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NUCLEAR METHODS FOR DETECTING A DIVERSION OF D_2O *

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ABSTRACT

The possibility of diverting D_2O for use in clandestine reactors capable of weapons grade nuclear material production has created interest in safeguarding D_2O. As part of a program to investigate the technological aspects of safeguarding D_2O, several proposed nuclear based non-destructive measurement methods have been proposed which do not require the D_2O containers to be opened. These nuclear techniques all proved workable and could lend themselves, in principle, to field application. They are neutron transmission measurements, neutron capture gamma detection, photoneutron production, and measurements of the lifetime of neutrons injected in the barrel. Since much of the instrumentation is common to several methods, more than one technique may be employed to give added assurance.

I. INTRODUCTION

The Arms Control and Disarmament Agency (ACDA) is supporting a study of the technical requirements for implementation of international safeguards at a heavy-water production plant. In this paper, preliminary results from several nuclear nondestructive assay (NDA) experiments are presented. These were investigated as possible techniques for determining D_2O or its adulteration by H_2O. In particular it was desired to find methods which could quickly and easily distinguish heavy water from light water without opening the container and which were adaptable for field use by inspectors.

The present effort concentrated on developing methods for assaying the "standard" 210 liter (55 gallon) steel drums, in which heavy water is often stored. Large numbers of such drums are often present at production plants, at heavy water reactors awaiting commissioning, etc. However, it will be evident that the methods discussed are also adaptable for in-flow monitoring in a D_2O production plant, either for process control or safeguards purposes. The particular diversion scenario considered in developing the methods to be described below, was the siphoning off of amounts of D_2O from each of many barrels (in order to obtain a reasonable amount, i.e., a significant fraction of what is needed for a small clandestine reactor), and replacing the withdrawn amount with H_2O, together with some material to bring the weight and water level up to the nominal value. It is assumed that barrel weight and fill level are checked as a matter of course, the latter by an external technique. The problem then resolves itself into determining whether the nominally pure D_2O (generally 99.75% D_2O, 0.25% H_2O) has been adulterated, and, possibly, to what extent. This adulteration would have to be done in quantity enough to divert 10-20 tonnes of D_2O, considered the necessary amount to operate a reactor capable of producing one significant quantity of plutonium ($\sim$ 8 Kg) in one year.

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The basis for the use of nuclear methods to distinguish D_2O from H_2O are the very different nuclear properties of D and H (and hence for D_2O compared to H_2O). Moreover, the fact that energetic gamma rays and neutrons easily penetrate the stainless steel drum and the water, allows the drums to be probed without opening the seals. The relevant parameters for the two nuclides are listed below:

Table I Selected Nuclear Parameters for D_2O and H_2O

	D_2O	H_2O	units	Ratio (D_2O/H_2O)
M (migration length)	170.	5.9	cm	29:1
Σ_a (absorption mean free path)	7.5×10^{-5}	1.88×10^{-2}	cm^{-1}	1:2500
Σ_t (transport mean free path)	2.36	0.44	cm^{-1}	5.4:1
σ (n,γ) cross section	$\sim$ 0.5	330	mb	1:660
σ (γ,n) cross section	$\sim$ 1	0.0	mb	∞
Capture γ energy	2.226	6.270	MeV	1:2.8
α_o reciprocal of the infinite medium die-away time	19.0	4.77×10^3	s^{-1}	1:250

There are several available basic nuclear measurement techniques which exploit these differences:

(1) Neutron transmission - the different moderating and absorption properties will strongly affect the transmission of neutrons through the respective media.

(2) Neutron capture gamma rays - while both absorb slow neutrons to emit gammas via the respective reactions $n + H \rightarrow D + \gamma$, $E_\gamma = 2.226$ MeV and $n + D \rightarrow T + \gamma$, $E_\gamma = 6.270$ MeV, the reaction with H is not only more than 600 times more probable, but the respective gamma energies are different enough to be easily distinguishable.

