SOME RESEARCH APPLICATIONS OF RADIOISOTOPE TECHNIQUES AT NCEL

By

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ABSTRACT

Specific applications of radioisotope techniques in three areas of the research program at the U. S. Naval Civil Engineering Laboratory (NCEL) are studied and evaluated. The areas include measurements of the diffusion of isotopically labeled water in concrete; the measurement of small distortions in a hollow sphere; and the measurement of film thicknesses, both liquid and solid, when covering other materials.

The preferred technique for the study of the migration of water in concrete involves the introduction of heavy water (D₂O) into a concrete specimen and the measurement of its concentration at some later time by sectioning the specimen, extracting the water, and measuring the concentration of heavy water therein with a mass spectrometer.

The second study indicates the feasibility of measuring the internal distortions of a hollow concrete sphere to within 0.001 inch by observing variations in counting rate with variations in separation between a source of alpha radiation and a detector installed on opposite sides of the cavity.

The final investigation is concerned with the measurement of thin films, either of liquids as in a thin-film heat exchanger, or of solids such as protective coatings. The preferred method is based on the selective back-scattering of beta rays from the materials under study.

The first two techniques can be applied with a minimum of effort, but the last method would probably require some further development before its full effectiveness is realized.
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INTRODUCTION

This note describes attempts to determine the feasibility of applying radioisotope techniques to problems in three areas of investigation at the U. S. Naval Civil Engineering Laboratory. These problems have arisen during the normal pursuit of the Laboratory's programs and were selected because the experimenter was interested in improving either the amount of information to be learned from the experiment or the ease with which it could be performed. Also, there appeared to be a reasonable expectation that application of the techniques would effect the desired improvements. There is no attempt here to treat the subject exhaustively, but merely to select those problems where modest beginnings could be made.

Each problem is treated separately in the discussion which follows. The first two problems, concerned with measuring the migration of water in concrete and with measuring the deformation of a hollow concrete sphere under external pressure, were found in the Laboratory's Materials Division; the last problem, which considers methods of measuring thin films, was found in the Energy Division. A similar problem, found in the Chemistry Division, of measuring the thickness of protective coatings is included as an integral part of the problem of measuring thin films.

The study is thus divided into three parts. Each part, which is devoted to a specific problem, starts off with a summary. This is followed by some preliminary remarks, which are followed, in turn, by a discussion of physical arrangements, principles of operation, and limitations of the applicable methods. There is a brief discussion of some of the equipment, but no real attempt has been made to introduce actual working descriptions.

PART I. MEASUREMENT OF THE MOVEMENT OF WATER IN CONCRETE

Summary:

In Part I an examination is made of the feasibility of using isotopically labeled water, $D_2O$, $H_2O^+$, and $H_2O^-$, for the study and measurement of the migration or diffusion of water through concrete at any time after it has been placed in the forms and the labeled water inserted in the aggregate in some known distribution. Analysis is made by taking sections of the concrete, extracting the water, and measuring the ratio of the amount of labeled water to normal water with a mass spectrometer. Expected mass spectra are tabulated for different concentrations of the labeled water.

The method described appears to be effective, to make possible an important study of the behavior of concrete not otherwise as easily attainable, and to be capable of extension to other related investigations. Infrared spectroscopy also offers possibilities.
INTRODUCTION

The application of "tagged" atoms to research into the behavior and properties of concrete has been briefly described in a study made by Tracerlab, Incorporated in 1959, and by Leonard B. Gardner in 1964. Both of these surveys considered the use of radioactive hydrogen (tritium) for the tagging of water used in the concrete studies. Detection of the tritium was by Geiger counters, scintillation counters, or similar devices. The purpose of this study is to investigate the possibility of using water tagged either with a heavy isotope of hydrogen (deuterium) or oxygen, but isotopes that are not radioactive (so-called stable isotopes) which are detected by a mass spectrometer. The use of radioactive tracers require care in handling the materials and conducting the experiments because of hazards to health and safety, whereas these hazards are virtually nonexistent when using the stable isotopes. The increase in ease of doing a given experiment when using stable isotopes for labeling is therefore evident, provided the ultimate objectives of the experiment may be realized with their use. Stable isotopes are frequently cheaper than corresponding radioisotopes, and their use in experiments does not entail the extensive licensing requirements of the latter.

PROCEDURE AND ANALYSIS

Use of Hydrogen Isotopes

Two types of experiments are contemplated. In the first the problem is to determine the extent of any water migration in concrete. During the casting of an experimental piece, and while the mix is still partially fluid, the experimenter can place small samples of tagged water (microcuries of water containing tritium, or 100 mg or more of water containing deuterium) at various desired locations throughout the mix and establish an arbitrary distribution of tagged water in the whole piece. Then after any desired lapse of time (presumably after initial set) the piece may be sectioned and samples taken at appropriate locations for measuring the concentration of tagged water. If tritium is used, the sample is exposed to or placed in a counter, and the concentration of tagged water determined from the count rate. If deuterated water or another form of nonradioactive heavy water is used, the determination must be made by some mass-sensitive device. In this way, any migration can be followed through the piece. In both cases, it would seem desirable to extract the water from the sample for measurement.

The second type of experiment considered is one involving the transmission of water vapor or other liquids or gases through a piece of concrete. The procedure in the past has been to cement the sample of concrete into a container so that the sample forms a barrier between the bottom of the container and the top, which is usually exposed to the ambient atmosphere. One then introduces the proper atmosphere (frequently water with enough air to equalize the pressure) into the bottom of the container and measures its loss through the concrete sample. If one is interested in the movement of water through the concrete, the gross amount can be found by weighing.
If exchange occurs between the water chemically bound in the concrete and the vapor which is diffusing through the concrete, this simple procedure will be unable to detect it. The use of tagged water, however, appears to make the verification or denial of this hypothesis possible, and to give some quantitative estimates of the amount of diffusion. In this case, the atmosphere at both the top and bottom of the container must be controlled and enclosed so that samples may be withdrawn from time to time for analysis. The extension of these concepts to atmospheres other than water seems straightforward, and if the age of the sample of concrete is of no importance the same experimental arrangement would serve for several different atmospheres.

