Much remains to be learnt, but from animal studies, as well as careful observation of human experience both in United States of America and Japan, it appears that 100 roentgens of gamma radiation to the whole body may shorten life expectancy by an amount between half an year and two years. Similar proportionate shortening of life span appears to exist for many experimental animals as well.

(b) Genetic Effects.

In considering the question of genetic effects certain general scientific principles have been well established. It is known that gamma radiation to the germ cells is effective in producing those alterations in genetic characteristics called mutations. (2) These are characteristics not previously present in that particular line of inheritance. A second general important conclusion is that for purposes of genetic analysis the effect depends upon total doze and is not importantly determined by the rate at which the radiation has been delivered. A third generality is that within the range of actual experimentation the frequency of mutations is approximately proportional to the amount of radiation.(4)

The mutations produced may lead to serious abnormality in some cases or to trivial departures from the normal in others. In general these departures from normal are seriously detrimental.(5) Most modern geneticists look with concern on any appreciable increase in the frequency of mutations in man. Not enough is as yet known of human mutation frequency or the susceptibility to mutational change induced by radiation in human species. Because of this area of ignorance there is general agreement among geneticists that radiation exposure should be kept to the lowest possible amount, although there is appreciable disagreement concerning the quantitative aspects of the ultimate effect upon the population. (5)

With respect to all these biologic effects, both somatic and genetic, it is a good working principle that as the fraction of the total population exposed increases there should be a dimunition in the maximum amount of individual exposure permitted. Frequencies of injury that might cause little concern if they applied to small groups of people may be deserving of the most serious attention when large populations are involved. Thus the standards for permissible exposure need to be more stringent when large blocks of population are irradiated than when the exposure is limited to a comparatively small number of persons.

2.4. Permissible Dose.

The problem of how to define a permissible dose becomes more difficult than that of specifying what dose had been tolerated by a few people. After much delibrations the International Commission on Radiation Protection defined permissible dose as "a dose of ionizing radiation that in the light of present knowledge is not expected to cause appreciable bodily injury to a person at any time during his lifetime". "Appreciable bodily injury" is defined as "any bodily injury or effect that a person would regard as being objectionable and/or competent medical authorities would regard as being deleterious to the health and well being of the individual". (5) In attempting to give a numerical value to the permissible dose the fact that man is and always has been bombarded by the ionizing cosmic rays and the radiations from the natural radioactive elements must be taken into consideration. These radiations provide a lower limit that is inescapable. (about 10 mr per wee). (5)

The upper limit of permissible exposure is based on the experience of radiation workers (mostly medical) over the past 60 years, and on such animal experiments as seems to be pertinent. There are several factors that make the interpretation of the human experience difficult. First, we have no real measure of the level of exposures prior to about 1942. Second, the long latent period between cause and effect - sometimes 25 years, - makes it difficult to connect the two. Third, the biologic variability of man is so great that some people known to have been overexposed have had no complication while others not known to have been overexposed have died of such conditions as leukemia that probably were due to such exposures as they suffered.

2.5. Critical Organs.

Reviewing the experience of man and the results of animal experimentation revealed clearly that some organs of the body are critical to the problem of permissible exposure. The skin is the most exposed organ of the body. It was first known to be injured and is one most frequently involved.

It was recognised early in the study of radiation effects that any peripheral blood change was one of the first objective signs of the effect of X-rays. The hemopoietic tissue was thus established as a critical organ, the changes in which can be followed by studies of the peripheral blood. The other critical organs which are important are the gastro - intestinal tract(G.I.tract), lungs, kidneys, Thyroid, Liver, Bone spleen and wounds, in particular cases. (5)

The fact that why only one out of the several organs described above becomes the critical organ depends upon the type of exposure, that is, whether the exposure is due to external radiation, injection or through a surface wound etc.,and also upon the concentrations of the radioactive material in air or water or food and the kind of radioactive material, that is, whether it is U^{238} or I^{131} etc.

2.6. Relative Biological Effectiveness.

The Relative Biological Effectiveness (RBE) of radiations of various types and energies is a necessary factor in determining the permissible dose of radiation

TABLE 2. R.B.E. VALUES.

(Ref. 5)

Average Specific Ionization (Ion pairs/µ of water)	RBE
100 or less	1
100 to 200	1 to 2
200 to 650	2 to 5
650 to 1500	5 to 10
1500 to 5000	10 to 20

TABLE 4.

(Ref 6)

Element	NCRP. MPC for water in curies per litre.	Standard Recommended by the Radiological Congress in London in 1950, in curies/litre.
Ra^{226}	5×10^{-11}	4×10^{-11}
I^{131}	$0{\cdot}5 \times 10^{-8}$	3×10^{-8}
Co^{60}	5×10^{-8}	1×10^{-8}
P^{32}	1×10^{-7}	2×10^{-7}
Na^{24}	1×10^{-5}	$0{\cdot}8 \times 10^{-5}$
$Sr^{90} + Y^{90}$	1×10^{-9}	$0{\cdot}8 \times 10^{-9}$

for other than X-rays up to 250 KV.(5). The biological effectiveness of a radiation is the relationship between a given biological reaction and the dose of radiation required to produce it. Since for both man and animals the effects of X-rays up to 250 KV. are known with greater precision than for other types, the biological effectiveness of such rays is arbitrarily established as the base line. The relative biological effectiveness of a given radiation is the factor that relates its biological effectiveness to that of 250 KV. X-rays(5).

Since the biological effectiveness is known to be related to the density of ionization along the paths of the ionizing particles, this characteristic can be used to determine the RBE. The linear ion density per unit of path is called the specific ionization. Various combinations of photon or particle radiations with differing energies may have similar specific ionizations. Thus, a very high energy proton may have the same specific ionization as a low energy electron. (8). It is thus better to relate RBE to the specific ionization rather than to any particular type of radiation. The average specific ionization of 250 KV X-rays is 100 ion pairs per micron of water and by definition the RBE is 1.

The problem of relative biological effectiveness is complicated by the fact that, when determined by different biological reactions, even in the same organism the

RBE is not always the same. When mammals only are used the differences are not great enough to seriously complicate protection problems.(5).

In practice if one wishes to determine the permissible dose of a given radiation, he first finds its specific ionization. From the data collected by the International Commission on Radiological Protection and reproduced below he obtains the RBE for that radiation. The basic permissible dose in tissue roentgens for 250 KV X-rays multiplied by this RBE factor gives the permissible dose in rems. The rem is a unit defined as "the absorbed dose of any ionizing radiation which has the same biological effectiveness as 1rad of X-radiation with an average specific ionization of 100 ion pairs per micron of water, in terms of its air equivalent, in the same region". In practice, a dose in rems is equal to the dose in rads multiplied by the appropriate RBE. Its greatest usefulness is in integrating exposures involving various kinds of radiations.(5)

2.7. Standards for Maximum Permissible Concentration of Radioactive Elements in Drinking Water.

Table No. 3 shows the maximum permissible concentrations of radioactive isotopes with other details

TABLE 5

[illegible]

MAXIMUM PERMISSIBLE CONCENTRATION
IN CURIES/CC

[illegible]	AIR	WATER
$Kr^{[illegible]}$	1×10^{-14}	[illegible] $\times 10^{[illegible]}$
$R[illegible]$	1×10^{-11}	[illegible] $\times 10^{-9}$
$Sr^{90} + Y^{90}$	1×10^{-12}	[illegible] $\times 10^{-9}$
[illegible]	5×10^{-12}	5×10^{-9}
Sr^{89}	1×10^{-11}	[illegible] $\times 10^{-8}$
Ca^{45}	$5 \times 10^{[illegible]}$	5×10^{-8}
$Ba^{140} + La^{140}$	[illegible] $\times 10^{-11}$	5×10^{-8}
P^{32}	1×10^{-10}	1×10^{-7}
$Cs^{137} + Ba^{137}$	5×10^{-10}	$5 \times$ [illegible]
$[illegible]^{95} + Nb$	5×10^{-10}	5×10^{-7}
Ag^{110}	5×10^{-10}	1×10^{-6}
C^{14}	5×10^{-9}	1×10^{-6}
Mn^{56}	1×10^{-9}	1×10^{-6}
S^{35}	5×10^{-9}	1×10^{-5}
Na^{24}	5×10^{-9}	1×10^{-5}

like half-life etc. For detailed information regarding the standards one is advised to consult the Handbook-52 of the National Bureau of Standards, "Maximum Permissible Amounts of Radioisotopes in the Human Body and Maximum Permissible concentrations in Air and Water". For sale by Superintendent of Documents, Washington 25, D.C. (1953) and ICRP/54/4, Recommendations of the International Commission on Radiological Protection W.Binks, Secretary, London S.W.7, England (1954). However, work has also been done in USSR on these lines and Tables 4 and 5 show some of the Standards.

Some insight into the rationale of current standards pertaining to maximum permissible concentration of radioactive isotopes in drinking water may be obtained by consideration of mathematical model shown in Appendix A. The model simulates some of the characteristic features of isotope intake and decay in vital organs of human beings and animals.

While the use of the formulation set fourth in Appendix A leads to quantitative results for limiting concentrations, it must be realized that a number of rather arbitrary assumptions are entailed. These are chiefly resident in the concepts of, and the numerical values assigned to the permissible weekly dosage, W, the biological half-life, T_b, and the distribution factor, f_w. The MPC values will doubtlessly be subjected to considerable revision in the future as more extensive experimental evidence becomes available. The rate at which this refinement proceeds will in part be dependent upon the frequency of accidents involving over exposure. At present the critical values such as those listed in the last column of Table III must be regarded as the best obtainable under the circumstances, and many of them may prove to incorporate little, if any, safety factor.

The formulation applies to steady state contamination of drinking water with radioactive isotope. Various unsteady state formulations have been used that are of a form more immediately applicable to evaluation of the hazard from radioactive fallout. As an operational guide in continental weapons tests, the AEC has used the following criterion: to determine whether the concentration of radioactivity in water is below the dangerous level for drinking, the observed activity level is extrapolated to the value it would have at three days after fission by "1.2 decay law"; if the extrapolated value is less than 0.005 micro curies per milliliter the water may be safely used for any length of time provided the water has not been contaminated by antecedent detonations and is isolated from subsequent fallout.

2.8. Discussion on Maximum Permissible Standards.

It is well recognised that the validity of maximum permissible standards for internal exposure to radioisotopes is not comparable with the validity of standards for external radiations. The International Commission on Radiological Protection (ICRP), in 1950, did not "consider that there is sufficient information to make firm recommendations concerning maximum permissible exposures to internal radiation from radioactive isotopes".

In 1953, the sub-committee on Permissible Internal Dose of the National Committee on Radiation Protection(USA) published maximum permissible exposure limits for 70 internal emitters. Recognising that, "in some cases there is considerable uncertainty about the maximum permissible values given ....", the Sub-Committee felt that it was "desirable to agree upon what are considered as safe working levels for these radioisotopes now rather than wait until more complete information is available.

Plutonium-239.

The presently accepted MPC for Pu^{239} in water is 1.5×10^{-6} $\mu c/ml$[2]. This is based, among other parameters, on an assumed absorption from the gastro-intestinal tract of 0.1 per cent of the ingested dose, a figure derived from a measured adsorption of 0.01 per cent, with a safety factor of ten included because measurements were made at relatively high plutonium concentrations.

Ruthenium - 106.

Current maximum permissible standards for internal exposure to Ru^{106} are based on limited observation on few rats. The absorption of ruthenium from the gastrointestinal tract is reported as 0.05 percent, and on this basis the MPC for Ru-106 in water is recommended as 0.1 μc/ml[2].

Kidney is presently considered to be the critical organ for ruthenium internal exposure, and the biologic half-life for ruthenium in kidney is listed as 20-days. Long-term studies on ruthenium retention suggest strongly that bone rather than kidney may be critical organ. While kidney accumulates the highest internal concentrations, the half-life for ruthenium retention in bone is of the order of 100-150 days. Data from chronic feeding studies is not yet available, but calculations based on existing data indicate that equilibrium concentration of ruthenium in bone and kidney should be approximately equal. On this basis, bone, in view of its greater radiosensitivity should be considered the critical organ. Assuming bone to be the critical organ the MPC value for Ru^{106} becomes 1×10^{-3} μc/ml. This is a factor of 100 lower than the presently recommended MPC. It is recommended that this value be tentatively accepted.

Iodine-131.

This isotope is not important for the public health engineer so far as damages due to this through water contamination is concerned. It usually causes trouble in the thoroid gland (the critical organ for this) and the intake of the material is by eating vegetables and other food products from I^{131} - contaminated lands. The milk of cows fed on such pastures as have been contaminated by the I^{131} shall also be a source of this isotope for man.

Phosphorus- 32.

The presently recommended MPC for P^{32} in water is 2×10^{-4} μc/ml^2. While this is a reasonable, perhaps even conservative, limit for drinking water, it is inapplicable to biologically populated waters. Here the remarkable concentration is small aquatic organisms and their use in food chains by fish and man necessates lower limits. At 2×10^{-4} μc/ml, plankton receive dose rates up to 70 rads/hour, and some fish may receive up to 50 rads/hour. Humans eating one lb of these fish per day would receive about 200 rads per week, at some seasons of the year. The MPC for P^{32} in such waters should not be higher than 3×10^{-6} μc/ml, or better, 3×10^{-7} μc/ml to include the conventional factor of safety of 10^7.

Sodium- 24.

The drinking water MPC for Na^{24} is listed as

8×10^{-3} μ c/ml[2]. For the protection of large fish, the MPC should not exceed 2×10^{-6} μ c/ml[7].

2.9. Appendix A.

Mathematical Model of Uptake and Radiation in Human Tissue (Ref. (1))

The following notation is used -

Q = the steady state rate of flow of blood or lymph through the critical organ or tissue, grams/day.

m = mass of the critical organ or tissue, grams.

c = the natural concentration of the (stable) element in the critical organ or tissue, grams of element/critical tissue in grams.

X_o = the concentration of radioactive element under consideration in the blood delivered to the organ; atoms/gram of blood.

X = concentration of radioactive element leaving the organ, atoms/gm of blood.

Y_o = the concentration of the stable element of the same chemical species delivered to and removed from the organ in the blood, atoms/gm of blood. It is presumed that $Y_o \longrightarrow > X_o$.

W = the radiation exposure to the critical organ.
= 0.3 rep/week for gamma and beta radiation.
= 0.015 rep/week for alpha radiation.

$\Sigma (bE)$ = the effective energy of radiation per distegration, in millions of electron volts, mev.

$(MPC)_w$ = the maximum permissible concentration of the isotope in drinking water, microcuries per milliliter.

A = $(6.03)(10^{23})/$ (gram molec. Wt. of element).

L = the number of grams of water injested per day per person.

f_w = the fraction of element injested that is delivered to the critical organ.

λ = the decay constant of the radioisotope, per day.

= $0.693/Tr$, where T_r is the radioactive half-life in days.

T_b = the biological half-life of the stable elements, days.

= (0.693) Acm/QY_0.

T = $Tb\ Tr/(Tb + T_r)$ = the effective half-life of the radioisotope.