(3) Neutrons from photodisintegration - gamma rays in the several MeV range can cause photodisintegration of D (the inverse reaction to n capture by H mentioned above), but there is no corresponding reaction with H. While the count rates are predictably low, the discrimination ratio is infinite, and hence possibly workable in a suitable low neutron background environment.

(4) Neutron lifetime - the lifetime (die away time) of neutrons injected into the respective media are measurable with standard techniques, and are quite different.

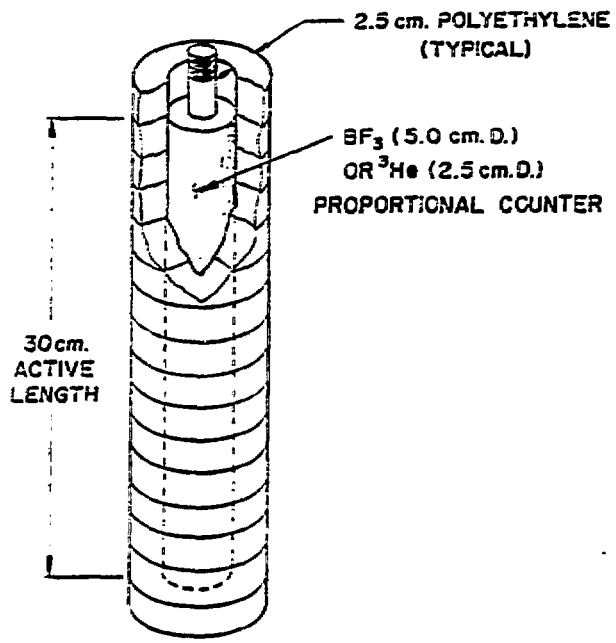
These methods can be used to complement one another. The use of more than one of these techniques for inspection would make it disproportionately more difficult to defeat as a safeguards measurement. Fortunately, as will be seen in the following, much of the equipment needed is common to more than one method, and it is not too hard to conceive of a set or kit of instrumentation simultaneously making the measurements appropriate to two or more of these methods. In the following, preliminary work on each of the methods discussed in rough order of their complexity.

II. NEUTRON TRANSMISSION

In this series of measurements, both 210ℓ and 60ℓ drums were used. Some contained D_2O (> 99.7%), some H_2O and one contained a 74% D_2O mixture.

A 20 Ci source (^{241}Am-Li), providing about 2×10^5 n/sec with average energy $\sim$ 400 keV, was placed on one side of the drum, up against it, and centered vertically. At the corresponding position, directly opposite the source, a BF_3 neutron counter (sensitive to thermal neutrons) was placed. The dimensions of the counter were approximately 5 cm in diameter and 30 cm in active length. Measurements were made both with the bare counter and with a moderating 2.5 cm thick polyethylene sleeve (Figure 1). It was found that the bare counter provided better discrimination between H_2O and D_2O, since the sample itself provided adequate moderation.

Figure 1: SINGLE NEUTRON COUNTER AND MODERATOR



The neutron counter signals were amplified conventionally and displayed on a multichannel analyzer, although a simple discriminator-scaler would have sufficed. Results for a series of 100 runs are presented in Table II. In this type of measurement, the effects of absorption, scattering and moderation are combined to give what can be considered an effective transmission

rate for the particular source, and source-drum-detector geometry.

TABLE II
TRANSMISSION MEASUREMENTS

SAMPLE	COUNTING RATE (sec^{-1}) (MODERATED)	(BARE)
210ℓ D_2O	136	157
210ℓ H_2O	92	66
210ℓ Ratio $\frac{D_2O}{H_2O}$	1.48	2.38
60ℓ D_2O	192	194
60ℓ H_2O	129	81
60ℓ D_2O 74%	157	159
60ℓ Ratio $\frac{D_2O}{H_2O}$	1.49	2.40
NO SAMPLE (drum away)	362	98
NO SOURCE (Background)	1	3

The rejection ratio does not depend on the size, but is much improved for the bare detector. The 74% point for the bare case is close to what a linear interpolation would give (159 vs. 165).

The difference between light and heavy water is seen to be substantial.