Examination of this method in some detail is the main emphasis in this investigation, together with some findings as to available analyzing equipment and its costs.

There are several classes of mass spectrometers commercially available, discussed later on in this note. Some operate on direct-current principles while others use radiofrequency systems. There are at least two classes of DC spectrometers, those with superimposed electric and magnetic fields, and those where the electric and magnetic fields are separated. The last type will be selected here as an example, but the others can serve equally well.

The mass spectrometer in this class would be supplied with water vapor extracted from the samples of concrete. From this vapor it would form ions, and then sort these ions according to mass. The output of the mass spectrometer would be proportional to the current of ions to the detector. The method of sorting the ions by mass is accomplished with an electric and a magnetic field; the first accelerates the ions from rest to a high velocity, and the magnetic field produces a curved trajectory whose radius of curvature varies directly with the mass of the ion. Consequently, in order to measure the current of an ion of predetermined mass, both the electric field and the magnetic field must have proper values. With a simple instrument of this class, the mass of the singly charged ion is given by the equation

$$M = \frac{a}{B^2} \frac{V}{V}$$

where $M$ is the ionic mass, $V$ is the potential drop of the electric field, $B$ is the magnetic field intensity, and $a$ is a constant depending on the radius of curvature of the instrument, other design factors, and various physical constants.

The easiest way to create the ions necessary for the analysis is to bombard the molecules of vapor with an electron beam. Control of the energy and number of these electrons is desirable and is usually provided in mass spectrometers. In the instant case, electrons of sufficient energy on impact with molecules of $\text{H}_2\text{O}$ will produce positive ions with five different masses, as shown in Table 1. Negative ions are not normally detected in a mass spectrometer.
### Table I. Ion Masses for H₂O Bombardment

<table>
<thead>
<tr>
<th>Ion</th>
<th>Mass</th>
<th>Relative Intensity (very approximate)</th>
</tr>
</thead>
<tbody>
<tr>
<td>H⁺</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>H₂⁺</td>
<td>2</td>
<td>0-2</td>
</tr>
<tr>
<td>O⁺</td>
<td>16</td>
<td>1</td>
</tr>
<tr>
<td>OH⁺</td>
<td>17</td>
<td>10</td>
</tr>
<tr>
<td>O₂H⁺</td>
<td>18</td>
<td>100</td>
</tr>
</tbody>
</table>

Similarly if D₂O is the molecule bombarded, the corresponding masses will be those shown in Table II.

### Table II. Ion Masses for D₂O Bombardment

<table>
<thead>
<tr>
<th>Ion</th>
<th>Mass</th>
<th>Relative Intensity (very approximate)</th>
</tr>
</thead>
<tbody>
<tr>
<td>D⁺</td>
<td>2</td>
<td>1</td>
</tr>
<tr>
<td>D₂⁺</td>
<td>4</td>
<td>0-2</td>
</tr>
<tr>
<td>O⁺</td>
<td>16</td>
<td>1</td>
</tr>
<tr>
<td>O₂D⁺</td>
<td>18</td>
<td>10</td>
</tr>
<tr>
<td>O₂₂⁺</td>
<td>20</td>
<td>100</td>
</tr>
</tbody>
</table>

The relative intensities in these tables are crude estimates of the relative ion currents to be found in a mass spectrometer for each particle, all other conditions remaining the same. In practice, in any given instrument they should be measured and the experimental values used.

If the sample to be analyzed contains both light- and heavy-water vapor, all the masses listed in both tables will be present, and the two sets of intensities will appear in direct proportion to the concentration of the respective particles at the point of ion formation. However, it is clear that several masses simultaneously occur in both tables, so that the measured intensities will be complicated by the superposition of these intensities arising from each of the primary, or parent, molecules. For instance, if α is the fraction of D₂O molecules present in the source, the remainder being H₂O molecules, the number of ions of mass 18 produced from the H₂O will be proportional to 100(1 - α), and the number of ions of the same mass produced from the D₂O will be proportional to 10α, with the same
factor of proportionality. The total intensity registered by the mass spectrometer for mass 18 will be proportional to the sum of these two quantities, 100(1 - 0.9α). With this introduction we can easily calculate the percentage contribution from each molecule to each mass intensity. This is shown in Table A-1 in the Appendix. The relative yield shown in the last column would be the expected mass spectrum of a sample where the fraction, α, of the molecules are D₂O.

The concentration, α, of D₂O in the original sample is the quantity sought and it may, in general, be quite small. There are only two masses, 4 and 20, in this example which would depend only on the D₂O concentration, since the intensities arising from H₂O for the other masses would completely obscure any contributions from D₂O. In this case, the concentrations of D₂O could be obtained from the ratio, R₂₀, say, of the yield of mass 20 to the yield of mass 18. Then

\[ \alpha = R_{20} \]

and as a check, from the yield of mass 4,

\[ \alpha = 0.5R_4 \]

where R₄ is the ratio of the yield of mass 4 to mass 18. Alternatively, the ratio \( R'_{20} \) of the yield of mass 20 to mass 17 will give a more sensitive evaluation of \( \alpha \):

\[ \alpha = \frac{R_{20}}{R'_{20} + 10} \]

Greater accuracy in the determination of the concentration \( \alpha \) may be obtained by using all the yields properly normalized, and the expressions listed in Table A-1 with a least-squares fitting procedure, provided \( \alpha \) is not too small. If \( \alpha \) is small (\( \leq 0.01 \)), the simpler methods of calculation will probably give as close results as the more complicated calculations. It would appear possible to measure heavy-water concentrations of perhaps \( \alpha \approx 10^{-5} \) molar percent of the sample.