Assume a steady state with complete mixing and free exchange of both isotopes of the element within the organ so that the distribution is uniform throughout and in dynamic equilibrium with the concentration in the fluid leaving the organ. An activity balance is given in the following equation -

$$Q = (X_0 - X) = \lambda A c m (X/Y_0) \cdots\cdots (1)$$

$$(X_0 - X) = \lambda X (T_b/0{\cdot}693) = X(T_b/T_r) \cdots\cdots (2)$$

$$\text{or} \quad 1 - X/X_0 = T_b/(T_b + T_r) \cdots\cdots (3)$$

The radiation damage in the organ depends upon the disintegration rate within the organ, and the energy release per disintegration.

$$W = K_1 m^{-1} Q (X_0 - X) \Sigma (bE) \cdots\cdots (4)$$

Where K_1 is a scale factor depending upon the units of measurement used. The amount of active element delivered to the organ in steady state is -

$$K_2 L (MPC)_w f_w = \lambda Q X_0 \cdots\cdots (5)$$

Where K_2 is a scale factor depending on the units employed. Eliminating Q, X, and X_0 from equation (4) using equation (3) and (5),

$$W = \frac{K_1 K_2 L (MPC)_w f_w T_r T_b \Sigma (bE)}{0{\cdot}693 m (T_b + T_r)} \cdots\cdots (6$$

Finally, solving for the maximum permissible concentration of the isotope with appropriate values of the scale factors, and setting W = 0.3 rep/week, and L at an average value of 2.2 kilograms per day, the following relationship is obtained:

$$(MPC)_w = \frac{2{\cdot}34 \times 10^{-7} m}{\Sigma (bE) T f_w} \cdots\cdots (7$$

In this steady-state form the formula is equivalent to equation G-6 of Handbook 52 of the National Bureau of Standards and has been used to establish the maximum permissible concentration in drinking water of most of the radioactive fission products (Table III., last column).

For example, for promethium ^{147} the following parameters apply

$\Sigma(bE)$ = 0.067 mev., m = 7,000 grams (bone);
T_r = 1.46×10^3 days; T_b = 100 to 150 days;
T = 140 days; $f_w = 1.7 \times 10^{-4}$;

Substituting these values in equation (7) a value of 1 microcurie is obtained.

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CHAPTER 3.

DETECTION AND MEASUREMENT OF WATERBORNE RADIOACTIVITY

DETECTION AND MEASUREMENT OF WATERBORNE RADIOACTIVITY

3.1. Introduction.

The rate of decay $\frac{dN}{dt}$ of any radioactive substance is equal to the number of radioactive atoms present multiplied by the decay constant, so that for any particular isotope, the measurement of the rate of emission is a measure of the amount of radioactive material present(1). Nearly all the methods for the detection and measurement of nuclear radiation involve one or the other of the following effects -

(1) The ionization of gases

(2) The scintillations, or flashes of light produced in certain "phosphors".

(3) The blackening of photographic plates.

The methods of using these phenomena can be broadly divided into two groups - integrating methods, in which the mean flux is measured, and methods of detection of individual particles. For example, the mean ionization current produced by a radioactive source can be measured, or a single particle can be detected by the electric pulse obtained on collecting the ions it produces.

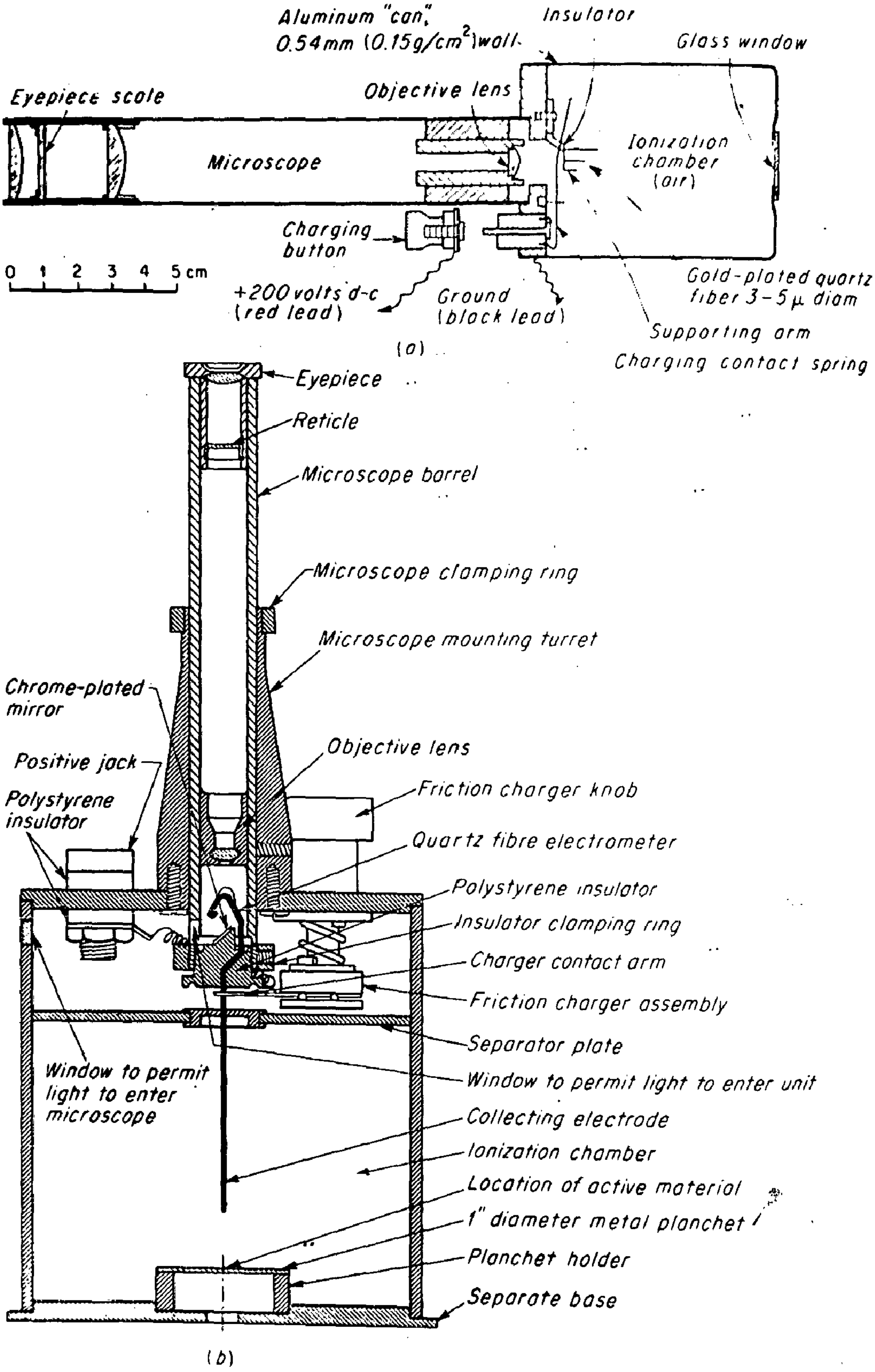

FIG. 1 (*a*) Schematic diagram of the Lauritsen electroscope. (*From C. S. Garner, J. Chem. Educ., vol.* 26, *p.* 542, 1949.) (*b*) Schematic diagram of the Landsverk electroscope. (*Courtesy of Landsverk Co.*)

3.2. Ionization of Gases.

Ionization is the process by which large numbers of positive and negative ions, or charged particles, are formed in a gas due to the passage of radiation (2). Usually an electron is stripped from an atom, producing a negative electron and a positive ion.

3.3. Integrating Methods based on Ionization.

One method of measuring activity is by means of an ionization chamber, which, in its simplest form, consists of a pair of parallel metal plates in air, with a potential of approximately 100 volts between them, and some means of measuring the ionization current produced. This ionization current produced is mostly weak (of the order of 10^{-10} to 10^{-14} amperes or less). The technique of measuring radiations usually resolves into measurement of small currents arising from the ion collection in the detector system.

(a) Electroscopes.

Modern forms of electroscope employ a gold-coated quartz fibre, the most well-known type being the Lauritsen electroscope. Diagrams of the typical electroscopes are shown in Fig. 1 Refer (2) (pages 46 and 47). The electroscope consists of an air-filled chamber containing a highly insulated electrode connected to a gold coated quartz fibre, the image of which thrown by a small light on to a graduated scale, is viewed through the eyepiece of a microscope attachment. Electroscopes specially designed for alpha, beta, or gamma radiation are available.

(b) Ionisation Chambers.

Ionization chambers are used in conjunction with current measuring devices, such as electrometer valves, direct-coupled amplifiers, or vibrating reed electrometers. They may be used for the measurement of alpha, beta, and gamma radiation, neutrons and other types of radiation or charged particles.(Fig.2).

An ionization chamber consists of a pair of electrodes with air or other gas in the intervening space. A potential of the order of 100 volts is maintained between the electrodes. This prevents the recombination of the ions produced by radiation. A potential gradient of about 10 volts per cm. is normally sufficient for collection of all the ions. The current then obtained is known as the "saturation current". Figures 3, 4 and 5 indicate the various ionization chamber arrangements.

(c) Electrometers.

The ionization current is measured by allowing it to flow through a high resistance (R in Figs. 5,6 & 7), the current I producing a voltage IR across R. This voltage can be measured with reasonable accuracy if it is of the order of 1 volt, so that for ionization currents, R must be 10^9 to 10^{14} ohms. Stable resistors of this order are expensive to manufacture, costing a few pounds each and 10^{13} ohms is about the highest so far manufactured.

To read the voltage across R, a suitable voltmeter of extremely high impedance, which does not reduce the effective value of R, is used. Special electrometer valves designed for this purpose are available. These have a very high input impedance, and a very low grid current. The circuit of a typical portable battery-operated instrument using such a valve is shown in Fig.6.

There are also circuits employing tetrodes and pentodes. The above type of circuit, though simple, has a number of disadvantages, so that it is only used in small portable radiation meters.

3.4. Counting Instruments.

For the measurement of alpha, beta or gamma radiations at tracer levels, techniques involving the detection and counting of individual particles or photons are almost universally employed. Among the gas ionisation devices are the pulse ionisation chamber, proportional counter and Geiger-Muller counter. Scientillation methods are available for counting alpha and beta particles, and

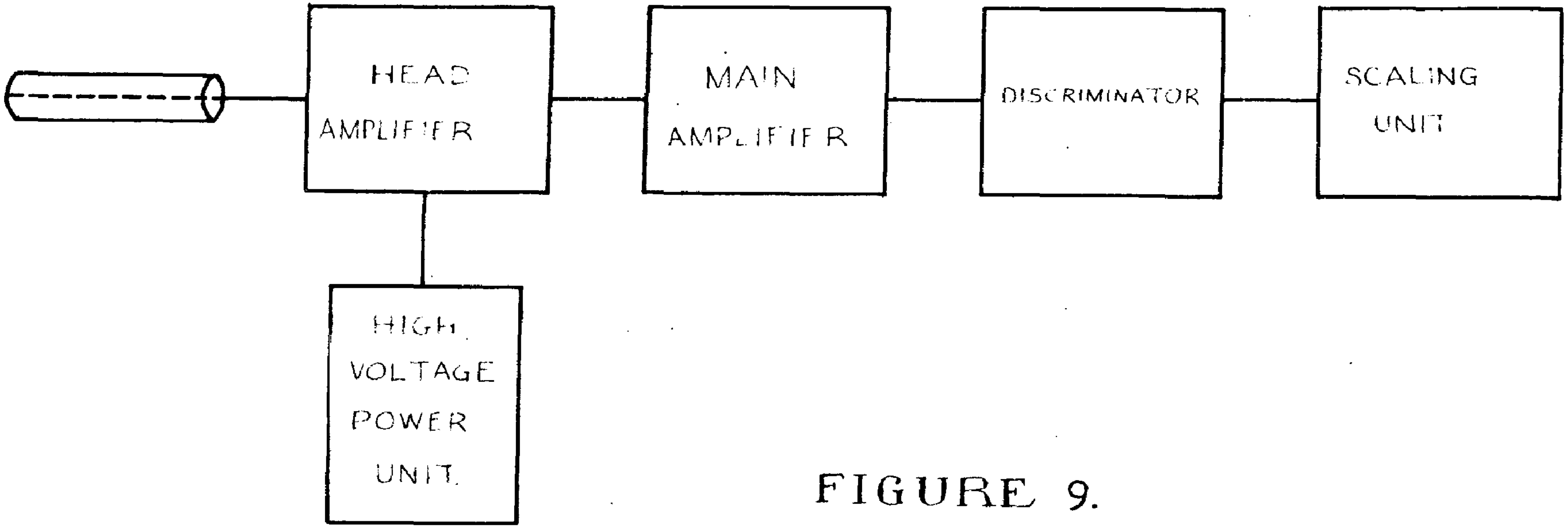

FIGURE 9.

BLOCK DIAGRAM OF PROPORTIONAL COUNTING SYSTEM.

are especially popular for gamma ray counting. Photographic techniques involving the use of nuclear emulsions may also be used for the detection and identification of individual particles.

(a) Pulse Ionisation chamber.

These have a disadvantage of high stability, and are frequently used for the most accurate work. They require the use of highly stable, sensitive electronic equipment.

(b) The Proportional Counter.

The proportional counter takes its name from the proportionality between the output pulse and the initial ionization. The design of the counter and the voltage applied are such that a very high voltage gradient exists in the vicinity of the positive electrode. Under this voltage gradient, the liberated electrons undergo a high acceleration, and are themselves capable of causing further ionization. Under suitable conditions gas multiplications of 1,000 or more are possible, the total ionization (and hence the pulse size) being strictly proportional to the initial ionization. Fig. 8 shows the effect of increasing voltage on the current produced, and the different regions which are commonly used for counting purposes. Fig. 9 shows the electronic equipment required for the satisfactory operation of a proportional counter.

(c) Geiger - Muller Counter.

The Geiger - Muller counter consists of a cylindrical cathode, normally one or two centimeters in diameter, along the centre of which is a wire anode. The intervening space is filled with a gas, or mixture of gases, which readily ionise, together with a small proportion of a quenching vapour. The detailed design of Geiger - Muller counters depends almost entirely on the purpose for which they are required. For the counting of solid sources, the end window type is most popular. The window may be aluminium alloy (7 mg/cm^2) mica (1.5 to 2.5 mg/cm^2) or may be a thin glass bubble (0.5 to 1.0 mg/cm^2). For medium and high energy beta particles (above 0.5 MeV) and for gamma counting, thin glass walled counters may be used. These are normally about 1 cm. diameter, with a glass wall of 20 to 40 mg/cm^2 thickness, the actual thickness depending somewhat on the length of the counter. The tube is coated on the inside with graphite to form the cathode

For the counting of radioactive liquids, the counter takes the form shown in (c) of Fig. 10. This has a capacity of 10 millilitres in the annular space. In such a counter, 10 ml of a 3% solution of a uranium salt will give approximately 10,000 counts per minute. This type is also available with a ground - glass stopper.

Another very useful liquid counter is the dip type shown at (d) and there are other types containing a thin-walled tube or spiral, through which radioactive liquids may be drawn.

(d) Scientillation Counters.

Scientillation methods have, in the last few years, gained rapid popularity, specially in the counting of gamma emitters. Visual methods of observing the scientillations are slow and insufficiently sensitive for the detection of single beta particles and gamma photons. The development of photomultiplier tubes, consisting of photographic cells coupled directly to electron multipliers, have led to sensitive detection devices, which today are challenging the supremacy of gas ionization detectors, such as Geiger - Muller counters, in the detection and measurement of radioactive radiation. The introduction by Hofstadter of thallium activated sodium iodide brought about an important advance in the application of scientillation techniques for gamma counting and energy determination. Many other fluorescent materials or "phosphors" are known today and the more important of these are:

1. Zinc Sulphide activated with silver (For alpha counting). Commercially available as Luminescent Powder, Type G 86, supplied by Levy-West Laboratories Ltd., of Wembley. (2) Anthracene (Most efficient phosphor for beta

INSTRUMENT	STATE AND TERRITORIAL LABORATORIES NUMBER WITH INSTRUMENTS	 TOTAL NO OF INSTRUMENTS	PUBLIC AND PRIVATE UTILITIES NUMBER WITH INSTRUMENTS	 TOTAL NO OF INSTRUMENTS
ller Survey Meter	17	28	11	15
Chamber	18	58	6	66
troscope or Electrometer	11	40	13	19.
er Muller Scaler	9	13	4	4
rtional Counter	5	7	4	4
llation Counter	4	6	1	1
Monitoring Equipment	2	3	4	5
rs	9	55	3	10
or more of any type	23		19	

PLICATION.	STATE AND TERRITORIAL LABORATORIES WITH INSTRUMENTS. TYPE A	 TYPE B	 TYPE C	PUBLIC AND PRIVATE UTILITIES WITH INSTRUMENTS TYPE A	 TYPE B	 TYPE C
ERGENCY RADIOASSAY OF WATER	2.	2	5	5	1	13
SONAL PROTECTION MONITORING.	9	12	3	0	5	0
RADIOASSAY OF WATER AND AL PROTECTION MONITORING	6	3	3	5	0	0

counting in the form of a single crystal).