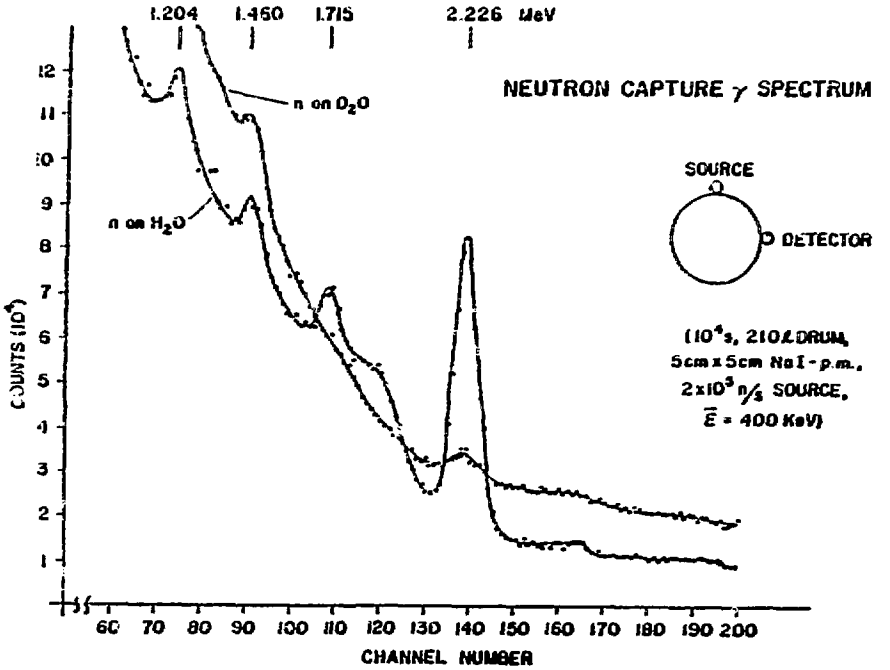
III. NEUTRON CAPTURE

The same ^{241}Am-Li source as in Part II was placed against the barrel. An unshielded 5 cm x 5 cm NaI-photomultiplier tube gamma detector was placed against the drum and near the neutron source, but shielded from it. This was used to monitor capture gamma rays from the drum. The γ spectrum was analyzed by a multichannel analyzer.

The $H(n,D)\gamma$ capture gamma rays were clearly visible after only a few seconds of data collection, while the $D(n,T)\gamma$ capture gamma was for practical purposes not seen for reasonable counting times.

The most interesting region of a typical gamma spectrum resulting from the neutron bombardment of H_2O on a 210ℓ drum is shown in Figure 2. The peaks at 1.715 and 1.204 MeV are respectively the one and two annihilation radiation escape peaks from the 2.226 MeV characteristic neutron capture gamma ray of H, while the 1.460 MeV peak is due to ^{40}K in the building material of the laboratory where the experiment was set up. These three peaks rest upon a steeply falling background which is presumably due to multiply-scattered capture gamma rays from the laboratory environment. The 2.226 MeV peak is fortunately quite free of interferences and well defined.

Figure 2: γ-SPECTRUM FROM NEUTRON CAPTURE ON D_2O AND H_2O



The spectrum of gamma rays seen by the detector, when a 210ℓ barrel of D_2O is bombarded with neutrons, is shown superimposed on that for H_2O. The small 2.226 MeV peak is a few times larger than can be accounted for on the basis of the 0.23% H_2O content in the "pure" D_2O drum. It is probably mainly due to H in the environment, but the different energy spectrum of neutrons in the D_2O environment may also play a part. The effect of this is merely to have a non-zero intercept in the calibration curve of D_2O content versus "2.226 MeV peak response".

A curiosity of these spectra is that the background is distinctly higher for the D_2O than for the H_2O. This is presumably because D_2O creates a different ambient neutron flux in its vicinity than does H_2O. Thus a careful Compton background subtraction is required to obtain a true net count for the 2.226 MeV capture gamma rays.