There are some sources of error in the method outlined above. The background gases in mass spectrometers can contribute to the measured ionic yields. For instance, air as a background gas will contribute to the intensity of the ion of mass 16 by the breakup of O₂, CO₂, and CO under electron impact. Most mass spectrometers have hydrocarbon vapors which are difficult to pump out or are constantly supplied by internal leaks. These release hydrogen ions, both H⁺ and H₂⁺, under electron bombardment. The intensity at mass 20 can be disturbed by doubly charged argon (A''') or neon, both found as components of air. The former can be controlled by the use of
electrons with energies less than about 40 ev, and the latter occurs in such low concentrations that it rarely causes any trouble. Consequently, for high accuracy, it is necessary to evaluate these background ions so that their effects can be carefully eliminated. Fortunately, for low values of \( \alpha \), the mass 4 to 20 positions are quite clear, and this is the region where measurements are most effective. The naturally occurring isotopes of oxygen should not cause any difficulty since they would not generally be in the form of water.

Use of Oxygen Isotopes

Instead of using tagged hydrogen to label the water, it is equally possible to use tagged oxygen. Oxygen has three isotopes, \(^{16}O\), \(^{17}O\) (occurring naturally about 4 parts in 10,000), and \(^{18}O\) (occurring naturally about 1 part in 500), which may be used to label water.

The analysis may be carried forward in the same way as for deuterium. The expected ion intensities are given in Table A-11 in the Appendix.

Of the three possibilities, \( D_2O \), \( H_2O^{17} \), and \( H_2O^{18} \), the first and last are preferable since each gives two ions which fall in clear positions in the mass spectrometer. The other gives only one, the parent ion \( H_2O^{17} \) at mass 19. One may therefore expect greater accuracy in the determination of concentrations by using either \( D_2O \) or \( H_2O^{18} \).

Of the two which seem better, \( D_2O \) or \( H_2O^{18} \), there seems to be little choice as far as the measurements are concerned. The advantage of \( H_2O^{18} \) is that the two mass numbers 19 and 20 are both in the high mass region of the mass spectrum and are in greater yield, particularly mass 19, than the mass 4 ion from \( D_2O \), for the same concentration of each molecule.

Figure 1 shows theoretical mass spectra which may be expected in the analysis of mixtures of \( H_2O \) and \( D_2O \) where \( \alpha \) represents the molar fraction of \( D_2O \) in the complete sample. The bars represent the yields of the various ions (on a logarithmic scale for purposes of illustration) as if the mass spectrometer were adjusted to sweep either its magnetic field or its accelerating electric field continuously from the lower masses to the higher. If pure \( H_2O \) were being examined, the bar graph in the upper left-hand corner of the figure would be applicable. There would be no peak at either mass 4 or mass 20. For pure \( D_2O \), a spectrum similar to that shown in the lower left-hand corner would be expected. In this case, there would be no peak at either mass 1 or mass 17. In practice, however, unless the sensitivity of the instrument were greatly reduced, there probably would be small peaks at both of these positions arising from the background gases in the instrument, one important constituent being water vapor.
Evaluation and Relative Analyses

The applications of these procedures to the study of the diffusion and migration of water in concrete may introduce other complications. In most of the regions where the D2O is found, its concentration would probably be low, the bulk of the water being the lighter H2O. Under these conditions the exchange of deuterium and hydrogen may take place. That is, there may be an appreciable concentration of HDO molecules through the interchange of H and O between two neighboring molecules, one D2O and the other H2O. The introduction of this third molecule into the sample being analyzed would augment the yields at masses 1 and 17, produce two new peaks at masses 3 and 19, and diminish the peaks at masses 4 and 20. This exchange would tend to reduce $\alpha$ below its proper value. On the other hand, a deuterium atom might be passed from molecule to molecule and reach distances farther from its starting point than the rest of the molecule to which it was originally attached, thereby tending to increase the apparent motion of the D2O. This problem is common to hydrogen labeling, including the use of radioactive tritium. The reactions require a water-water vapor interface, high temperatures, or some other form of molecular activation and should cause little trouble in these analyses. This potential source of error would be completely avoided by the use of an isotope of oxygen such as $^{17}$O or $^{18}$O as a constituent of the labeled water.

The expected mass spectra for all three types of water mixed with H2O are shown in Table A-III for various values of the concentration parameter, $\alpha$. The yields were calculated from the intensity values listed in Tables I and II, but for any experimental measurements, the intensities in Tables I and II should be measured and Table A-III recalculated.

This method of analyzing water has been used extensively in other laboratories and in industry. Its feasibility for use in the problems outlined above seems assured. It seems rather clear that with an experimental arrangement similar to that described above, there are several new types of investigations of the properties of concrete which could be undertaken with every expectation of success.

Use of Infrared Spectroscopy

If deuterated water is used, there is another method of determining its concentration in light water through measurements of infrared absorption. This method is considerably simpler than the mass spectrometric method if sample sizes are sufficiently large ($> 0.1$ gram) and the deuterium concentration is not too low ($< 10\%$). Equipment suitable for such measurements is available at NCEL in the Chemistry Division. The relatively large sample required, however, poses difficulties as compared to the microgram to milligram requirements for a mass spectrometer.
THE MASS SPECTROMETER

A mass spectrometer suitable for general analytical determinations should suffice for the experiments considered here. It is thought desirable that peaks be resolved to at least mass 100 so that application to other types of analysis, such as hydrocarbon measurements, would be possible. It should also be rugged enough to be easily moved, and easy to adjust. Most commercial units considered would satisfy the second set of requirements, but several models on the market cannot resolve the larger masses.