3. Clear Plastics, such as polystyrene or polyvinyl toluene etc. constitude efficient organic scientillators. For up-to-date information on organic phosphors the literature supplied by manufacturers is useful; for example Messrs Mash and Thompson Ltd.

4. Sodium iodide activated with thallium. (This is the best organic scientillator for gamma counting).

A Task Group report on the nuclear instrumentation has been published in the Jour of American Water Works Association (4). This report is of value in appreciating the techniques and instruments at present employed in the United States of America. Useful data as regards the efficiencies of various instruments and their costs in dollars makes the Report especially interesting. Refer Table No.3.

3.5. The Choice of Counting Equipment.

Let some of the factors influencing the choice of counting equipment be considered. One important factor will almost undoubtly be that of cost, and if we hope at some time to be able to carry out different kinds of counting work, then versatility in equipment will be of importance. If, on the other side, it is inteded to carry out one type of investigation only, then for economy and simplicity, we require equipment which will do that work alone.

Assuming that we know what type of work we wish to carry out, and have selected likely isotopes, or we may be interested in a particular element, and have selected the most suitable radioisotope of that element, we will be in a position to decide whether we wish to do alpha, beta or gamma counting, or require to count one isotope in the presence of others.

The form of our material will help in deciding whether we wish to do solid, liquid, or gas counting - or we may require all three. Radiography or autoradiography may be preferred to counting methods. The energies involved will have a profound influence - we cannot count low energy beta particles, such as those from tritium in a normal liquid counter. The maximum levels of activity which can be safely employed may influence the shielding required around the detector. If large numbers of samples are to be counted we may well be advised to think in terms of automatic counting equipment, or at least in terms of equipment which will count a sample for a predetermined time. Where accuracy is not of major importance, a counting ratemeter may be more convenient to use than a scaling unit, particularly if a continuous recording is to be made of activity levels.

(a) Alpha Counting.

For alpha counting, the scintillation counter is recommended as the first choice, because of the wide range of counting which it will accept, and the fact that a high gain amplifier is not required. The low background counting rates (below 10 counts per hour) permit very low counting rates to be measured, and the high resolution (less than 1 sec) allows of counting rates of several thousand per second. Alpha activity can be measured in the presence of 10^6 times as much beta activity without significant interference due to beta build-up.

The alternative choice for alpha counting is the flow type alpha proportional counter, the most popular model being the Type 1077B. Using argon gas, the normal operating voltage is about 800 volts. Geiger - Muller counters with very think windows may also be used for approximate work.

(b) Beta Counting.

For the counting of solids, the Geiger counter is probably the most suitable type of counter to use. The very popular 1 in. diameter end-window version is available with organic quenching and with 7 mg/cm^2 metal window, or 2 mg/cm^2 mica windows from the G.E.C. or 20th Centuary Electronics, or with bromine quenching and a 2 mg/cm^2 mica window from Mullards.

For counting liquids, special Geiger counters, either halogen or organic vapour quenched, are available. These normally hold about 10 ml of solution. They are only suitable for the higher energy beta radiations, because of absorbtion by the walls, and by the liquid itself. Where practicable liquid counters are very convenient to use.

Bromine-quenched types are preferred so long as they are not required for coincidence work, as there is normally a delay of a few microseconds between the incidence of the ionizing particle and the output pulse. The bromine-quenched types are more robust electrically, will withstand a temporarily revised voltage, or too high a voltage, require a lower operating voltage than the corresponding organic-vapour-quenched type, have a wider temperature range, and a theoritically infinite life. The beta counting efficiency is about 95% of that of the corresponding organic quenched type, which is very nearly 100%. Other considerations of shape and size depend on the work in hand.

For counting low-level beta particles, below about 0.1 MeV, there is a choice of three methods -

(1) Gas counting as a proportional or Geiger counters;
(2) Liquid scientillation counting using internal phosphor;
(3) As a solid in flow type proportional counter.

(c) Gamma Counting.

Scientillation counting, using a sodium iodide crystal as a phosphor - is to be recommended, because of its high efficiency, which may be 50% or more, while the gamma counting efficiency of a Geiger counter is usually much below 1%. Higher counting rates are possible with the scintillation counter than with the Geiger counter as the resolving time can be made much less than 1 sec. If the activity is sufficiently high for adequate counting rates to be obtained using a Geiger counter, then this method may prove the more convenient.

3. 6. Remarks.

For detailed study and practical work it is necessary to know the sources of errors, their statistics, their measurement and methods adopted to minimize or completely eradicate them. The choice and preparation of radioactive sources also requires a considerable attention. The above descriptions of the instruments are very elementary and are just enough for a basis for detailed reading. Inclusion of these topics and detailed treatment of instruments shall make this chapter a very volumenous one.

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CHAPTER 4.

REMOVAL OF RADIONUCLIDES FROM WATER BY WATER TREATMENT PROCESSES
(PART I)

REMOVAL OF RADIONUCLIDES FROM WATER BY WATER TREATMENT PROCESSES.

4.1. Introduction.

Before a method of treatment for the removal of radioactive materials from water can be selected or provided, the sanitary engineer, consulting engineer or water works official must know several things. These include:

(1) The concentration of radioactive material in the water to be treated;

(2) The composition of radioactive materials in the water;

(3) The maximum permissible concentration (MPC) of the individual components comprising the radioactive materials; and

(4) The efficiency of the particular process or combination of processes that might be considered for the removal of the various radioelements.

To obtain information on item (1) (the concentration) the engineer or official must know what radioisotopes are used on the watershed and in what amounts, and the concentration in which they are discharged. In case of atomic fallout the intensity per square foot should be known. This will aid in the interpretation of counting data from a reasonable number of samples. If only a single isotope is found Item (2) (the composition) presents no problem. Where several radioisotopes are detected, however, radiochemical analysis must be made, and these become difficult as the number of radioisotopes in the mixture increases. Information on item (3) (MPC Values) will be found in "National Bureau of Standards Handbooks No.52 (1) Item (4), (efficiency of treatment process) will be considered later. If radioisotopes for which no information relative to the efficiency of removal afforded by various treatment processes has been published are encountered, it may be possible for water works operator to carry out studies to determine the behaviour of these radioactive materials in their passage through the plant.

The percentage of removal that must be provided by a particular water treatment process is a function of the initial concentration of the radioactive element and of the MPC value for that radioisotope, as given in the Handbook (1). Let it be assumed for example that the initial concentration of strontium (Sr^{89}) in the raw water amounts to 1.0 microcurie per millilitre ($\mu c/ml$), or 3×10^{-5} ppm. The removal required to reduce the activity to the MPC value of $7 \times 10^{-5} \mu c/ml$ (2×10^{-9} ppm) is 99.993 percent, a very high efficiency of removal. Or if the efficiency of a particular process and the MPC value are known, the maximum concentration of that component in the water to be treated can be defined. Again, using the example cited above, if the treatment processes have an overall removal efficiency of 90 per cent, the maximum concentration of (Sr^{89}) in the untreated water cannot be more than 7×10^{-4} μ c/ml (2×10^{-8} ppm).

4.2. Radioactive Materials.

Before speaking specifically of water decontamination and citing experimental results, several characteristics of radioactive materials should be mentioned.

(1) The rate of disintegration or decay of radioisotopes cannot be modified by any physical, chemical, or biological processes. In the treatment processes employed, the radioactive materials are transferred from the liquid to the solid phase (chemical precipitates, suspended solids and sludges). The radioactive materials associated with these solids must still be disposed of in a safe manner.

(2) Chemically, the radioactive isotopes react similarly to their stable counterparts. Thus if a process removes the stable element, as with the precipitation of calcium in softening, radiocalcium will be carried down in the same manner. This statement may be expanded further to include substances such as strontium and barium which are

TABLE 1

CHEMICAL CONSTITUENT	DRINKING WATER STANDARDS (2)		MAX^M PERMISSIBLE CONCENTRATION		RADIO ACTIVE FORM
	ppm (mg/l)	μg/ml	μg/ml	μc/ml	
GROUNDS FOR REJECTION					
LEAD (Pb)	0·1	1×10^{-1}			
FLOURIDE (F)	1·5	$1·5 \times 10^{0}$			
ARSENIC (As)	0·05	5×10^{-2}			
SELENIUM (Se)	0·05	5×10^{2}			
CHROMIUM (Cr^{+6})	0·05	5×10^{2}	$5·2 \times 10^{6}$	0·5	Cr^{51}
RECOMMENDED VALUES					
COPPER (Cu)	3·0	3×10^{0}	$2·1 \times 10^{8}$	8×10^{2}	Cu^{64}
			$1·8 \times 10^{6}$	4×10^{3}	F
			$2·1 \times 10^{9}$	1×10^{4}	Fe^{59}
IRON & MANGANESE (Fe + Mn)	0·3	3×10^{-1}	$9·3 \times 10^{9}$	0·2	Mn^{56}
MAGNESIUM (Mg)	125	$1·25 \times 10^{2}$			
ZINC (Zn)	15	$1·5 \times 10^{1}$			
CHLORIDE (Cl)	250	$2·5 \times 10^{2}$	$8·9 \times 10^{2}$	2×10^{3}	Cl^{36}
SULPHATE (SO_4)	250	$0·83 \times 10^{2}$ (as sulphur)	$1·2 \times 10^{-7}$	5×10^{3}	S^{35}
PHENOLIC COMPOUNDS (C_6H_5OH)	0·001	$1·5 \times 10^{4}$	$6·7 \times 10^{4}$	3×10^{3}	C^{14}

QUANTATIVE COMPARISION OF DRINKING WATER STANDARDS AND MAX^m PERMISSIBLE CONCENTRATION OF RADIOISOTOPES IN WATER.

chemically related to calcium - that is a process designed for the removal of calcium will also remove some of the strontium and barium.

(3) The amounts of radioelements that must be considered are exceedingly small. To illustrate this, Table 1 presents a comparison of the permissible levels of specific elements in the US Public Health Service "Drinking Water Standards" (2) with the concentrations indicated for radioisotopes of these elements. It will be seen that the permissible levels from the standpoint of radioactivity are far lower than the concentrations permitted on the basis of the "Drinking Water Standards".

(4) Because the radioactive elements are present in such low concentrations, the usual qualitative and quantitative procedures cannot be used in the identification of these elements. Because these materials are radioactive, however, this characteristic may be utilized to detect, measure and identify them.

4.3. Conventional Processes.

Conventional Water treatment processes include coagulation, settling, filteration, and disinfection. In addition, auxiliary treatment such as aeration, softening, or iron and manganese removal may be included. In the course of the experimental studies carried out at Oak Ridge National Laboratory, Oak Ridge, Tenn., by the Public

TABLE [illegible]. REMOVAL OF I BY AUXILIARY PROCEDURES

DOSAGE				FINAL CHARACTERISTICS			PERCENTAGE REMOVAL.	
TURBIDITY ppm	ALUM ppm	NaOH ppm	Auxiliary Chemical ppm	p^H	ALKALINITY ppm	TURBIDITY ppm	COAGULATION & SETTLING	COAGULATION SETTLING & CENTRIFUGING
103	25·7	38·9	5–15	5·8–7·1	17–68	0–15	64–72	74–78
103	25·7	38·9	3·5	7·1	65	0	38	56
17·3–274	25·7	38·9	0·12–3·8	5·8–7·5	19–74	1–40	42–74	48–76
	123·5		0·013–0·13	6·0–6·9			55–90	
	247		0·13–1·1	6·3			90–92	
	100·64		0·13	6·3			95·5	

TABLE 5. EFFECT OF INCREASED p^H ON REMOVAL OF MIXED FISSION PRODUCTS BY FILTERATION

p^H	PERCENTAGE REMOVAL
2·5	14·6
6·2	63·1
8·6	80·7
10·5	85·1
12·2	88·6

ISOTOPE.	CLAY ADDED P.P.M	COAGULANT ADDED P.P.M.	FINAL pH	PERCENTAGE REMOVAL.
Cs^{137}-Ba^{137}	0	1		0-6
	100	1		35-65
Sr^{89}	0	1-5	6·7-7·8	0-6
	100	0·5-6	6·7-10·7	0-51
Cd^{115}	0	1		40-60
	100	1-5		60-95
Ba^{140}-La^{140}	100	1-6	7·5-8·2	28-84
Sc^{46}	100	1-6	6·5-8·2	66-98
Y^{91}	0	1·5	6·8-7·1	83-93
	100	1-6	7·0-10·2	34-99
Zr^{95}-Nb^{95}	0	1-5		70-98
	100	1		95-99
P^{32}	100	0·5-1·5	6·8-8·8	97-99
Cr^{51}	100	1-6	7·6-8·8	73-98
W^{185}	100	1-6	7·5-8·4	5-91
I^{131}	100	0·5-2	6·9-9·0	0-10
Ce^{144}	0	1-1·5	7·2-7·8	81-94
	100	0·5-2·5	7·0-7·8	85-96
F.P.M. 1	100	1-3	7·2-8·8	61-84
F.P.M.	0	1·5	4·3-10·2	9-71
	100	1-5	4·3-10·2	12-73
F.P.M. 3	0	10	9·9-10·0	4·6

TABLE 4. COAGULATION & SETTLING RESULTS
(JAR TEST STUDIES).

Phosphorus (P^{32}) has been removed from contaminated waters (4, 5), but considerable care had to be given to the coagulation steps (5). Removals in excess of 98 percent have been reported. Iodine (I^{131}), which has found most extensive use in medical therapy and research, cannot be removed by coagulation with alum or iron because the element is in the form of an anion. The addition of small amounts of activated carbon (4), copper sulphate(4), or silver nitrate (4, 6), however, has increased removals of this anion from water to about 96 percent. Some of these results are indicated in the Table 3.

Coagulation will be helpful also in the removal of radioactive material attached to or adsorbed on the natural turbidity found in many surface waters. Since coagulation will remove much of turbidity, the activity attached to it will also be removed. The effect of added turbidity on the removal of certain radioisotopes is illustrated by the data in Table 4.

As might be expected, p^H exerts an appreciable effect on removal. Table 5 shows this effect on the removal of mixed fission products by filteration, and

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in the second experiment, the error was calculated from the variation between duplicated samples.

Health Service, the Engineer Research and Development Laboratory (Fort Belvoir), the Tennessee Valley Authority, and the Oak Ridge Laboratory, the conventional processes that showed promise for removal of radioactive contaminants were evaluated. Nonconventional processes including clay adsorption on metal powders, electrodialysis with permselective ion-exchange membranes, and evaporation were also investigated.

4.4. Coagulation.

Coagulation involves the formation of chemical flocs that adsorb, entrap or otherwise bring together suspended matter, particularly suspended matter that is colloidal. The addition of coagulating chemicals will result also in the precipitation of some of the soluble constituents in the waste as metal hydroxides, generally as hydroxides of aluminium or iron, because alum and iron salts are the most common coagulants.

Coagulation followed by settling has been studied rather extensively at the Oak Ridge National Laboratory, where standardized jar test procedures were used and aluminium sulphate, ferous sulphate, and ferrie chloride were evaluated. To conserve time and effort, statistical procedures were employed in planning and carrying out the experiments. A reexamination and recalculation of an earlier 3^4 factorial experiment, in which the simultaneous effects of turbidity, coagulant doze, p^H, and activity in counts per minutes (cpm) were determined, indicated that a one third replication of the 3^4 experiment would provide equally reliable interpretation of the factor interactions. (3) * Through the use of this technique, it was possible to reduce the number of jar tests from 243 to 54 for each series, or to two-ninths of the original effort. The result of these studies are summarised in Table 2.