The H_2O barrel gives about 60 counts/s above background in the peak. For measuring a 5% concentration of H_2O in a barrel, a net 3 counts/s (relative to a 30 count/s background) implies that a 100 second run could give better than a 3σ deviation from zero, and could measure concentration to ±1% absolute, apart from limitations due to systematic errors.

IV. PHOTODISINTEGRATION

In order to use the gamma capture reaction, a gamma source of at least $10^5\gamma$/sec over the threshold of 2.226 MeV must be used. A ^{228}Th source would be ideal, since a daughter in the chain, ^{208}Tl, has a 2.61 MeV gamma ray. The controlling half-life (of ^{228}Th) is 1.9 years. Such a source was not immediately available, so a substitute sample of ^{24}Na was used. This was produced by neutron bombardment of ^{27}Al in the BNL High Flux Beam Reactor.

The energy is 2.75 MeV, not far from the 2.61 MeV of ^{228}Th which was envisioned for field use.

The source was placed next to the sample and a neutron counter (as above in Part II) was used to measure the rate.

The D_2O sample gave $\sim$ 0.45 counts/sec. The H_2O sample gave less then 1% of this rate. Thus, a good potential for a quantitative measurement, directly sensitive to D_2O exists. The source intensity was measured at 160 mR/hour at the mouth of the small shielding cask used.

About 1000 seconds would give 450 counts for a "pure" D_2O sample. A 5% D_2O sample would only give $\sim$ 22 counts in this time. Rates could be increased by using a stronger source, but this could require much heavier shielding.

In a few seconds, the two cases can be distinguished to several standard deviations. Somewhat longer runs should be able to give quantitative results on mixes.

V. NEUTRON DIE AWAY TIME

This method is distinctly more complicated theoretically and experimentally, but has an advantage in that it doesn't require transport of gamma and neutron sources, since it makes use of an pulsed accelerator. The basic principle involved is measurement of the lifetime of neutrons in a drum of D_2O or H_2O. Both geometric (the size of the drums) and nuclear properties will affect the neutron lifetime, but the size of the drums is such that nuclear properties play an important role.[1]

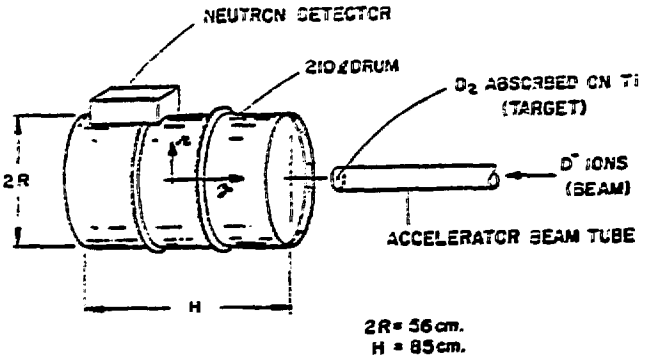
Fast neutrons from a pulsed source enter the barrel. A significant fraction of them become thermalized and form an initial distribution throughout the drum which then decays with time due to capture by the materials in the media and leakage through the drum surface. A thermal neutron detector placed near the drum samples neutrons leaking out. The instantaneous count rate is taken to be proportional in some sense to the density of the thermalized neutrons in the drum.

The spatial and temporal behavior of the neutron density in the drum is given in the "one group diffusion approximation" by the solutions of the equation

$$\frac{\partial \rho}{\partial t} = S + D\nabla^2\rho - \Sigma_a v\rho$$

where S is a source term, the number of neutrons produced ($1/cm^3s$), D is the Fick's law diffusion constant (cm^2/s), $\Sigma_a = n\sigma_a$ (cm^{-1}) is the macroscopic cross section for absorption, n is the number of absorbing centers per cm^3, σ_a is the cross section for absorption (cm^2) v is the neutron speed representative of the thermalized neutrons, and t is the time (s). Considering the time origin (t=o) to be after the neutron pulse is over, S=o. Then the solutions of the differential equation appropriate to the geometry of this experiment (see Figure 3) are of the form