Among those with limited mass range is the Omegatron, a radio-frequency type of spectrometer made by Sylvania for $3,000 to $4,000. It would require additional equipment in the form of a vacuum pumping system and possibly a large magnet. The manufacturer's descriptions indicate that it would have difficulty resolving argon (mass 40) from CO₂ (mass 44). The Aerovac Corporation also makes an instrument with rather low mass resolution and limited sensitivity.

The Bendix Corporation produces a spectrometer for about $10,000, but its properties have not been determined. It uses crossed and coextensive electric and magnetic fields.

VEECO and the General Electric Company also make satisfactory instruments. The VEECO spectrometer costs about $8,000, but needs a vacuum system and a sample system. These are DC-type mass analyzers with separated electric and magnetic fields.

Varian Associates and Ulteck make quadrupole, RF-type spectrometers, the former charging from $10,000 to $15,000 per unit.

There are two foreign firms which also make satisfactory mass spectrometers, but their prices are not known at this time. AEI makes a high-grade instrument which is a complete unit, and Atlas Werke makes a quadrupole instrument.

At the request of the Laboratory, three companies have provided prices and literature on instruments capable of analyzing the samples of mixed waters described above. The Consolidated Electrodynamics Corporation quoted a price of $34,000 for a cycloidal type spectrometer for petroleum and residual gas analysis as a complete unit. They did not believe, however, that it is suited to the type of analysis considered here.

Alpha Scientific Laboratories, Inc., manufactures an instrument which separates masses up to 88. The principle of its operation is similar to that of the cyclotron. It comes complete with tube, magnet, and electronics for $1,700, but requires a vacuum system, a sample system, and an output electrometer-amplifier and recorder, if permanent records are to be obtained. The vacuum system and detector system are already available at the Laboratory, but a suitable system for introducing the sample to be analyzed would have to be constructed.
Machlett Laboratories, Inc., manufactures a mass spectrometer tube only, which is capable of analyses up to mass 60. All the peripheral equipment needed to make an operative instrument would have to be obtained elsewhere. This would include a sample system, a vacuum system, and the necessary electronic circuitry. The price of the tube only is $425. The operating principle is one of sorting a beam with respect to time by a radio-frequency oscillator so that no magnet is needed. If an operating unit is assembled, it will probably be approximately competitive in cost with the unit put out by Alpha Scientific Laboratories.

None of the firms responding make measurements of D<sub>2</sub>O-H<sub>2</sub>O concentrations on a commercial basis. This problem of contracting for the analyses with agencies already equipped to perform them was pursued further. Inquiries were made at the Argonne National Laboratory where it is known that heavy-water analyses are made. They reported, however, that their analytical procedures could only take care of small concentrations of light water in heavy water (from 5% down). No other leads were found to any agency that routinely makes analyses in the range of 5% to 95% heavy-water concentrations. One is left with the conclusion, therefore, that the analyses will have to be carried out at NCEL if the experiments are to be performed.

PART 2. MEASUREMENT OF THE DEFORMATION OF A HOLLOW SPHERE

Summary:

The application of radioisotopes to the measurement of small structural displacements in this Part is concerned with measuring the deformations of a hollow sphere when exposed to external pressure. The method employs a small source of alpha radiation and a solid-state detector mounted on opposite sides of the spherical cavity in such a way that deformations of the surface are registered as changes in the counting rate of the detector. The method is found to be feasible for displacement measurements of about 0.001 inch, either positive or negative.

INTRODUCTION

The Materials Division at NCEL has encountered a problem of measuring the deformation of a hollow concrete sphere when under stress produced by an external pressure. Mechanical devices, like strain gages, have been subject to malfunctioning, and there is always the uncertainty of whether a method which involves moving parts is performing properly. The use of alpha rays as a means of measuring small displacements in a volume with limited access appears attractive because the necessary equipment which must be introduced into the cavity is minimal and there are no moving parts. Signals from the detector can be brought to the outside conveniently since only two small wires are required.
PRINCIPLES OF OPERATION

The use of alpha particles for measuring the thickness of materials where the density is known, or the density where the thickness is known, has been recognized for some time. One of the principal developments which contributed to the feasibility of such a device was the discovery of the gold-plated silicon detector. These detectors can be made comparatively small (0.5 inch square by 1/8 inch thick or smaller) and can respond to the impact of alpha particles by emitting pulses on the output electrodes which are proportional to the amount of energy expended by the particle in the detector. These pulses can be amplified and counted with standard circuits. If, therefore, one brings such a detector close to a source of alpha particles, the number captured in the detector per second may easily be determined. Since about 1 inch of air is usually adequate to stop almost all of the alpha particles emitted by most substances, the counting rate registered can be used to measure either the distance between the source of radiation and the detector or the amount of absorbing gas (pressure) between them.

Every alpha emitter ejects alpha particles with a sharp, well-defined energy characteristic of the emitter. This energy may be used up either in emerging from the matrix comprising the source of radiation, in the material intervening between the source and the detector, or in the body of the detector itself. If, therefore, one uses a "thick" source (such as a piece of uranium 1/16 inch thick or more), alpha particles emerge from the surface with all possible energies up to the energy of ejection. The number of particles with any prescribed energy between zero and the energy of ejection is approximately the same. Because of this constancy of numbers with energy, there is a corresponding linear relationship between the number of alpha particles with penetration distance from the source in an absorber (such as air), so that the counting rate of the detector will be simply related to the separation from the source.