Note: * A 3^4 factorial experiment means that four independent variables were investigated at three separate levels. In the original factorial experiment the simultaneous effects of turbidity, coagulant dosage, p^H, and initial radioactivity dosage were evaluated at three levels. In the second factorial experiment,evaluation were made of different coagulants (aluminium sulphate, ferric chloride, and ferrous sulphate), initial radioactivity dosage (at three levels), tap water (variation over a 3-day test period), and coagulant dosages of 1,2, and 6 grains per gallon (gpg) (17, 34 and 103 ppm, respectively). The one third replication means that a third of the total experiment was carried out on each succeeding day of the test until the experiment was completed - that is, for three days. In the original experiment, the individual error was calculated from the variation found in running triplicated samples, while,

TABLE 6.

COAGULANT DOSAGES	PERCENTAGE REMOVAL	
	p^H RANGE 2·5 4·4	p^H RANGE 7·5 8·4
$FeSO_4$		
1 g.p.g	10	75
2 g.p.g	15	84
6 g.p.g	30	98
$FeCl_3$		
1 g.p.g	4	92
2 g.p.g	5	90
6 g.p.g	14	94
Alum		
1 g.p.g	3	74
2 g.p.g	4	91
6 g.p.g	9	92.

EFFECT OF pH ON REMOVAL OF RUTHENIUM.

Table 6 indicates the effect of p^H on the removal of ruthenium by coagulation. Increased chemical dosages do not necessarily result in greatly increased removals of activity. In coagulation of a fission products mixture with ferric chloride, for example, a twenty-fold increase in the coagulant doze (from 10 to 200 ppm) resulted in less than 14 percent increase in removal of radiation materials (from 79.9 to 88.6 percent removal).

The results of the research undertaken by Conard P. Stranb, Roy J. Morton and Oliver R. Placak for the Atomic Energy Commission (Under Contract No. W-7405-Eng-26) at the Oak Ridge National Laboratory (4) require a special mention. In this research the effectiveness of coagulation was studied for removal of a series of individual radioisotopes. Specific concentrations of coagulating chemicals, turbidity, and radioisotopes were added to a given quantity of water and the mixtures were subjected to a 3-minute rapid mix and 27-minute slow mix. After mixing, the floc was allowed to settle for 30 minutes. Samples of the supernatant liquor and sludge were extracted, dried under infrared heating lamps and counted in a Geiger - Mueller end-window counter. A total of 53 series, each of 5 jar tests, were made; 7 series of tests in the study of Ce^{144}, 8 of Y^{91}, 10 of Sr^{89}, 13 of I^{131}, 10 of P^{32} and 5 of iodine dissolver solution (mixed fission products waste).

[illegible]

REMOVAL OF SPECIFIC RADIOISOTOPES BY ALUM & NaOH COAGULATION.

RADIOISOTOPE	DOSAGE		FINAL CHARACTERISTICS			ACTIVITY TOTAL		REMOVALS (%)	
	ALUM ppm	NaOH ppm	pH	ALKALINITY ppm	TURBIDITY ppm	INITIAL million cpm	FINAL million cpm	COAGULATION & SETTLING	COAGULATION, SETTLING & CENTRIFUGING
Ce^{141}	25·7	45·6	7·7	[illegible]	4	1·610	0·1700	82·6	89·1
Y^{91}	25·7	45·6	6·6		6	0·190	0·2000	0	0
	25·7	15·2	6·8	26	1	0·028	0·0180	32·1	35·7
Sr^{89}	25·7	45·6	6·7	32	1	0·820	0·8400	0	0
I^{131}	8·6	12·9	7·5	91	3	0·081	0·0690	1·3	14·8
	17·1	26·0	7·3	115	5	0·094	0·0800	6·3	14·9
P^{32}	17·1	26·0	7·3	71	4	0·159	0·0130	90·2	91·7
	17·1	26·0	[illegible]	60	2	0·152	0·0088	81·1	94·2
	17·1	26·0	7·4	70	2	0·033	0·0022	93·2	93·3
	17·1	26·0	7·5	85	5	1·230	0·0530	87·5	93·9
	8·6	13·0	7·7	77	4	0·115	0·0154	86·5	86·5
	8·6	13·0	7·6	88	2	0·110	0·0088	90·0	92·0
	8·6	13·0	7·6	91	2	0·097	0·0066	93·2	93·2

TABLE 8.

REMOVAL OF SPECIFIC RADIOISOTOPES BY ALUM & Na_2CO_3 COAGULATION.

RADIOISOTOPE	DOSAGE		FINAL CHARACTERISTICS			ACTIVITY TOTAL		REMOVALS (%)	
	ALUM ppm	Na_2CO_3 ppm	pH	ALKALINITY ppm	TURBIDITY	INITIAL (million cpm)	FINAL (million cpm)	COAGULATION & SETTLING	COAGULATION, SETTLING & CENTRIFUGING
Ce^{144}	17·1	203·7	7·8	83	3	1·050	0·092	64·8	[illegible]
	25·7	15·2	7·6	12·6	4	1·610	0·088	73·3	94·5
	25·7	15·2	7·2	12·4	4	0·300	0·057	82·7	81·0
Y^{91}	25·7	15·2	6·8	13	4	0·092	0·006	82·6	93·4
		15·2	7·1	27	0·4	0·030	0·005	93·3	83·3
Sr^{89}	25·7	15·2	7·8	32	3	1·700	1·600	5·8	5·8
	25·7	15·2	6·7	22	2	0·160	0·170	0	0
I^{131}	17·1	26·0	7·1	73	4	0·051	0·038	3·9	25·4
	17·1	25·9	8·7	44	2	0·940	0·980	0	0

The results obtained in routine coagulation studies for the removal of radioactive Ce^{144}, Y^{91}, Sr^{89}, I^{131} and P^{32} are reported under several headings -

(1) Alum and Na OH,
(2) Alum and $Na_2 CO_3$,
(3) Alum and Na OH in the presence of turbidity,
(4) Alum and $Na_2 CO_3$ in the presence of turbidity,
(5) $Fe Cl_3$ and Na OH in the presence of turbidity, and
(6) $Fe Cl_3$ and $Na_2 CO_3$ in the presence of turbidity.

As the condition of tests varied somewhat from run to run all information obtained has been summarised in Tables 7-13. The procedure used in the coagulation studies consisted of the addition of radioisotopes, coagulant and turbidity to tap water. This addition was followed by 3-minute rapid mixing, 27-minute slow mixing, 30 minute settling and centrifuging of the settled samples where indicated. An examination of the data in these Tables will show that:

1. The use of alum or iron with Na OH or $Na_2 CO_3$ in the presence of turbidity always gave greater removals than did the use of the coagulating chemicals alone.
2. The use of $Na_2 CO_3$ as compared with Na OH as a coagulant, with or without added turbidity, resulted in greater removal efficiencies and was particularly advantageous for Y^{91}, as removals

[illegible]

	DOSAGE			FINAL CHARACTERISTICS			ACTIVITY DATA		REMOVAL %	
RADIO ISOTOPE	TURBIDITY ppm	ALUM ppm	NaOH ppm	pH	ALKALINITY ppm	TURBIDITY ppm	INITIAL (micro c/ml)	FINAL (micro c/ml)	COAGULATION & SETTLING	COAGULATION, SETTLING & CENTRIFUGING
Ce^{144}	103	8.6	[illegible]	7.3	89	4	[illegible]	[illegible]	89.1	93.6
	103	25.7	[illegible]	7.0	64	4	[illegible]	0.1600	86.3	90.0
	103	42.8	88.1	7.1	59	4	1.540	0.0780	92.2	94.9
Y^{91}	103	8.6	13.7	6.8	73	20	0.190	0.1600	5.3	15.8
	103	25.7	18.2	6.8	26	3	0.028	0.0120	46.4	57.1
	103	25.7	45.6	6.8	73	20	0.190	0.1600	5.3	15.8
	103	34.2	25.8	6.9	25	2	0.028	0.0300	25.0	0
	103	42.8	38.9	6.8	23	2	0.028	0.0300	17.8	0
	103	42.8	88.1	6.8	69	3	0.190	0.1900	0.0	0
Sr^{89}	103	8.6	13.7	[illegible]	39	15	0.810	0.8300	0.0	0
	103	25.7	45.6	[illegible]	36	2	0.810	0.7500	0.0	1.4
	103	42.8	88.2	[illegible]	35	0	0.810	0.7600	0.0	6.1
I^{131}	34.3 ~~103~~	8.6	12.9	7.4	89	1	0.078	0.0780	0.0	0
	68.6 ~~103~~	8.6	12.9	7.5	91	4	0.079	0.0780	5.1	2.5
	103	8.6	12.9	7.5	103	4	0.095	0.0820	9.4	13.7
	103	17.1	26.0	7.3	107	5	0.100	0.0780	13.0	22.0
	103	25.7	38.9	7.3	113	4	0.098	0.0860	15.3	12.2
P^{32}	103	8.6	18.0	7.4	73	4	0.159	0.0044	94.5	97.3
	103	8.6	15.0	7.3	65	4	0.152	0.0022	94.3	98.6
	103	8.6	13.0	7.5	79	3	0.033	0.0009	96.6	97.3
	17.2 ~~103~~	8.6	13.0	7.5	90	2	0.110	0.0044	92.2	96.0
	17.2 ~~103~~	8.6	14.0	7.8	89	4	0.097	0.0044	95.5	95.3
	51.5 ~~103~~	8.6	15.0	7.5	90	[illegible]	0.110	0.0044	94.0	96.0
	51.5 ~~103~~	8.6	13.0	7.6	90	5	0.097	0.0044	93.2	97.7
	103	8.6	15.0	7.6	89	2	1.230	[illegible]	91.6	97.1
	103	17.1	26.0	7.3	68	6	0.159	0.0044	93.1	95.8
	103	17.1	[illegible]	7.1	61	4	0.152	0.0072	88.4	98.6
	103	17.1	[illegible]	7.4	67	3	0.033	0.0013	97.2	96.0
	103	17.1	[illegible]	7.4	84	2	1.230	0.0176	94.7	[illegible]
	103	25.7	[illegible]	7.2	67	5	0.159	0.0044	94.5	97.3
	103	25.7	[illegible]	6.8	52	5	0.152	0.0022	97.2	98.6
	103	25.7	[illegible]	7.1	61	3	0.033	0.0011	93.2	96.6
	103	25.7	[illegible]	7.3	75	2	1.230	0.0153	96.2	98.8

with Na OH were less than 57 per cent and, with $Na_2 CO_3$, were more than 83 per cent.

3. Chemical coagulation under the conditions reported was unsatisfactory for the removal of Sr^{89} and I^{131}.

4. Removals reported with $Fe Cl_3$ were slightly greater than those with alum, due probably to the broder p^H range for effective coagulation with the iron salt. In the studies with iodine dissolver solution (Table 13) higest removals were obtained following coagulation with lime, alum and sodium silicate.

Removals by Turbidity.

Most surface waters contain variable amounts of turbidity, usually in the form of fine clay or silt particles. Some radioisotopes, either through adsorption, ion exchange or a combination of both, will be removed by this turbidity. If the process is primarily one of adsorption, it may be assumed that the process will follow the Freundlich-type reaction, which is expressed emperically by the exponential relationship:

$$\frac{X}{M} = K\,C^n$$

in which:

$\frac{X}{M}$ is the adsorption per unit weight of adsorbent,

x equals $C_o - C$, which is the amount of material adsorbed, or the initial concentrations of adsorbable material minus final concentration of adsorbable material,

C is the concentration remaining (non-adosrbed fraction),

K and n are constants, depending upon the materials investigated.

This expression is modified here by denoting x, C_o and C as the concentration of activity (or radioisotope) in terms of the counts per minute per millilitre (cpm.per ml.) and by expressing M as the milligram per litre (ppm) of adsorbent (turbidity) added. In the use of this modified expression, the values x, C_o, and C represent only the radioactive fraction, as no consideration is given to stable isotopes present that may be adsorbed. The reported $\frac{x}{M}$ ratios may be considerably lower, therefore, than if an evaluation were made of the total adsorbable concentration, radioactive and stable.

Freundlich - type isotherms for Ce^{144}, Sr^{89} and Y^{91} are shown in Fig. 1, and proportional removals for various turbidity dosages are shown in Fig. 2 for Ce^{144}, Y^{91}, Sr^{89} and iodine dissolver solution. In these studies, a clayey soil of local origion was prepared as a slurry and added to tap water to produce the desired turbidities. The results illustrated in Fig. 1 and 2. are based on removals following 3-minute rapid mixing, 27-minute slow mixing and 30-minute settling.

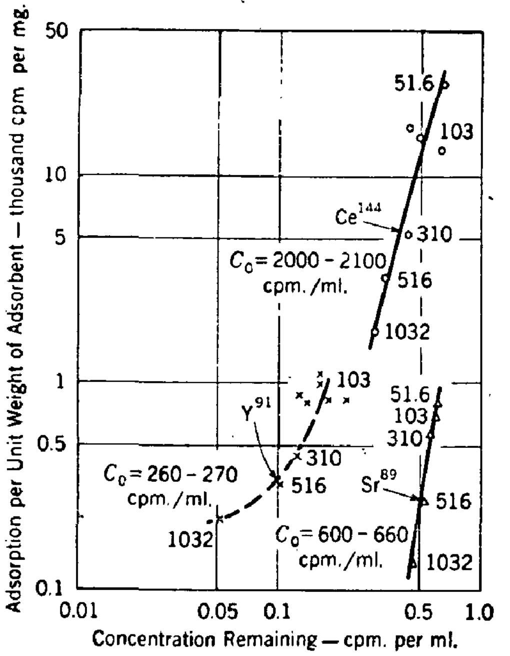

Fig. 1. **Log-Log Plot of Freundlich-Type Isotherms**

The removals of Ce^{144}, Y^{91} *and* Sr^{89} *from solution by clay turbidity are plotted.* C_0 *is the initial concentration of adsorbable material.*

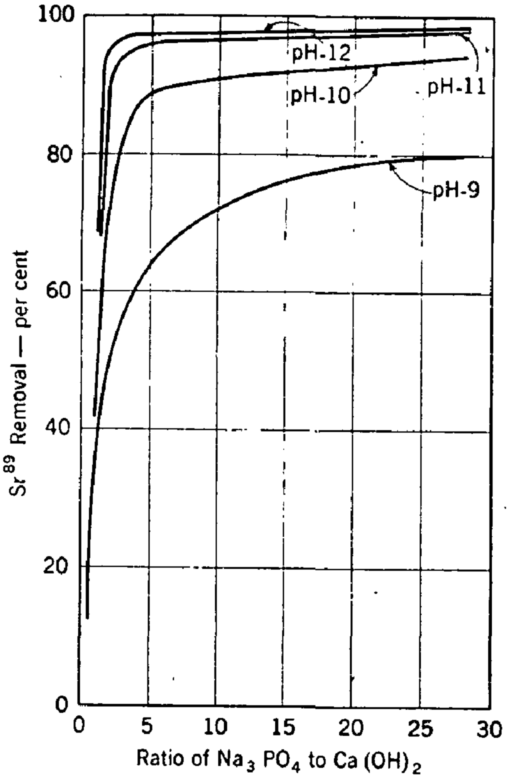

Fig. 3. **Removal of** Sr^{89}

The proportion of Sr^{89} *removed from solution is shown as a function of the ratio of milligrams of* Na_3PO_4 *to milligrams of* $Ca(OH)_2$ *at 9–12 pH.*