Figure 3: SET-UP FOR DIE-AWAY MEASUREMENT



(NOT TO SCALE)

$$\rho = \sum_{mn} A_{mn} J_o\left(\frac{j_m r}{R_e}\right) \left\{ \begin{matrix} \cos\left[\frac{(2n+1)\pi z}{He}\right] \\ \sin\left[\frac{2n\pi z}{He}\right] \end{matrix} \right\} e^{-\alpha_{mn} t}$$

This is seen to be a summation over several spatial modes each with its own time dependence (reciprocal time constant α_{mn} in s^{-1}), and amplitude A_{mn} depending on the details of the initial thermal distributions at time t=o. J_o is the Bessel functions of order zero, j_m are its zeros, $R_e = R + d$, and $H_e = H + 2d$, where d is the extrapolated distance given by $d = 0.71\ell_t$, ℓ_t being the mean free path for transport defined below. The inverse time constant is given by

$$\alpha_{mn} = \alpha_o + (\alpha_o L^2)B_{mn}^2 - C(B_{mn}^2) \approx \alpha_o + (\alpha_o L^2)B_{mn}^2$$

Here $\alpha = \Sigma_a v$; $L^2 = \frac{\ell_a \ell_t}{3} = \frac{\ell_a \ell_p}{3(1-\bar{m})} = \left[3(1-\bar{\mu})\Sigma_a \Sigma_s\right]^{-1}$, where $\ell_a = 1/\Sigma a$

is the mean free path for absorption, ℓ_t is given by $\ell_t = \ell_s/(1-\bar{\mu})$ with $\ell_s = 1/\Sigma_s$ the mean free path for scattering and μ the average (laboratory) scattering angle; $D = \Sigma_a v L^2$. This is given by

$$\mu = \frac{2}{3} \frac{\Sigma_i N_i \sigma_i / A_i}{\Sigma_i N_i \sigma_i} = \frac{2}{3} \left\langle \frac{1}{A_i} \right\rangle$$

N_i being the number per cm^3 of nuclei nuclide mass A_i and scattering cross section σ_i present; this is seen to be 2/3 of an average of the inverse nuclide masses. The quantity B_{mn}^2 is called the "buckling". For the geometry shown,

$$B_{mn}^2 = \begin{cases} \dfrac{(2n+1)^2\pi^2}{He^2} + \dfrac{j_m^2}{Re^2} & \text{modes symmetric in z,} \\ \dfrac{n^2\pi^2}{He^2} + \dfrac{j_m^2}{Re^2} & \text{modes anti-symmetric in z.} \end{cases}$$

C is called the diffusion cooling and can be calculated theoretically or obtained empirically by fitting experimental die away decay times as a function of buckling. In the present situation, the CB^4 term is negligible for H_2O; for D_2O it is of the same order of magnitude as α, but small compared to the $\alpha_o L^2 B^2$ term.

In practice the time constants decrease rapidly enough with increasing mode numbers n,m so that only the lowest order mode is seen. Under special circumstances one and even two higher order modes may be observed. The circumstances are for $1/\alpha_o$ to be large, that the geometry of the neutron source relative to the drum be such as to be favorable for the excitation of the higher order modes, and that the detector be placed in a position relative to the drum so that it can advantageously sample neutrons from the higher order modes leaking from the barrel. Even when higher order modes can be seen, the lowest order or fundamental mode is quite distinct and differs enough for reactor purity D_2O compared to H_2O to constitute a useful signature, as will be shown below.

Representative data for D_2O and H_2O in 210ℓ drums are shown in Figures 4 and 5 for the pulsed neutron source-drum detector geometry shown in Figure 3. The data represent a cumulative multichannel scale analysis of the detector count rate as a function of time after the start of the neutron pulse, for repeated pulses. The signal which controls the pulsing of the accelerator beam also started the multichannel scaler time sweep. Signals received from the neutron detector during the time sweep are stored in the appropriate time channel of the MCS.