SOME DESIGN CONSIDERATIONS

In order to exploit this principle for the measurement of displacement, one would expect rather simple design requirements. There are two isotopes which are suitable as sources and which are readily available. Plutonium-238 is preferred in the references to give a higher counting rate for any given setting, but natural uranium may also be used, and is much cheaper.

If the source were attached to one side of the inner volume of the hollow sphere, and the solid-state detector to a diametrically opposite point so that the two are parallel and spaced 1/4 to 1/2 inch from each other, the arrangement would approximate those of the references, and changes in the counting rate should be approximately proportional to changes in the separation of the points of support. There would be substantially no change in counting rate with lateral displacements.
With such an arrangement, one may expect to be able to detect changes in counting rates for changes in gas density of about \(6 \mu g/cm^2\), which, translated into air at normal temperature and pressure, corresponds to a displacement of 0.00125 inch. If deformations of this order are to be measured, however, care must be taken that the gas pressure surrounding the device is kept constant. Variations of atmospheric pressure, which might affect performance if the void communicates with the outside, can be rather simply eliminated by applying suitable corrections. The necessary electronic circuitry for the detecting system is available at NCEL. The gold-plated silicon crystal detector, however, may not be as readily available. The articles used in the references were homemade, but in the intervening period of time, one would expect them to be quasi-standard items available either from stock or by special order.

PART 3. MEASUREMENT OF LIQUID FILMS AND PROTECTIVE COATINGS

Summary:

This study examines the feasibility of using radioisotope techniques for measuring liquid film thicknesses in a tubular test section with a counterflow of gas and vapor, where the liquid flows in a film on the interior of the test section with thicknesses ranging from 0.030 to 0.000 inch. A method of using backscattered beta radiation with the detector and source adjacent and located in the vapor phase seems to offer the best possibility of making these measurements. The method appears to offer distinct advantages over more cumbersome and expensive optical methods but does not give detailed information about the film shape in small regions. Experimental investigation of the method is suggested.

INTRODUCTION

The measurement of thin films of liquids flowing down tubes or over baffles of varying shapes is important for the understanding of the fluid dynamics, which in turn is important in engineering problems of heat exchangers. These questions are assuming increased importance with the constantly increasing power densities used or contemplated in heat-generating installations such as power stations and desalting operations for seawater. The problem of heat transfer and removal has been recognized for many years, and its investigation is being actively pursued today.

Radioisotope techniques have been used in these studies (see Reference 6 for an extensive bibliography) but with only limited success. The most obvious method was to load the liquid with a radioactive material in solution and then scan the film with a suitable counter. If the film is flowing down the inside of a small tube, this method places rather severe size restrictions on the detector. Also, if the interior of the tube is carrying a counterflow of gas and vapor, the deposition of liquid on the detector will introduce absorption errors.
A second technique was to place a source of radiation on one side of a test section with an internal film, and measure the transmission of radiation through the complete system by a detector on the other side of the tube. This would give an average film thickness over the cross section of the test section and over a portion of its length, depending on the size of the detector. The method, however, would be difficult to use for water flowing down a steel pipe, for instance, because most of the absorption would be in the steel walls, and changes due to varying average film thickness would be comparatively small. Such techniques have been used to measure void fractions in columns of liquids, and as liquid-level indicators.

Three other methods which are not subject to some of these limitations, and offer possible approaches to the problem will be described in what follows. Two and perhaps all of these methods have been used in the past, but they seem to be the best suited to the problem at hand. The direction of flow of the gas and vapor through the tube is of no importance, of course.

PROPOSED METHODS

The Experimental Arrangement

The essentials of the experimental arrangement are shown in Figure 2, which forms the basis of the discussion which follows. A tube, preferably 1/2 inch in diameter, is connected to the bottom of a reservoir in which the liquid level is maintained at such a value as to produce the film thickness desired. The tube is about 1 foot long and does not dip into the accumulation of liquid underneath. A countercurrent of gas and vapor is maintained in the tube while the liquid is flowing. Usually air and water are considered for the formation of the film and the countercurrent streams. The temperature of the water may approach the boiling point, and the whole assembly may be enclosed so that the ambient pressure can be maintained at any desired level.

Film thicknesses of from zero to about 0.030 inch (75 mg/cm²) are to be measured, with the emphasis on thicknesses less than about 0.002 inch (5 mg/cm²).

Gamma-n Reaction in a Heavy-Water Film

The first method to be considered is probably the least attractive, but does have the advantage that the radioactive source and detector system can be insulated from the water and vapor, and in principle, the method can be applied to a tube with a small diameter.

The method consists of using heavy water as the working liquid, irradiating it with a strong source of high-energy gamma rays, and detecting the neutrons generated. The arrangement is shown schematically in Figure 3.
A source of radiation is mounted in a heavy shield with a small collimating hole, or horizontal slit if the whole cross section of the test section is to be examined, and provided with means for raising and lowering it. The remaining region surrounding the test section is filled with neutron-sensitive detectors. The drawing indicates the use of Geiger counters, filled with a gaseous form of boron, as an example, but scintillation counters as well as other types would be equally suitable. In operation, the radiation issuing from the collimating opening in the source shield illuminates a portion of the film (shown in the drawing as included between the planes defined by "H") in the test section. Some of the gamma rays react with the deuterium making up the heavy water and liberate neutrons by photodisintegration of the deuteron. The number of neutrons liberated per unit of time will depend on the total mass of heavy water in the illuminated region, but the neutrons will be ejected in all directions, so that the detectors should be as extensive as possible in order to catch as many neutrons as possible. Also, the neutrons will have high energies; thus they can travel great distances through almost anything, including the walls of the tube, without being captured. As a consequence, the counting rate in an arrangement as shown in Figure 3 would be small unless a very strong source were used. A better design would include a container filled with either ordinary water or an organic liquid or solid surrounding the test section with the detectors embedded in it. These materials have great slowing-down power for fast neutrons, and since slow neutrons are much more easily detected than fast ones, the counting rate would be greatly increased.