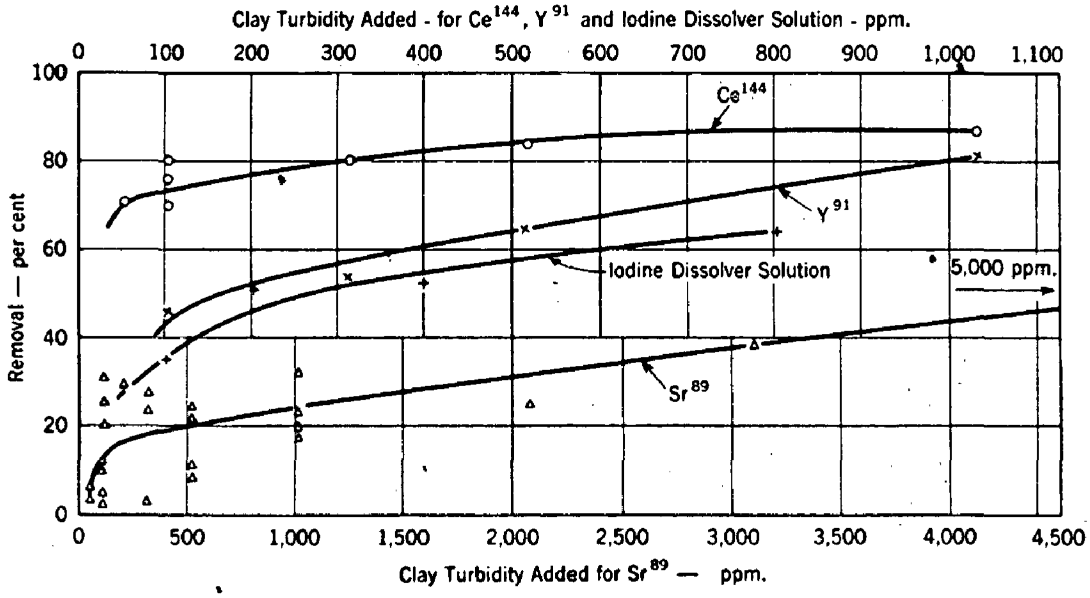

Fig. 2. **Removal of** Ce^{144}, Y^{91}, Sr^{89} **and Iodine Dissolver Solution**

More recent studies have shown that the rate of mix (rapid or slow) affects removal rates greatly. The addition of 100 ppm. of clay to a solution containing Ce^{144} followed by 5-minute rapid and 25-minute slow mix showed a removal of 25.2 per cent by the clay; with 30-minute rapid and no slow mix, a removal of 92.8 percent. The addition of 100 ppm. of clay to a solution containing Y^{91} produced similar results. With slow mix, only 38.8 percent of the Y^{91} was removed; with rapid mix, removal was increased to 93.4 percent. The effect of rapid or slow mix may have been more pronounced in the local clay slurry because it was of a size that settled out during slow mixing, thereby reducing the number of surfaces in contact with the radioactive solution. Rapid mixing overcome this and resulted in much better removals.

The data plotted in Fig. 1 and 2. show that wide differences in removal may be encountered dependent upon the particular isotope involved. Subsequent studies have further shown that the particular absorbent used also has a major effect on removal. Extreme caution should therefore be exercised in interpreting statements that have been made on overall removal of radioactive contaminants by suspended and colloidal matter.

4.5. Sand Filteration.

Except for removal by simple straining or sorption

TABLE 13. REMOVAL OF RADIOACTIVITY FROM [illegible] DISCOVERER SOLUTION.

TURBIDITY			LIME			LIME AND ALUM				LIME, ALUM, & Na_2SiO_3				
DOSAGE	Initial pH	Removal	Dosage	Initial pH	Removal	Dosage Lime	Dosage Alum	Initial pH	Removal	Dosage Lime	Dosage Alum	Na_2SiO_3	Initial pH	Removal
p.p.m		percent	ppm		percent	ppm	ppm		percent	ppm	ppm	ppm		percent
50	7·6	28·4	9	8·8	39·3	51	9	8·8	67·0	0	51	6	[illegible]	[illegible]
100	7·6	34·9	17		43·4	51	17	8·8	70·2	6	51	6	7·6	61·0
200	7·6	50·2	9	9·0	61·2	51	26	8·7	73·5	12	51	6	[illegible]	64·2
400	7·6	52·2	34		65·4	51	34	8·6	76·1	24	51	6	8·[illegible]	72·4
800	7·6	64·1	51	9·0	80·0	51	51	8·4	74·4	51	51	6	8·[illegible]	84·2
						0	51	7·2	56·0					
						6	51	7·4	50·2					
						12	51	7·5	60·5					
						24	51	7·8	62·9					
						51	51	8·4	70·7					

TABLE 14 LABORATORY SAND FILTRATION RESULTS.

ISOTOPE.	INITIAL ACTIVITY cpm/ml	VOLUME PASSED ml	pH OF EFFLUENT	PERCENTAGE REMOVED RANGE	PERCENTAGE REMOVED AVERAGE.
Cs^{137} – Ba^{137}	800	500	8·3	10–70	50
Sr^{89}	2,700	750	8·3	1–13	94
Cd^{115}	1,200	500	8·1	60–90	95
Ba^{140} – La^{140}	1,300	750	7·6	39–99	74
Ce^{146}	1,500	750	8·3	94–99	96
Y^{91}	5,700	750	7·0	84–89	87
Zr^{95} – Nb^{95}	3,400	500	7·8	91–96	93
W^{185}	2,700	750	7·1	3–18	8

by biological life contained in a "schmutz-decke", of slimy layer covering filters, sand filters have not been effective for the removal of radioactive materials (3). Their major function is to remove radioactivity previously incorporated in floc particles during coagulation. Some typical results from single-pass, down flow filteration through sand columns are shown in Table 14.

4.6. Softening.

Softening with lime and soda ash has been studied and, under proper conditions, effective removals of strontium, barium, cadimium, yttrium, scandium, and zirconnium-niobium have been obtained (3, 7). Removal efficiencies of 95-99 percent or higher are possible. For most satisfactory removal of strontium, Hoyt (3) found that excesses of both lime and soda ash were required. His results are summarised in Table 15. The strontium present was reported to be removed by coprecipitation with the calcium carbonate by the mechanism of mixed crystal formation. Data on the removal of stable strontium by municipal water treatment processes, including softening have been reported (8) and are given in Table 16.

As has been indicated above, lime soda softening is fairly effective in removing specific radioelements if relatively high dosages are employed. From the standpoint of emergency water treatment, this process is not generally

TABLE 15.

STRONTIUM REMOVAL BY LIME SODA ASH TREATMENT.

TREATMENT	PERCENTAGE REMOVAL
STOICHIOMETRIC AMOUNTS	75·0
20 ppm Excess lime soda ash	77·0
50 ppm Excess lime soda ash	80·1
100 ppm Excess lime soda ash	85·5
150 ppm Excess lime soda ash	97·5
200 ppm Excess lime soda ash	99·4
300 ppm Excess lime soda ash	[illegible]

TABLE 16.

TYPE OF TREATMENT	PERCENTAGE REMOVAL		NUMBER OF CITIES
	Avg.	Range	
Alum or Ferrous sulphate	12	10 - 31	7
Alum or Ferrous sulphate, plus lime	37	10 - 75	11
Alum or Ferrous sulphate + lime + soda ash	54	10 - 85	5
Alum or Ferrous sulphate, lime and phosphate	[illegible]	10 - [illegible]	[illegible]
Softening only (phosphate, ion exchanger)	[illegible]	69 - [illegible]	[illegible]
None (except [illegible], NH₃)	10		8

STRONTIUM REMOVAL BY CHEMICAL TREATMENT.

applicable because water works serving almost nowhere in India are equipped with a softening plant. Also, exceedingly high dosages would be required to effect substantial reduction of mixed fission products. Other water treatment processes with the exception of ion-exchange, are relatively ineffective in removing strontium, whereas lime-soda softening, under optimum condition, can effect removal in excess of 99 per cent.

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CHAPTER 5.

REMOVAL OF RADIONUCLIDES FROM WATER BY WATER TREATMENT PROCESSES
(PART II)

Mixed Bed Ion Exchange

Mixed Bed Ion Exchange

The main theme of this article has several facets. These may be summarised, however, as the removal of radioactivity by mixed bed ion exchange.

During experiments devoted principally to a study of the removal of mixed fission products from water by utilization of ion exchange, many problems arose including the size of ion exchange units; the flow rate; the effect of radioactivity on the capacity of units; the use of cation resin alone or the need for a mixed bed; the decontamination factors possible with such units; and the discarding of the resins at exhaustion rather than their regeneration.

Because of these questions and the present upsurge in the building of reactors by industries, it has seemed fitting to cover the removal of radioactivity by mixed bed ion exchange in a general way and to try to answer the questions.

5.1 Historical Resumae.

The first article to appear in the literature on the removal of radioactivity by mixed bed ion exchange was by Ayres (1) who used laboratory wastes to which the added

1.5 x 10^6 mixed fission product activity and poured these through a mixed resin. The waste was 0.015 N in acidity and contained 3,000 ppm total solids. A decontamination factor of 10^6 was obtained for a throughout equivalent to 59 gal per cubic ft. of mixed resin. Breakthrough was taken as the first appearance of activity in the effluent.

Launderdale and Emmons (2) investigated the removal of mixed fission products from tap waters using mixed bed ion exchange in conjunction with another bed filled with steel wool, clay, and activated carbon. They were able to pass the equivalent of 119 gal of water per cubic foot of mixed resin before the breakthrough point of 10 counts per minute per millilitre (cpm per millilitre) was reached. The Oak Ridge Tap Water contained 200 ppm total solids, and the p^H of feed was adjusted to 7.5 with sodium hydroxide.

Balcar and associates (3) studied removal of 1.5 x 10^3 cpm per millilitre mixed fission products added to Jersey City tap water, using as theirh breakthrough point 20 cpm per millilitre activity in the effluent. The throughput volumes varied between 230-295 gal per cubic foot of resin depending on the resins used. Jercy City tap water contained 90 ppm total solids. The same authors also studied the removal of Cs^{137}, Sr^{90}, Co^{60}, Ni^{63}, Ru^{106}, Ce^{144}, S^{35} and C^{14} from varieus salt solutions containing 1,000 and 5,000 ppm total solids.

6.2. Tap Water Radioactivity Removal

Intensive investigations have been made by the waste processing group at the Argonne National Laboratory on the use of mixed bed ion exchange for the removal of mixed fission products from tap water.

The three main objects of these investigations were (1) to see what the optimum conditions were for the removal of radioactivity; (2) to discover the capacity of the resins under these conditions; and (3) to find the most economical use of regenerant solutions.

The feed used was tap water to which 1 to 2 year old mixed fission products were added. This tap water contained 300 ppm total solids, 85 ppm total hardness as $Ca\ Co_3$ and had a p^H of 9.0 - 9.5.

Laboratory experiments using a 1½ in column containing 45 cu.in. of mixed resin (30 cu. in. of anion resin and 15 cu. in. of cation resin) indicated that an overall decontamination factor of 10^4 was obtained with neutral tap water which contained 10^5 cpm per milliliter of gross beta-gamma activity. The volume processed was equivalent to 440 gal. per cubic foot of the mixed resin. When a 3:1 ratio of the anion to cation resin was used, the capacity of bed was increased to 630 gal per cubic foot (3).

TABLE [illegible]

INCREMENTAL FEED VOLUME -(l)		AVERAGE ACTIVITY cpm/ml					DECONTAMINATION FACTOR			
THROUGH PRIMARY COLUMN	THROUGH SECONDARY COLUMN.	FEED	PRIMARY COLUMN EFFLUENT.	EFFLUENT THROUGH SECONDARY COLUMN WITH ANION CATION RESIN RATIO OF:			THROUGH PRIMARY MIXED-BED	OVERALL THROUGH SECONDARY COLUMN WITH ANION CATION RESIN RATIO OF:		
				2:1	1:2	CATION ONLY		2:1	1:2	CATION ONLY
FIRST CYCLE										
[illegible]	0·9	1·[illegible] $\times 10^5$	67	1·5	0·8	13·0	1,600	7·3 $\times 10^4$	1·4 $\times 10^5$	[illegible]
9·9	3·3	1·5 $\times 10^5$	7	2·0	1·0	4·2	2·0 $\times 10^4$	7·5 $\times 10^4$	1·5 $\times 10^5$	[illegible]
27·2	9·1	2·0 $\times 10^5$	2	1·2	0·9	3·4	1·0 $\times 10^5$	1·7 $\times 10^5$	2·2 $\times 10^5$	[illegible]
3·0	1·0	2·6 $\times 10^5$	39	1·0	1·0	31·0	6,600	2·6 $\times 10^5$	2·6 $\times$ [illegible]	[illegible]
1·2	0·4	2·6 $\times 10^5$	103	0	0	56·0	2,500	$>10^6$	[illegible]	[illegible]
44·0	14·7						1·4 $\times 10^4$	1·2 $\times 10^5$	[illegible]	[illegible]
SECOND CYCLE (After first Regeneration of Primary Bed)										
4·5	1·0	2·3 $\times 10^5$	460	1·2	2·5	49·0	[illegible]	[illegible]	[illegible]	[illegible]
5·0	1·5	1·7 $\times 10^5$	270	3·7	3·0	8·0	[illegible]	[illegible]	[illegible]	[illegible]
5·0	1·0	1·9 $\times 10^5$	78	2·0	8·0	6·0	[illegible]	[illegible]	[illegible]	[illegible]
18·0	6·0	2·4 $\times 10^5$	22	4·0	2·8	7·3	[illegible]	[illegible]	[illegible]	[illegible]
10·0	3·0	2·8 $\times 10^5$	8	2·3	4·0	15·0	[illegible]	[illegible]	[illegible]	[illegible]
1·0	1·0	2·8 $\times 10^5$	78	2·5	5·5	29·0	[illegible]	[illegible]	[illegible]	[illegible]
43·0	13·5						[illegible]	[illegible]	[illegible]	[illegible]

REMOVAL OF MIXED FISSION PRODUCT ACTIVITY FROM NEUTRAL TAP WATER BY A MIXED BED ION EXCHANGER.

The breakthrough point was the point at which the effluent had a resistivity of 200,000 ohms.

A problem usually arises as to whether it would be practical to use a mixed bed column in the same manner as a roughing filter - that is to remove gross contamination - fallowed with either a second mixed bed or a cation-exchange column alone for polishing the effluent. The results of such experiments are given in Table 1. A 1½ in dia. column which contained 46 cu. in. of mixed resin was used as the primary column. The effluent from this column was then split three ways; one third passing through a small mixed bed column containing a 2:1 ratio of anion to cation resin; one third through a mixed bed having a 1:2 ratio of anion to cation; and the third portion passing through the cation resin only.

Until breakthrough of the total solids the primary column gave an overall decontamination factor of 1.4×10^4 in the first cycle, whereas the two secondary mixed bed column gave an overall decontamination factor 1.2 and 1.9×10^5, respectively. The cation column showed an overall decontamination factor, including the primary mixed bed, of 2.3×10^4.

5.3. Capacity for Radioactivity Removal.

In order to determine the effect of continued reuse of a mixed bed resin for removal of radioactivity,

TABLE 4.

STAGE OF EXPERIMENT	VOLUME INCREMENT gal/cu ft		CUMULATIVE VOLUME gal/cu ft		INTEGRATED DECONTAMINATION FACTORS	
	MIXED BED	DUAL BED	MIXED BED	DUAL BED	MIXED BED	DUAL BED
BEFORE TOTAL SOLIDS BREAKTHROUGH	406	356	406	356	1·4 × 10^3	1·7 × 10^4
AFTER TOTAL SOLIDS BREAKTHROUGH	559	610	965	966	44·0	1·4 × 10^2
	203	203	1,168	1,169	3·7	15·5
	127	127	1,295	1,296	4·0	3·4

COMPARISION OF DUAL AND MIXED BED ION EXCHANGERS FOR REMOVAL OF RADIOACTIVITY.*

* COLUMNS USED WERE 5/8" IN DIAMETER CONTAINING 3 CU IN. CATION AND 6 CU. IN ANION RESIN. THE FEED WAS NATURAL WATER + 2·5 × 10^5 M.F.P. FLOWING AT 2 gpm/cu.ft.