The measured values of the fundamental mode time constant for D_2O and H_2O are seen to be well defined and distinctly different, they are, respectively, 713±5 and 153±5 μs. The higher order mode seen in the case of D_2O has a measured time constant of 184 μs compared to a calculated value of 440 μs. The values calculated on the basis of selected published data[2] are, for D_2O and H_2O respectively, 645 and 197 μs. The reasons for these discrepancies between calculated and measured values are not well understood at this writing. However, the measured die away times are reproducible.

The detector, illustrated in Figure 6, was designed to be preferentially sensitive to thermal neutrons leaking through the surface of the barrel (since bare "1/v" (BF_3) detectors are exposed there), but insensi-

Figure 4: DIE-AWAY SPECTRUM FOR D_2O

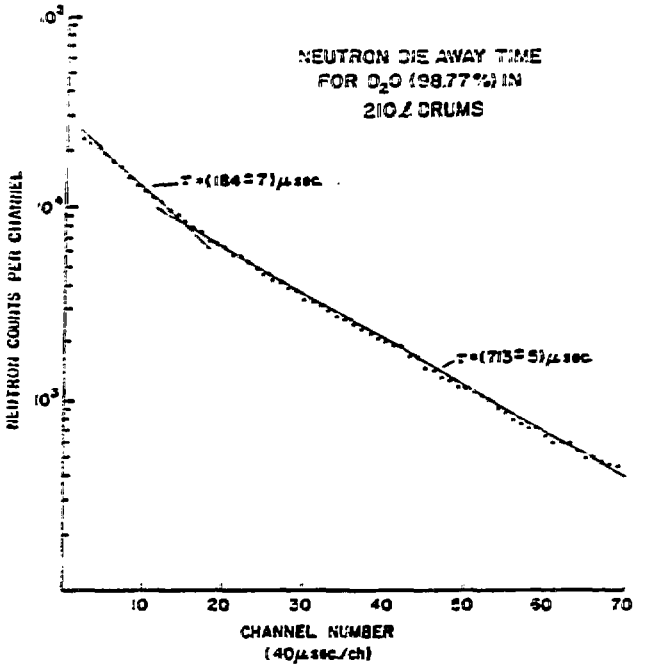


Figure 5: DIE-AWAY SPECTRUM FOR H_2O

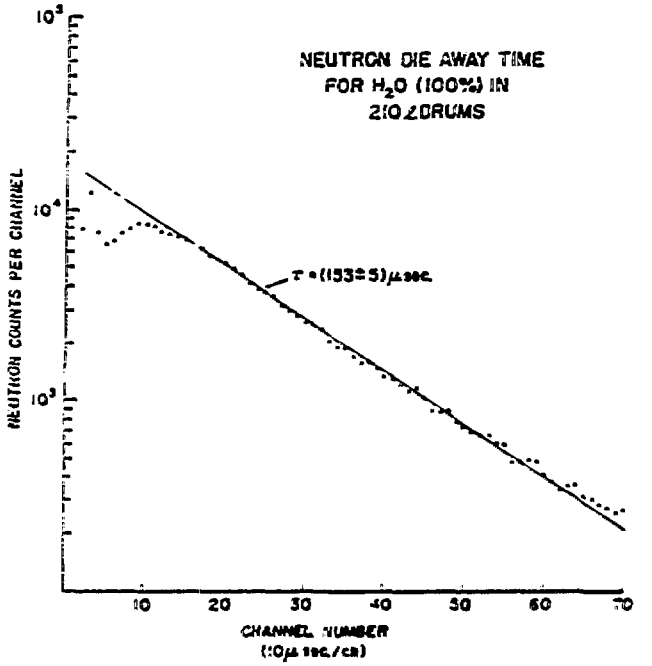
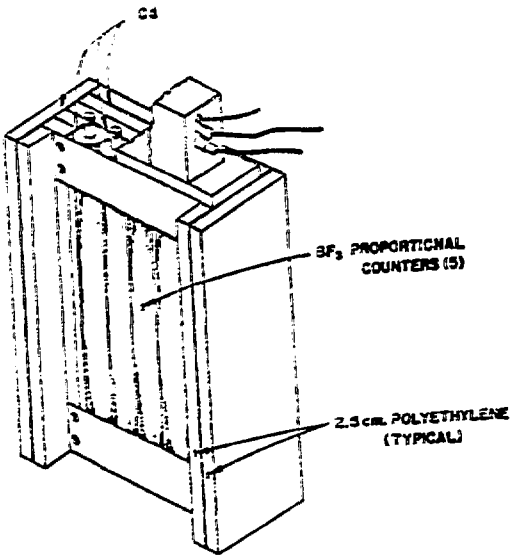