Scanning the test section would be accomplished by raising or lowering the source of gamma rays. It would not be necessary, in general, to change the position of the detectors, although close to the reservoir at the top there might be loss of neutrons leaking out in the vertical directions. As mentioned earlier, the source would have to be very intense, and the radiation emitted would have to be quite energetic for the method to work. Cobalt-60, for example, does not emit radiation of sufficient energy to liberate neutrons. The radioisotope sodium-24 is a suitable source material, but has an unfortunately short half-life. After 30 hours the source strength decreases to a quarter of its original value. This would imply that transit times from the generation of the isotope in a neutronic reactor to the completion of an intended set of operations or measurements would have to be a day or two only. Some very crude estimates indicate that for a film thickness of 0.001 inch a source strength of 10 curies might be needed to give a counting rate of one count per second.

The problem of source strength is difficult to solve. The only alternative to sodium-24 appears to be potassium-42 which has a similar half-life. All the other sufficiently energetic radionuclides have even shorter half-lives, most of them very much shorter, on the order of minutes or seconds and on down. If the half-life is conveniently long, the energy of the gamma is insufficient to liberate the neutron. This, of course, is in accordance with general relations for most radioactive isotopes; that is, the more energetic the radiation, the shorter the half-life.
Liquid-Film Absorption of Beta Rays

The second method to be taken up involves the detection of beta rays, and their absorption in traversing the liquid film. The same principles are applicable to alpha particles, but they are able to penetrate film thicknesses only up to about 0.001 inch of water (2.5 mg/cm²) if there are no other losses. The alpha particle, being relatively massive and carrying a double charge, loses energy very rapidly when penetrating dense matter. Beta rays, on the other hand, can penetrate more than 100 times the thickness, depending on the energy. In this case, there is a wide choice of beta-ray energies available, and the half-lives are conveniently long, so that disappearance of the intensity is not a problem. Furthermore, they are much more easily detected than neutrons, and source strengths in the millicurie range should prove satisfactory.

The experimental arrangements for using this principle are rather simple. The test section can be fabricated of a radioactive metal, or a suitable source of beta rays can be plated on the inside of the tube, or the tube can be made with a suitable metal which is nonradioactive and then the whole piece made radioactive by neutron bombardment in a reactor or suitable accelerator. The presence of gamma rays in the activated material should not be troublesome because the greater penetrating power of the gamma rays implies that the differences in film thicknesses would have little effect on them and they would present a uniform background which could be subtracted off. Also, most detectors do not respond as readily to gamma rays as to beta rays, so even the background should be small.

The detector would be placed in the center of the tube, with some collimating shielding around it so that only portions lengthwise of the test section would be "seen." Movement of the detector up or down the tube would permit measurements along the length of the section. Geiger counters which are responsive to beta rays are available in diameters down to about 1/4 inch. The presence of some sort of collimator might require a little more room, so that the minimum diameter of the test section would have to be determined from these limits, and from the acceptable amount of throttling of the countercurrent gas-vapor mixture. There would be the additional problem of condensation on the detector, which would give false indications of the true film thickness. Condensation might be eliminated by the use of an auxiliary heater around the sensitive part of the detector, or if the amount of liquid condensed reached some equilibrium value, suitable corrections could be introduced.

Other types of detectors are available for beta rays. A particularly attractive one is the gold-plated silicon surface-barrier detector which has been used for alpha particles. The effects of a moist atmosphere on these instruments is not known, but with sufficiently energetic beta rays, the penetration of a suitable encapsulating material should present no difficulties. These detectors, which are commercially available, could probably be made as small as the Geiger counters and would give improved performance.
Backscattering of Beta Rays by Liquid Films

The third method is similar to the second, but eliminates the need of a radioactive test section. The principle involved is the use of back-scattered beta rays to measure the film thickness. If a source of radiation and a detector are placed side by side, with some interposed shielding to reduce the background counting rate, and this assembly is brought close to a solid or liquid surface, the beta rays enter the material and some are scattered back into the detector. In contrast to the usual surface reflections, the particles enter more deeply into the substance before being scattered. Consequently, a film of liquid over a piece of metal will scatter differently than the piece of metal alone, the liquid film making the difference. The scattering would be still different if the metal were removed and the depth of the liquid were essentially infinite. Applications of this technique to the measurement of the thickness of air films show that thicknesses on the order of 0.05 mg/cm² (0.00002 inch of water) can be distinguished.

The apparatus necessary to measure film thicknesses using the back-scattering principle should be the simplest of any of the methods considered so far. The source, intermediate shielding, and detector would all be mounted together as a unit, in tandem and with a common axis. This unit would be inserted along the centerline of the test section and counting rates observed as the unit is raised or lowered. The complete source-detector unit could be removed completely from the test section for servicing or inspection and would be more easily handled than the equipment required in the other methods. If a Geiger counter is used as the detector, it would not be unreasonable to expect the whole source-detector combination to be incorporated in a volume about 1/4 inch in diameter by an inch long.