TABLE 2.

RUN NO	FEED pH	VOLUME gal/cu ft	FINAL EFFLUENT FRACTION cpm/ml	INTEGRATED DECONTAMINATION FACTOR
H 20 D	7·0	707	13	8·7 × 10^3
H 30 D	7·0	707	10	1·5 × 10^4
H 32 D	7·0	628	7	3·4 × 10^4
H - 35 C	7·0	628	7	1·4 × 10^4
H 26 D	2·5	393	10	2·0 × 10^4
H 36 D	2·5	314	0	1·0 × 10^5
H 38 D	2·5	414	41	4·0 × 10^4
H 40 D	2·5	393	17	2·8 × 10^4

EXPERIMENTAL MIXED BED [illegible]

a column of larger diameter was required to regenerate the resins effectively. Two 4 in diameter, 52 inches high lucite columns (Fig. 1) were constructed, in to which were measured 1,550 ml cation resin and 3,200 ml of anion resin equivalent to 0.168 cu.ft. of mixed resin. In one column the feed was tap water adjusted to p^H 7.0 and, in the other it was adjusted to p^H 2.5. Typical results are shown in Table 2. It is interesting to note that twice as much capacity was obtained at p^H7, as was obtained at p^H 2.5. The decontamination factors, however, are not as great as at p^H 2.5, undoubtedly because most of the radioactive nuclides would be in the anionic form at the lower p^H, whereas some, such as ziroconium would be colloidal at p^H 7.0. As the anion resin broke through first, probably because of the nitrate ion from nitric acid used to reduce the p^H to 2.5, it is recommended (3) that a 3:1 ratio of anion to cation resin be used in order to increase the capacity of the mixed bed.

5.4. Method of Regeneration.

On the basis of the above capacity for water containing 300 ppm of total solids it can be seen that it would be uneconomical to process only 400-800 gal per cubic foot of mixed resins if regeneration were not practiced - unless, of course, the resins required remote handling because of curie level activity. Regenerating solutions would be radioactive and would require further processing,

SOLUTION	VOLUME ml	VOLUME gal/cu ft	ANION OR CATION REGENERANT FLOW RATE gal/cfm	REGENERANT EFFLUENT pH	REGENERANT EFFLUENT cpm/ml
FIRST REGENERATION					
DISTILLED WATER FOR BACK WASH	750	7·5		1·2	$1 \cdot 1 \times 10^4$
4% SODIUM HYDROXIDE (ANION REGENERANT)	800	8	1·0	2·5	$2 \cdot 4 \times 10^5$
WATER RINSE	500	5		1·2	$8 \cdot 8 \times 10^4$
6N NITRIC ACID (CATION REGENERANT)	170	1·7	0·4	< 1	$4 \cdot 0 \times 10^7$
FIRST WATER RINSE	1,000	10		0·3	$1 \cdot 4 \times 10^6$
SECOND WATER RINSE	500	5		2·2	6×10^3
THIRD WATER RINSE	500	5		3·3	8×10^2
FOURTH WATER RINSE	500	5	— —	3·5	$5 \cdot 2 \times 10^2$
FIFTH WATER RINSE	500	5		7·2	19
	Σ 5,220	Σ 52·2			
SECOND REGENERATION					
BACK WASH	750	7·5		1·9	$8 \cdot 1 \times 10^4$
4% SODIUM HYDROXIDE (ANION REGENERANT)	1,375	13·75	1·0	11·4	$9 \cdot 2 \times 10^4$
DISTILLED WATER RINSE	500	5·0	1·0	11·8	$8 \cdot 2 \times 10^4$
6N NITRIC ACID (CATION REGENERANT)	170	1·7	0·4	0·7	4×10^6
FIRST WATER RINSE	500	5·0	2·0	0·3	4×10^6
SECOND WATER RINSE	500	5·0		1·2	$2 \cdot 9 \times 10^4$
THIRD WATER RINSE	500	5·0		2·8	2×10^3
FOURTH WATER RINSE	500	5·0		4·0	246
FIFTH WATER RINSE	500	5·0		4· 4·5	56
	Σ 5,295	Σ 52·95			

TABLE 3

REGENERATION OF PRIMARY MIXED BED ION-EXCHANGE COLUMN.

NOTE. TOTAL VOLUME OF RESIN WAS 45·3 CU. IN.

so it is essential that these solutions be kept to a minimum volume. Two processes seemed feasible; a two step process, or the simultaneous method of regeneration. In both processes the resins were separated by back washing, but instead of using clean rinse water for this purpose, the water on top of the resin was recirculated so as to use only a small additional amount of water. In the two-step process a 4 percent caustic solution was passed through the entire resin column, thus changing the entire resin column to the hydroxyl form and the cation resin to the sodium form. The liquid in the column was drained to the cation-anion interface and 6 N nitric acid was run on to the cation resin by means of a tube. Several distilled - water ~~resins~~ rinses followed, in each case using a minimum volume. Instead of rinsing until a p^H of 6-7 was obtained, rinsing was discontinued when the effluent had a p^H of 3-4. The results of laboratory experiments are shown in Table 3, and a schematic drawing of apparatus is shown in Figure 2.

In the simultaneous method of regeneration, after the resins are classified by the method just mentioned, the caustic is passed down the anion column, 6N nitric acid is passed upflow through the cation column, and both are removed at the interface. (Fig.3). As the volume of 4 per cent caustic was 9 litres for the 0.168 cu.ft. of mixing resins, and as only 1 litre of 6N nitric acid was required, 8 litre of caustic were passed through the column before any nitric acid was added. The equipment manufacturer recommended that water be passed up flow through the cation bed until it was time to add the nitric acid, but experimental work indicated that that was unnecessary. Again the object was to keep the regenerant solutions to a minimum. In all of these preliminary tests employing the simultaneous method of regeneration, the capacity of the resins was not impaired by these procedures when the experiments were run with inactive solutions.

When using tap water to which radioactivity has been added, it was found that continuous cycles at p^H 7 showed some erratic results. Further investigation indicated that in order to obtain good removal of radioactivity when the feed was at $p^H 7$, it was advisible to add so called a decontaminating step. This was accomplished by passing 6N nitric acid, before regeneration, down flow through the column at the rate of 0.4 gpm per cubic foot. The amount used was equal to that required to cover the resin bed. When the feed is at p^H 2.5 this decontaminating step is required only after every fourth or fifth cycle. The extra step also adds to regenerant volume, and it is therefore recommended that the mixed bed regeneration in the usual manner until a marked decrease of capacity for activity removal appears and, at this time, to employ the decontaminating step. It might also be possible to use less concentrated nitric acid for this particular step

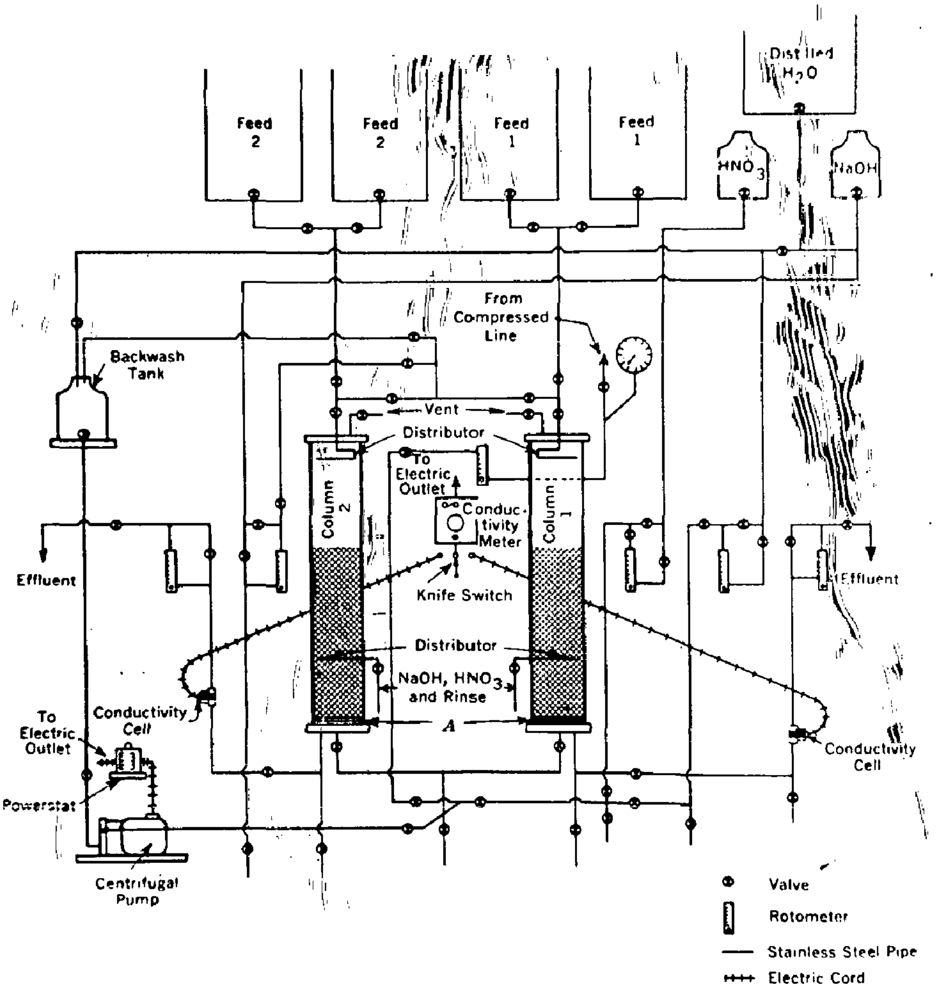

Fig. 4 Pilot Plant Mixed Bed Ion-Exchange Unit Piping Diagram

although 6N nitric acid was found to be optimum for renerating the cation resin (3).

5.5. Efficiency of Regeneration

In a series of 10 laboratory experiments using the two-step regenerating process and neutral tap water containing 10^5 cpm per milliliter mixed fission product activity as feed, the average regeneration efficiency was 70 percent (3). The formula for calculating percentage regeneration efficiency is:

$$\text{Regeneration efficiency} = \frac{\text{Capacity after regeneration}}{\text{Determined initial capacity}} \times 100$$

In a series of 10 experiments using the small 4 in diameter pilot plant columns, considerably better results were obtained by the simultaneous method of regeneration. The new resin bed processed 650 gal of tap water per cubic foot of mixed resins prior to total solids breakthrough. Six cycles using the two-step process showed capacities of 321, 262, 298, 220, 494 and 619 gal per cubic foot, or an average of 57 percent efficiency. Subsequently cycles using the simultaneous method showed capacities of 821, 792, 750, and 780 gal per cubic foot, or an efficiency of 121 per cent.

5.6. Effect of Flow Rate

The manufacturer's recommendation for rate of flow through a mixed bed ion exchange unit is 2 gpm per cubic foot. The same recommendation is given for a cation exchanger, but some work (4) has shown that flow rates as high as 10 gpm per cubic foot do not impair the capacity of a cation exchanger for radioactivity removal. With a mixed bed exchanger (3), however, the capacity was greater at the 2-gpm per cubic foot rate (340 compared to 280 gal per cubic foot), and the integrated decontamination factors were approximately 2 to 5 times greater at the 2-gpm than at the 5 gpm per cubic foot rate. In these experiments tap water adjusted to p^H 2.5 was used, and 10^5 cpm per milliliter mixed fission products were added. It is therefore concluded that, for the best removal of radioactivity by mixed bed ion exchange, the flow rate should not exceed the 2-gpm per cubic foot rate, and a lower rate might be even more desirable.

5.7. p^H, Resistance and Activity

Laboratory wastes from ion - exchange experiments which contained Na, Ca, Fe^{+++}, and Al nitrates equivalent to about 300 ppm calcium carbonate and activity ranging from 0.1 to 2,200 cpm per milliliter, mainly cesium-137, with some mixed fission product activity was passed through a mixed bed column. The column was 2 in. in diameter and contained 0.014 cu.ft. cation resin and 0.028 cu.ft. anion resin. During this run, p^H, resistivity, and activity, of the effluent were noted by analysing a composite effluent after a day's operation. A correlation

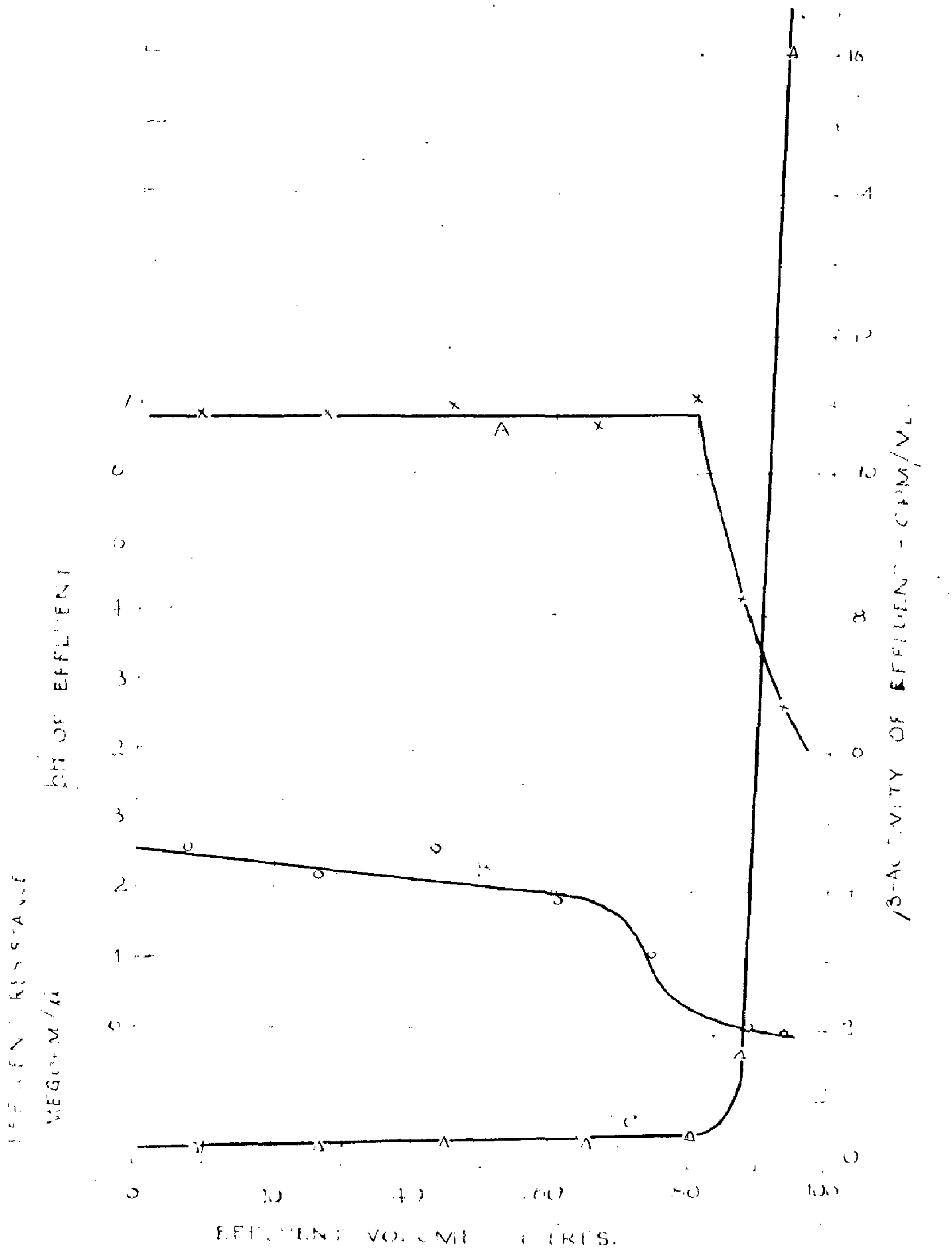
EFFLUENT RESISTANCE MEGOHM/Ω
pH OF EFFLUENT
β-ACTIVITY OF EFFLUENT - CPM/ML
EFFLUENT VOLUME LITRES
0
20
40
60
80
100
1
2
3
4
5
6
7
0
1
2
3
0
2
4
6
8
10
12
14
16

FIG. 4

was noted among these variables at the point of exhaustion of the resin column for activity. Breakthrough point was taken as 5 cpm per milliliter in the effluent; at this point the resistance was at the low point and the p^H dropped from 6.8 to 3.5. It is noted that immediately following this, the activity level rose vertically. The results are shown in Fig. 4.