Figure 6: NEUTRON COUNTER ARRAY FOR DIE-AWAY MEASUREMENT



tive to fast and slow neutrons forming the room background by virtue of the alternate layer of 2.5 cm polyethylene and 0.8 mm Cd. It is now realized that this detector is more sensitive than it needs to be and that its performance in other respects would be improved if it were made more compact.

The estimated pulsed neutron intensity impinging on the drum is of the order of 500 n/μs. The data in Figure 4 took on the order of 10^3 seconds to accumulate. The pulse durations used in this work ranged from 10 to 40 μs, with no apparent difference as regards results. In principle, increasing the pulse durations up to the order of the decay time would increase the count per beam pulse. The pulse repetition period was of the order of 3 ms, dictated by the interest in following the decay for many time constants.

The present experiment made use of a tandem Van de Graaff to accelerate D-ions which bombarded a target of deuterium absorbed on titanium. The resultant $D(D,{}^3He)n$ reaction produces neutrons of about 7-8 MeV for the bombarding energy used. Applying this method in the field would require use of a sealed beam accelerator of the type employed in bore hole logging. These make use of the $D(T,{}^4He)n$ reaction, which yields $\sim$ 14 MeV neutrons. Sealed beam tubes are themselves portable and easily capable of the neutron intensities required. The power supplies which they are used with are normally bulkier than one would like, but there are possibilities of trading off unneeded intensity for a smaller power supply than the ones useful in bore hole logging.

VI. CONCLUSIONS

The program described above was undertaken with the aim of providing attributes measurements of heavy water versus light water; that is, to find

methods of telling heavy from light water to a high confidence level (> 99%). All four methods seem able to do this. However, there is the possibility that some methods may provide additional quantitative information as well. Perhaps most promising in this regard would be some combination of two or all of the neutron transmission, capture gamma, and photodisintegration methods. The capture gamma ray method is highly sensitive to the presence of hydrogen, and could give usually accurate answers in minutes, given a calibration run with a known barrel. The latter method is sensitive to deuterium, but can only be reasonably successful in a zone of low neutron background (less than $\sim$ 0.5 counts/s into the detector.) Calibration is necessary here as well. Since the rate is likely to be low, with sources easily handled by an inspector, somewhat longer runs will be needed for a quantitative measurement (perhaps on the order of 15-30 minutes).

Combining these methods necessitates both a neutron source and a gamma (> 2.226 MeV) source, each with an intensity of at least a few times 10^5 particles/second. Also required would be a NaI gamma detector with, at least, a single channel analyzer, and a neutron counter, with appropriate ancillary electronics. The transmission approach requires only a neutron source and a neutron counter plus electronics. Only a few minutes counting may be necessary to determine attributes; quantitative information on mixtures may be feasible in the range of higher fractions of D_2O.

The die-away time method also gives a clear attribute signal for 210ℓ. drums. Further work needs to be done to determine the ability of this method to provide a good variables measurement of fractions of D_2O and H_2O in a mixture. This, too, will be most effective for higher fractions of D_2O. The pulsed neutron source has the advantage that when not in use the only radioactivity it has is the small amount in the tritium target.

These methods will have to be compared with non-nuclear techniques for distinguishing D_2O from H_2O in large barrels. Once of the other possibilities involves use of acoustics, which we are currently investigating.

VII. ACKNOWLEDGEMENTS

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LIST OF FIGURES

LIST OF TABLES