The collimation problems in the backscattering method would be considerably simpler than in the second method involving the radioactive test section. Most of the backscattering would be localized in the plane midway between the source and the detector, that is opposite the intermediate shield. This would mean that the depth of the film would be measured over a relatively small region in the axial direction of the test section, rather than being averaged over a considerable length. By suitable masking, the beta rays could be made to emerge in substantially a single direction, so that details around the circumference of the test section could be revealed. Finally, the source strength necessary to give adequate counting rates would be an order of magnitude less than that necessary for a radioactive test section, making for convenience of handling and reducing the problem of personnel exposure. The method would, however, suffer from the difficulty of fouling since it would be placed in the countercurrent stream of gas and vapor.
Measurement of Protective Coatings

Much of what has been stated above can be applied without much modification to the measurement of thicknesses of protective coatings such as paints and lacquers. The experimental environment is usually much less stringent, and ample space is available.

Either of the last two methods described may be applied. If the protective coating is applied to a subbase of steel, as an example, the steel may be made radioactive, or plated with radioactive iron. The radioactive iron emits beta particles of such an energy that a portion are absorbed in the coating, the amount of the absorption depending on the thickness of the coating. As a result of wear or weathering, the coating would become thinner and larger numbers of beta rays would escape and be detected. This technique, with some modification, has been applied to the study of wear on flooring materials. It was found that more detailed measurements could be made in less time than by using other methods.

When the base material is wood, or a wood product, this procedure is not very effective. The method indicated for examining paint on wood is the backscattering technique. Most paints carry a pigment which is usually lead or titanium, to as much as 30% (molar percent), and these heavy metals would have higher scattering powers for beta rays than the wood base which is composed almost exclusively of low-atomic-weight materials. Any thinning or loss of the surface covering would reduce the amount of backscattering and give a measure of the amount lost. On the other hand, titanium-pigmented paint on steel might give disappointing results with the backscattering method because of the proximity of the atomic numbers of titanium and iron. At the present time, apparently, there is no satisfactory method for the nondestructive examination of painted wood; so if backscattering techniques should prove to be feasible, there would be clear advantages in their use.

Other Methods

During the course of gathering pertinent information and interviewing persons interested in this study, other methods were suggested. One interesting possibility involved constructing the test section out of uranium metal. Natural uranium metal, when in equilibrium with its decay products, emits all three types of radiation and offers the possibility of using combinations of the methods mentioned above. The barrier-type detectors respond preferentially to charged particles, such as alpha and beta rays, so that the gamma-ray background would not be particularly troublesome. A detector placed along the center of the test section would respond to the beta rays for thicker films, but to both the alpha and beta rays when the film thickness is less than about 2 mg/cm². With suitable electronic circuits, it is possible to separate the detector responses to these two types of radiation. The sensitivity to changes in film thickness close to zero would be greatly enhanced by the alpha particles.
CONCLUSIONS AND RECOMMENDATIONS

Measurement of the Migration of Water in Concrete. The study of the migration of water in concrete by means of water tagged with a heavy isotope of either hydrogen or oxygen with concentrations determined by a mass spectrometer appears to be feasible and the design of suitable experiments straightforward. Mass spectrometers adequate for measuring the required concentrations appear to be available from several companies at costs ranging from $2,000 to $35,000. Infrared spectroscopy also offers possibilities of measuring the necessary concentrations; most of the equipment is on hand at NCEL.

Measurement of the Deformation of a Hollow Sphere. The use of alpha particles from selected radioisotopes for measuring small displacements in awkward locations, such as inside a hollow sphere, appears to be practical for displacements in excess of about 0.001 inch. Improvements in this lower limit can be expected with further development. The use of thin sources which emit more nearly monochromatic radiation appears to offer a possibility. Experimentation with a view to application or further refinement would seem to be implied.

Measurement of Liquid Films. Of the three methods examined for measuring the thickness of liquid films flowing in a tubular test section, the method which appears to offer the best chance of success employs the principle of selective backscattering of beta rays from the film. The source-detector apparatus can be designed into a small compact unit and placed at adjustable positions along the axis of the test section.

Of the various methods using radioisotope techniques found in the literature, the backscatter method appears to give greater sensitivity to changes in film thickness while avoiding most of the difficulties of the other methods, and requires minimum strength in the source of radiation. It also appears to be considerably simpler than some of the optical methods in use, although it is probably not capable of yielding as detailed information about small areas of the film.

In view of the obvious importance of film measurements in the investigation of heat exchangers and allied equipment, and in the nondestructive evaluation of protective coatings, and because of the apparent simplicity of the backscatter method, it is suggested that preliminary experimental evaluation of the method should be undertaken.

ACKNOWLEDGMENTS

The performance of this study was made possible by the interest of several members of the NCEL staff, both in bringing the problems to the attention of the writer and in helping evaluate possible solutions. He is particularly indebted to Mr. D. F. Griffin of the Materials Division for assistance with Parts 1 and 2, to Mr. Earl Beck of the Energy Division and Mr. J. B. Crilly of the Chemistry Division for their interest in Part 3, and to Dr. L. B. Gardner of the Physics and Mathematics Division for helpful discussions about backscattering methods.
Appendix

SUMMARY OF FORMULAS AND IONIC YIELDS FOR DIFFERENT MASSES AND HEAVY-WATER CONCENTRATIONS

Table A-1
Summary of formulas for computing the relative yield of ions for a given mass from the concentrations, $\alpha$, of $D_2O$ in a mixture of $D_2O$ and $H_2O$.

The various expressions are based on the values of intensities listed in Tables I and II, and are relative to the yield for mass 18 which has been taken arbitrarily as 100.