5.8. Mixed and Dual Bed

As it is easier to regenerate a dual bed than a mixed bed, a study was made of the capacity and decontamination obtained using the two. For the dual bed, 3 cu. in cation resin was used in series with 6 cu.in. anion resin. The same amounts of resin were used in the mixed bed. The feed was neutral tap water to which was added 2.5×10^5 cpm per milliliter mixed fission products. Until breakthrough of total solids, the decontamination factor for the two-bed system was 1.4×10^3 and for the mixed bed, 1.7×10^4 with capacities of 406 and 356 gal per cubic foot, respectively.

If decontamination factors of only 50 are required however, an additional 550 gal per cubic foot of resin can be passed through the two-bed system before regeneration is required. With the mixed bed, a decontamination factor of 140 can be obtained with the passage of 610 gal per cubic foot beyond total solid breakthrough before regeneration is required. These data indicate that there is not much difference in total capacity of resins whether a two-bed or mixed bed system is used, but decontamination factors are approximately 3-10 times greater with the mixed bed system. These data are shown in Table 4.

5.9. Cation & Mixed Bed.

The amount of radioactivity removed by a cation bed is as different from that removed by a mixed bed as is softened water from demineralised water. From a purely chemical point of view, it would appear that, as most radioactive ions are cations, it would be logical to expect a cation exchanger to give almost as good decontamination as a dual or mixed bed system. A cation bed, however, does not give a decontamination factor better than 10-15 unless the only ions present are radioactive ones - still several orders of magnitude under mixed bed results.

It should be noted from Table 1 that the decontamination obtained by the use of a cation resin only on the effluent from a mixed bed resin was considerably less than when the same effluent was passed through a second mixed bed column.

In order to compare the specificity of various ion-exchange materials, a group of four laboratory columns was set up to use the same feed. The columns

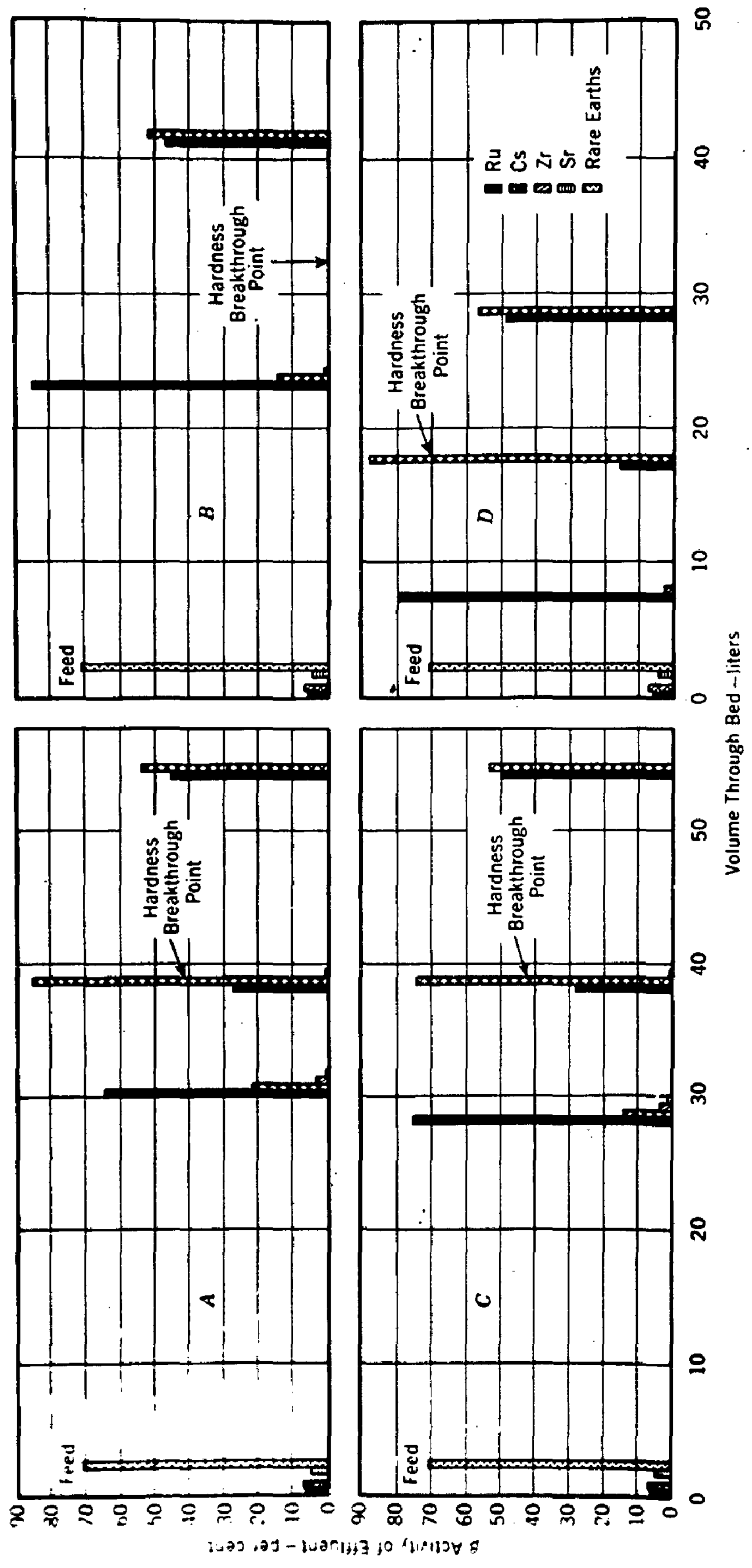

Fig. 5. Fission Product Distribution in Feed and in Effluent From Exchange Columns

contained the cation resins in the hydrogen and sodium forms, a regenerated batch of the hydrogen form of this resin, and a mixed bed composed of a 2:1 ratio of anion to cation resin in the hydroxyl and hydrogen forms respectively. The feed was tap water adjusted to p^H 2.6 to which was added sufficient 2-year old mixed fission product solution to provide about 3 x 10^5 beta cpm per milliliter. Samples of the effluent were taken at frequent intervals, and p^H, total hardness, and beta and alpha activity determined. One effluent sample was analysed for fission product distribution before hardness breakthrough, one at hardness breakthrough, and one after breakthrough.

All of the cation columns contained 3 cu.in. of resin and the mixed bed 1.5 cu.in. of hydrogen form cation and 3.0 cu.in. of hydroxyl form anion resin. The volume passed through the cation columns was 86 litres. Total hardness broke through with both the sodium and hydroxyl forms of the cation resin at 38 litres and between 30-34 litres with the regenerated resin. The integrated decontamination factor at hardness breakthrough was between 21 and 85 for alpha, and from 15 to 18 for beta. The overall integrated decontamination factors varied between 15 and 19 for alpha and between 7.5 and 7.8 for beta.

With the mixed bed columns the decontamination factors of 290 and 7.4 x 10^3 for alpha and beta activity, respectively were obtained prior to the total solids

TABLE 5

PE OF SIN	VOLUME OF RESIN (CUBIC INCHES)	HARDNESS BREAKTHROUGH VOLUME (LITRES)	FRACTIONS	THROUGHPUT VOLUME (LITRES)		ACTIVITY † cpm/ml		DECONTAMINATION FACTOR		INTEGRATED DECONTAMINATION FACTORS	
				INCREMENT	TOTAL	α	β	α	β	α	β
			Feed			290·0	$3·5 \times 10^5$	–	–		
tion H^+	3	38	1-10	30	30	7·0	$1·5 \times 10^4$	41	23	–	–
			11-13	8	38	12·[illegible]	$5·8 \times 10^4$	24	6	36	14·6
			14-32	16	54	31·0	$8·8 \times 10^4$	9	4		–
			33-40	32	86	19·0	$4·9 \times 10^4$	15	7	17	7·8
tion Na^+	3	38	1-9	28	28	12·0	$1·5 \times 10^4$	24	23		
			10-13	10	38	20·0	$3·9 \times 10^4$	15	9	21	16·0
			14-35	16	54	29·0	$1·0 \times 10^5$	10	3	-	-
			36-43	32	86	23·0	$5·1 \times 10^4$	13	7	15	7·5
enerated ation H^+	3	30-34		23	23	2·8	$1·2 \times 10^4$	104	29		
			9-18	18	41	[illegible]	$7·3 \times 10^4$	21	5		
			19-30	45	86	21·0	$5·1 \times 10^4$	14	7	19	7·8
ed bed H^+ & n OH^-		4 ‡	1-3	4	4	0	46		–	>290	$7·4 \times 10^3$
	4·5	12	1-4	1	5	0	192	>290	1,800	–	–
			6-11	12	17	1·7	$1·9 \times 10^4$	171	18	242	27·0
			12-22	11	28	6·7	$7·1 \times 10^4$	43	5		–
			23-3[illegible]	17	45	4·8	$4·2 \times 10^4$	60	8	74	9·2

COMPARISON OF CATION AND MIXED BED RESINS FOR REMOVING ALPHA AND BETA ACTIVITY FROM TAP WATER*

pH OF FEED WAS 2·5.

PROPORTIONAL COUNTER (62 % YIELD FOR BETA, 5[illegible] FOR ALPHA) WEIGHTED AVERAGE ACTIVITY OVER RANGE SHOWN.

TOTAL SOLIDS BREAKTHROUGH.

breakthrough point which correspond to about 4 litres of effluent. For this column which contained half as much cation resin as the columns containing cation resin alone, hardness broke through at 12 litres. After total solids broke through, the activity in the effluent increased markedly. These data are shown in Table 5.

5.10. Activity Distribution in Effluents.

The distribution is shown in Figure 5. The effluent activity discharged from exhausted cation resin columns of all three types was mainly ruthenium and cesium. Cesium was retained fairly well on the column until hardness breakthrough, at which time cesium broke through and from this point was being eluted. Ruthenium was only partly retained on the columns throughout these experiments. A more thorough discussion of the work with cation resin columns was reported by Swape and Anderson (4). The behaviour of the mixed bed column was similar, except that very little activity broke through prior to total solids breakthrough, but after this point was reached, ruthenium was the first element to elute followed by a small amount of cesium.

TABLE 6.

FRACTION NUMBERS	Σ VOLUME		INTEGRATED DECONTAMINATION FACTORS.							
			α ACTIVITY				β - γ ACTIVITY			
			REGENERATED USED RERIN		NEW RESIN		REGENERATED USED RESIN		NEW RESIN	
	ℓ	gal/cu ft.	RUN 1	RUN 2.	RUN 1	RUN 2	RUN 1	RUN 2.	RUN 1	RUN 2.
8	250	392	$3\cdot6 \times 10^{3}$	[illegible]	$6\cdot0 \times 10^{3}$	$1\cdot4 \times 10^{3}$	$6\cdot8 \times 10^{3}$	$7\cdot1 \times 10^{3}$	$5\cdot9 \times 10^{4}$	$1\cdot7 \times 10^{4}$
1-9	300	471	$2\cdot9 \times 10^{3}$	$1\cdot5 \times 10^{3}$	$6\cdot0 \times 10^{3}$	$1\cdot8 \times 10^{3}$	$7\cdot6 \times 10^{3}$	$8\cdot3 \times 10^{3}$	$5\cdot9 \times 10^{4}$	$1\cdot6 \times 10^{4}$
1-10	350	550		528	$1\cdot2 \times 10^{4}$	$1\cdot9 \times 10^{3}$	$8\cdot0 \times 10^{3}$	$9\cdot1 \times 10^{3}$	$3\cdot8 \times 10^{4}$	$1\cdot5 \times 10^{4}$
1-11	400	628		625	$3\cdot0 \times 10^{3}$	$1\cdot9 \times 10^{3}$	$8\cdot6 \times 10^{3}$	$8\cdot3 \times 10^{3}$	$4\cdot2 \times 10^{4}$	$1\cdot5 \times 10^{4}$
1-12	450	707		691	$2\cdot4 \times 10^{3}$	$1\cdot9 \times 10^{3}$	$9\cdot2 \times 10^{3}$	$9\cdot1 \times 10^{3}$	$4\cdot5 \times 10^{4}$	$1\cdot5 \times 10^{4}$
1-13	500	786	$2\cdot3 \times 10^{3}$	760	$2\cdot2 \times 10^{3}$	$1\cdot9 \times 10^{3}$	$1\cdot0 \times 10^{3}$	$1\cdot0 \times 10^{4}$	$4\cdot2 \times 10^{4}$	$1\cdot5 \times 10^{4}$
1-14	550	864	$2\cdot7 \times 10^{3}$	866	$1\cdot8 \times 10^{3}$	$1\cdot6 \times 10^{3}$	$1\cdot1 \times 10^{4}$	$1\cdot1 \times 10^{4}$	$4\cdot2 \times 10^{4}$	$1\cdot5 \times 10^{4}$

INTEGRATED DECONTAMINATION FACTORS OBTAINED ON A MIXED BED ION EXCHANGER USING A LOW-SOLIDS WATER.

NOTE. THE COLUMNS WERE 4" IN DIAMETER AND CONTAINED 0·168 CU. FT. OF MIXED RESINS (2:1 RATIO OF ANION TO CATION RESIN). THE FEED WAS DISTILLED WATER ADJUSTED TO pH [illegible] WITH 4 M NaOH "SPIKED" WITH ~10^{5} C.PM PER MILLILITER FISSION PRODUCT ACTIVITY. THE RATE OF FLOW WAS 2 GPM PER CU. FT.

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CHAPTER 6.

REMOVAL OF RADIONUCLIDES FROM WATER BY WATER TREATMENT PROCESSES
(Part 3).

Miscellaneous Non Conventional Processes

REMOVAL OF RADIONUCLIDES FROM WATER BY WATER TREATMENT PROCESSES

(Part 3)

Non Conventional Processes.

Several non conventional ~~other~~ methods have been studied for the removal of radioactive constituents from water. These are of particular interest as possible methods for emergency treatment or other non-conventional applications. The most promising of these, phosphate coagulation has effected removals of up to 99.9 per cent.

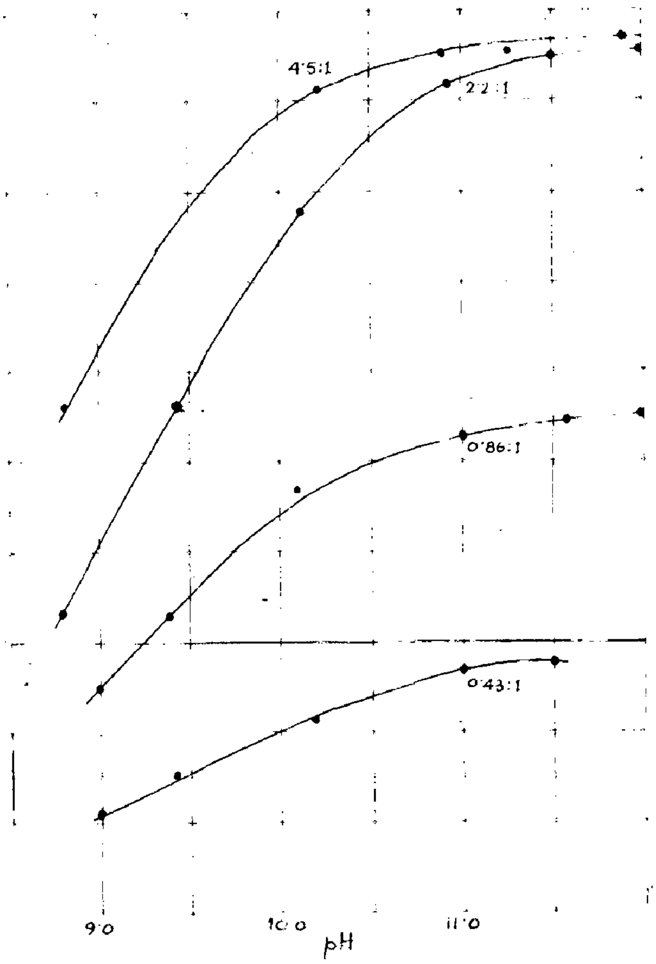
6.1 Phosphate Coagulation.

According to Launderdale, (1), following important conclusions are obtained as a result of experimenting with calcium phosphate -

1. Essentially all the cerium, yttrium, and zinc were removed from solution by phosphate precipitation. The data can probably be extended to include other elements of similar chemical properties.

2. Approximately 10% of the tungesten, 67% of the antimoney, and 95% of the strontiums were removed by the phosphate treatment.

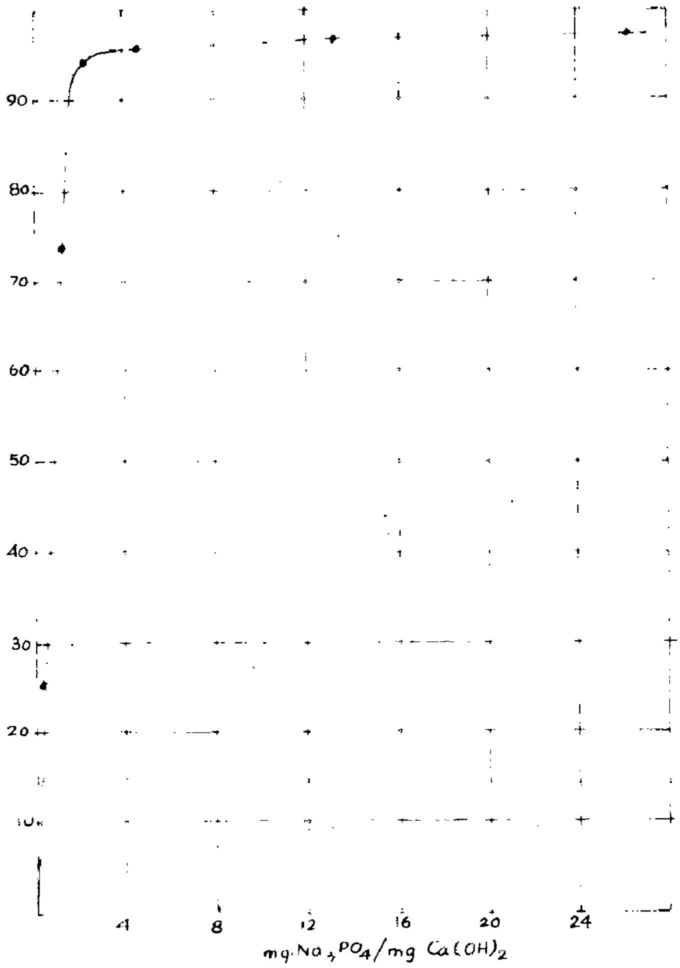
FIGURE 1.



STRONTIUM REMOVED

at Different Ratios Of Sodium Phosphate to Calcium hydroxide

FIGURE 2.



STRONTIUM REMOVED AS FUNCTION OF RATIO OF $Na_3PO_4/Ca(OH)_2$. pH ≥ 11·3.

3. Efficiencies of removal greater than 99%, or decontamination factors greater than 100, have been obtained with a mixture of fission products by using clay in conjunction with the phosphate, followed by sand filteration. The efficiency of the process when used with a mixture of radioactive isotopes will depend on the composition of the mixture.

4. The greatest concentrations of activity in the floc were obtained under conditions of high p^H and with an excess of phosphate. Figures 1 and 2 illustrate the relationship between p^H and the phosphate concentration.

Fig. 1 was obtained when the ratio of phosphate to calcium was kept constant and p^H variable. Six such tests were made using ratios of 0.43, 0.86, 2.16, 4.32, 12.90 and 25.8 mg of sodium phosphate to 1.0 mg of calcium hydroxide. In each case the p^H was adjusted with nitric acid or sodium hydroxide. The p^H has a marked influence on the percent of strontium removed until a p^H of approximately 11.3 is reached. At p^H values of 11.3 little effect due to p^H was observed.

In Figure 2 are shown data obtained by maintaining the p^H of each sample above 11.3 and verying the ratio of sodium phosphate to calcium hydroxide between the values of 0.43 to 1 and 25.8 to 1. The per cent of stron-

TABLE 1.

REMOVAL OF RADIOACTIVE CONTAMINANTS BY METALLIC DUST *

CONTAMINANT	STOCK SOLUTION pH.	INITIAL ACTIVITY cpm/ml.	PERCENTAGE REMOVAL			
			Fe	Zn	Cu	Al
Ru^{106} – Rh^{106}	8·0 - 8·2	1,975 - 3,950	99·6	98·4	93·7	92·8
Y^{91}	7·9 - 8·0	1,610 - 3,230	98·0	98·0		
Zr^{95} – Nb^{95}	7·2 - 8·3	1,210 - 4,470	99·1	97·9	99·1	99·8
P^{32}	7·8 - 8·4	128 - 12,000	99·8	98·1		84·2
I^{131}	6·5 - 8·2	4,288 - 6,689	37·2	45·7	42·1	23·2
$Ce^{141,144}$ – Pr^{144}	7·9 - 8·1	3,915 - 4,491	99·9	99·9	99·5	99·8
Ba^{140} La^{140}	7·9 - 8·2	1,571 - 4,792	92·2	74·5	47·8	30·2
Co^{60}	8·0 - 8·2	1,571 - 4,792	92·2		47·8	30·2
FPM-6 //	7·2 - 8·0	4,730 - 10,375	85·8	76·6	92·1	89·4
FPM-5 #	2·0 - 7·2	2,890 - 4,220	55·5	39·6	49·3	8·2
Cs^{137} – Ba^{137}	8·0	13,085	8·6			

* Tests used filtered samples, 90 min contact and 1000 ppm metal dust.

// FPM-6 was a 20 day old iodine dissolver solution.

\# FPM-5 was a mixed-fission product waste containing mainly Cs^{137} - Ba^{137} & Ru^{106} - Rh^{106}.

TABLE 2.

EFFECT OF ADDED CLAY ON REMOVAL OF RADIOISOTOPES.

RADIOISOTOPE	PERCENTAGE REMOVAL		
	100 PPM CLAY	750 PPM CLAY	5,000 PPM CLAY
Ba^{140} La^{140}	41	58	85
Cd^{115}	3	30	64
Ce^{144}	70 - 80	86	
Cs^{137}	38	87	98·0
Sc^{46}	53	91·7	96·9
Sr^{89}	2 - 12	14 - 22	49 - 52
W^{185}	0	4	49
Y^{91}	22 - 45	56 - 70	93·6
[illegible] Ni	[illegible] 5	99·0	98·0

tium removed increased rapidly as the ratio of phosphate to calcium was increased. A sharp break occurred at a ratio of 2.2 to 1, which corresponds to a 46% excess of phosphate. Above the ratio of 2.2 to 1 only a slight increase in the removal of strontium was noted.

6.2 Electrodialytic Cell.

Studies utilizing an electrodialytic cell with permselective ion-exchange membrances (2) indicated the necessity of removing all colloidal radioelements prior to concentration of soluble radioisotopes in the cell. Passage of various radiocolloids through a molecular filter membrance has shown that the larger colloids such as Zirconium^{95}- Niobium^{95} were removed completely, whereas others, such as Ruthenium^{103} and Cesium^{104}, were removed only in part. Therefore if the dialytic cell is to be effective for the removal of the soluble radioactive materials, the radiocolloids should be removed beforehand.

6.3 Metal Powders etc.

Other methods suggested for the removal of radioactive materials include the use of metal powders and added clay materials. Laboratory jar test studies, using various metal dusts to remove specific radioactive contaminants, were carried out by Lacy (2), and data are shown in Table 1. In general the best results were obtained with the iron dust with removals exceeding 90 per cent, except with fission products mixture iodine, and cesium. From these results Lacy concluded that adsorption was the primary mechanism involved in the removal of the radiocontaminants. Other mechanisms such as metallic displacement may, however, be responsible for some of the removal reported.

Considerable work has been reported (3, 4, 5) on the use of clay materials for the removal of specific radioisotopes from water. Some results are indicated in Table 2. Although, this method is feasible, it involves working with large volumes of clay, both initially and also as contaminated mass of material. Furthermore if combined with chemical coagulation, still larger volumes of radioactively contaminated sludge must be handled for disposal. The adsorption of radioisotopes on clays has its greatest potential value where the geologic and hydrologic conditions are such that the ground itself will retain radioactive materials and reduce the danger that underground water supplies will be contaminated.

6.4 Evaporation.

Evaporation, although is not a municipal water treatment method is included in this discussion because it is the most effective process presently available for concentrating the radioactive materials. Under proper conditions, a condensate very low in activity can be produced. High decontamination factors are possible, but the cost is

[illegible]	NUMBER OF OBSERVATIONS	TREATMENT	PERCENTAGE REMOVAL	
			AVG	RANGE
[illegible]	5	Alum Coagulation	28	23 – 32
[illegible]	5	Ferrous Sulphate Coagulation	32	15 – 44
[illegible]	5	Phosphate Coagulation	[illegible]	66 – [illegible]
[illegible]	4	Excess lime soda ash	[illegible]	[illegible] 78
[illegible]	1	Variable Clay (100 to [illegible] ppm)		39 – [illegible]
[illegible]	20	[illegible] Coagulation	70	43 – 83
Composite	6	Phosphate Coagulation	70	46 – 78
Composite	6	Excess lime Soda ash	37	19 – 48
Composite	[illegible]	Serial Treatment – Clay and Phosphate Coagulation	90	82 93
Composite	6	Serial Treatment – Clay and Excess lime soda ash softening	84	82 – 88

Preliminary Tests

TABLE 4

MOVAL OF MIXED FISSION PRODUCTS† ISOTOPES.

XTURE	TREATMENT PROCESS	PERCENTAGE REMOVAL
M 1	Coagulation and Settling plus clay	[illegible] 84
M [illegible]	Coagulation and Settling	9 71
	Coagulation and Settling plus clay	71
	Coagulation and Settling plus clay	46
	Coagulation and settling plus filteration plus clay	[illegible] 73
M 3	Coagulation and settling	[illegible]
M 4	Coagulation and Settling	[illegible]
	Mixed Cation Anion exchange slurry	[illegible] 99
PM [illegible]	Coagulation and Settling	51 59
	Mixed Cation Anion exchange slurry	38
	Metallic Dust Slurry	8 56
PM 6	Metallic Dust Slurry	17 – 92

ee page for details.

relatively great. Under emergency condition, however, evaporation should prove very satisfactory as a domestic method.

6.5 Removal of Mixed Fission Products.

The previous discussion has centered on the removal of specific radioisotopes from aqueous solutions. With a fission products mixture, or any mixture of radioisotopes, however, the efficiency of removal is a function of the individual radioisotopes comprising the mixture. Removals in laboratory experiments to date have been in the order of 70-80 percent with alum or iron coagulation (3). A series of studies on actual radioactive process waste waters, at the Oak Ridge National Laboratory disclosed that removals up to 90 per cent could be obtained with excess lime-soda ash softening and phosphate coagulation, when clay was added for removal of the cesium in the waste (4). Results of laboratory tests with this waste material are summarised in Table 3. Other experimental data with fission products mixtures are given in Table 4.

6.6. Summary and conclusions.

Unless related to the initial concentration of the radioactive materials present, the percentage removal values cited during these discussions have little significance. It is only when the efficiency of removal is considered in terms of the initial concentration and the maximum permissible concentration values that safety of the treated water for human consumption can be definitely determined. Generally speaking, water treatment processes will not effectively reduce the activity in water to acceptable safe limits except where the initial levels of activity are very low, certainly several orders of magnitude below the 1.0 μc/ml level.

In order to minimise the contamination of water supplies, radioactive materials should be retained as close to their source of production as possible. The practice at present is predominantly tank storage. As has been pointed out, the transfer of activity from the liquid to the solid phase by water treatment processes does not eliminate the problem, since suitable storage or disposal fecilities must be provided for radioactive sludges, filter wash water and other contaminated by-products.

To protect large population centres dependent on surface waters which may become contaminated by radioactive materials plans should be made for auxiliary water sources from possible non-contaminated supplies. For this purpose, existing ground water supplies may be used, other interconnected supply may be piped in, or special provisions may be made for bringing water from the outside. These precautionary measures will be effective only in those areas where the contamination from radioactive materials in low enough so that there will be no hazard from external radiation.

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CHAPTER 7.

Problem's Future Scope in India.

It has been pointed out in Chapter 2 that radioactive fallout may take place anywhere irrespective of the distance of the site of nuclear explosion. A number of factors influence fallout - prevailing winds, rainfall, temperature currents and other climatic, topographic and geographic factors - are a few from the big list. It would be interesting to note that fallout due to nuclear bomb explosions carried out by the Government of the United States of America occured in the United States itself and was recorded. The extensive tests carried out at the Harvard University testify the above statement. It, therefore, is very likely that radioactive fallout might have taken place in India as well. This might have raised the levels of radioactivity in many surface sources of supply. In order to know the effects of nuclear tests on the levels of radioactivity in sources of water supplies a country wide radioassay programme must be organised under the auspices of the Atomic Energy Commission or the Central Public Health Research Institute or any other suitable organisation. The data collected in this manner shall be of great value for all times to come so far as the radiological quality of water is concerned.

Besides fallout, contamination is being caused by the reactors set up by the Government of India at Trombay in the form of reactor wastes. This is limited to Bombay and the surrounding country only, but a glance at the Third Five Year Plan shall show that we are going to establish a nuclear power station either in Delhi, Uttar Pradesh, Punjab or Rajasthan depending upon the recommendations of the high power committee set up for the selection of site for this power station. Not only this but many research establishments dealing with radioactive materials in the fields of medicine, soilstudies, botany and engineering are expected to come up in the recent future. The highlights of the programme are the construction of a plutonium plant, the production of radioisotopes so as to supply the needs of the entire country, the production of large quantities of cobalt 60 for medical therapy and research. During the Third Plan the Atomic Energy Establishment will replace the present temporary laboratories by new ones. These will include in particular, a Modular Laboratory for housing the Divisions which will handle only small quantities of radioactive material and a large Radiological Laboratory for high activity work with hot cells, in which hundred thousand curie amounts can be handled. A plant for the treatment of used fuel elements and the extraction of plutonium is under construction at Trombay and will be completed in the first half of 1963. A plant for treating low active waste will be constructed next to the Canada - India Reactor. These and many more proposals speak of the rapid progress India is making in the Peaceful Uses of Atomic Energy and consequently bring out the importance in the near future of radioactive conditioning of water supplies as an after effect of the contamination caused by the above establishments.

It is also worth pointing out that the industries and private concerns have not yet started making use of Atomic Energy but from the trend of development it is expected that they shall also do so in the recent future. This shall add to the contamination and furthur stress the importance of Radioactive conditioning of water supplies, and accessing the levels of radioactivity in various sources of supply. Because we have a future which has a large expansion in this field, the measurements made at this time shall have a special importance in appreciating the contamination caused in future. The research data included regarding the treatment processes shall also be useful. The data is very exhaustive and gives details regarding a large variety of radioisotopes and their response to various treatment processes and it is hoped that it shall be of use for a good number of years to come. These recommendations should, however, not be contradicted with the economy of the country at any stage.