<table>
<thead>
<tr>
<th>Mass</th>
<th>Percentage of Total Ion Current for a Particular Ion From $H_2O$</th>
<th>From $D_2O$</th>
<th>Combined Yield Relative to Mass 18 = 100</th>
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<tr>
<td>1</td>
<td>100</td>
<td>0</td>
<td>$\frac{1 - \alpha}{1 - 0.9\alpha}$</td>
</tr>
<tr>
<td>2</td>
<td>$\frac{100 - \alpha}{1 - 0.5\alpha}$ 50 $\frac{\alpha}{1 - 0.5\alpha}$</td>
<td>$\alpha$ $1 - 0.9\alpha$</td>
<td>$\frac{2 - \alpha}{1 - 0.9\alpha}$</td>
</tr>
<tr>
<td>4</td>
<td>0</td>
<td>100</td>
<td>$\frac{2 - \alpha}{1 - 0.9\alpha}$</td>
</tr>
<tr>
<td>10</td>
<td>100(1 - $\alpha$)</td>
<td>100$\alpha$</td>
<td>$\frac{1}{1 - 0.9\alpha}$</td>
</tr>
<tr>
<td>17</td>
<td>100</td>
<td>0</td>
<td>$\frac{1 - \alpha}{1 - 0.9\alpha}$</td>
</tr>
<tr>
<td>18</td>
<td>$\frac{100 - \alpha}{1 - 0.9\alpha}$ 10 $\frac{\alpha}{1 - 0.9\alpha}$</td>
<td>$\alpha$ $1 - 0.9\alpha$</td>
<td>100</td>
</tr>
<tr>
<td>20</td>
<td>0</td>
<td>100</td>
<td>$\frac{100 \alpha}{1 - 0.9\alpha}$</td>
</tr>
</tbody>
</table>
Combined yields of the ionic masses relative to mass 18, arbitrarily given the value of 100, for the labeled molecules \( \text{H}_2\text{O}^{17} \) and \( \text{H}_2\text{O}^{18} \) in light water, in terms of the concentration parameter, \( \alpha \), for \( \alpha \leq 0.5 

<table>
<thead>
<tr>
<th>Mass</th>
<th>Yield Relative to Mass 8 = 100</th>
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<tr>
<td></td>
<td>From ( \text{H}_2\text{O}^{17} )</td>
</tr>
<tr>
<td>1</td>
<td>( \frac{1}{1 - 0.9\alpha} )</td>
</tr>
<tr>
<td>2</td>
<td>( \frac{2}{1 - 0.9\alpha} )</td>
</tr>
<tr>
<td>16</td>
<td>( \frac{1 - \alpha}{1 - 0.9\alpha} )</td>
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<tr>
<td>17</td>
<td>10 ( \frac{1 - \alpha}{1 - 0.9\alpha} )</td>
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<tr>
<td>18</td>
<td>100</td>
</tr>
<tr>
<td>19</td>
<td>( \frac{100 \alpha}{1 - 0.9\alpha} )</td>
</tr>
<tr>
<td>20</td>
<td>( \frac{100 \alpha}{1 - 0.9\alpha} )</td>
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Note: For \( \alpha \geq 0.5 \), the mass 19 and mass 20 yields, respectively, exceed the mass 18 yield, and the spectrum should be renormalized. This has been done in Table A-111.
Table A-III

Relative ionic yields for the mass in the mass spectra for different values of concentration parameter, $\alpha$, for three different mixtures of isotopically labeled water in light water.

The yields are calculated relative to the yield for the mass with the largest abundance, which has been assigned the arbitrary value of 100. The tabular values were computed from the relative intensities for D$_2$O and H$_2$O listed in Tables I and II and from similar quantities for H$_2$O$^{17}$ and H$_2$O$^{18}$. Experimentally, these values would have to be measured and Table A-III recalculated to be applicable to actual measurements.

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</tr>
<tr>
<td>1</td>
</tr>
<tr>
<td>2</td>
</tr>
<tr>
<td>4</td>
</tr>
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<td>16</td>
</tr>
<tr>
<td>17</td>
</tr>
<tr>
<td>18</td>
</tr>
<tr>
<td>20</td>
</tr>
<tr>
<td>H$_2$O$^{17}$ in H$_2$O</td>
</tr>
<tr>
<td>1</td>
</tr>
<tr>
<td>2</td>
</tr>
<tr>
<td>16</td>
</tr>
<tr>
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</tr>
<tr>
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<td>19</td>
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Continued
Table A-III (Continued)

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<th>0.2</th>
<th>0.5</th>
<th>0.8</th>
<th>0.9</th>
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<td></td>
<td></td>
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<tr>
<td>H₂O¹⁸ in H₂O</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
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REFERENCES


Figure 1. Theoretical mass spectra for D₂O in H₂O.
Figure 2. The essentials of thin-film formation.
Gamma-Ray Source
Lead Shield

Figure 3: Measuring thin films with the gamma-n reaction in heavy water.
## SOME RESEARCH APPLICATIONS OF RADIOISOTOPE TECHNIQUES AT NCEL

### ABSTRACT

Specific applications of radioisotope techniques in three areas of the research program at the U. S. Naval Civil Engineering Laboratory (NCEL) are studied and evaluated. The areas include measurements of the diffusion of isotopically labeled water in concrete; the measurement of small distortions in a hollow sphere; and the measurement of film thicknesses, both liquid and solid, when covering other materials.

The preferred technique for the study of the migration of water in concrete involves the introduction of heavy water (D₂O) into a concrete specimen and the measurement of its concentration at some later time by sectioning the specimen, extracting the water, and measuring the concentration of heavy water therein with a mass spectrometer.

The second study indicates the feasibility of measuring the internal distortion of a hollow concrete sphere to within 0.001 inch by observing variations in counting rate with variations in separation between a source of alpha radiation and a detector installed on opposite sides of the cavity.

The final investigation is concerned with the measurement of thin films, either of liquids as in a thin-film heat exchanger, or of solids such as protective coatings. The preferred method is based on the selective backscattering of beta rays from the materials under study.

The first two techniques can be applied with a minimum of effort, but the last method would probably require some further development before its full effectiveness is realized.
### Security Classification

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