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COMPARISON OF CESIUM IODIDE AND SODIUM IODIDE FOR UNDERWATER RADIATION DETECTION SYSTEMS

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ABSTRACT

Comparison is made between NaI and CsI scintillation detectors for application to an ultrasensitive underwater detection system. A series of measurements are reported on the response of each crystal to the gamma rays from ^{40}K distributed in water, the attenuation of the integrated count rate by aluminum and steel absorbers surrounding the detector and the background count rate from contaminants on the crystal-phototube assembly and pressure housings. It is assumed in the discussion that NaI crystals will always have to be protected from thermal and mechanical shock by a pressure vessel and that CsI, which is much less susceptible to these types of shock, can be exposed to the hydrostatic pressures in the ocean without damage. The minimum detectable change in ^{40}K concentration is calculated, based on the measured signal and background counts for a CsI crystal and several NaI assemblies with steel and aluminum pressure vessels. It is concluded that, for deep submergence work with detectors of equivalent size, a CsI system is the most sensitive and when compared in terms of a specific detection capability, the least expensive. To complete the evaluation, the assumption that CsI can be operated to all depths without a pressure vessel must be confirmed.

SUMMARY

Problem

Sensitivity of underwater detection systems is limited by detector sizes that are practical, the shielding effects of the water and any pressure vessel to gamma rays, and the background count rate from radioactive contaminants in a complete detector assembly. A comparison is desired of the sensitivity of underwater detection systems using NaI detectors which must be protected by a pressure vessel and an unshielded CsI detector of equivalent size.

Findings

Measurements were made of representative crystals of NaI and CsI showing the count rate from a standard solution containing radioactive potassium, the attenuation of this signal by the aluminum or steel walls of a pressure vessel and the count rate from radioactive contaminants in pressure vessel materials and the crystal assemblies themselves. Calculations based on these count rate measurements of signal and background are shown for an "unprotected" CsI detector and several NaI assemblies with pressure vessels.

It is shown that for deep submergence work, using detectors of equivalent size, a CsI system is more sensitive. This is in spite of the fact that the present day CsI crystals have an excessive background count from ^{137}Cs contamination in the crystal. In addition, the report concludes that to attain a specific detection capability, a CsI system is the least expensive. Tests of the response of CsI to hydrostatic pressure are recommended to complete the evaluation.

INTRODUCTION

The natural radioactivity in the ocean potentially affords an additional measurement of the physical, chemical, and biological changes in the ocean. In this context, it is desired to develop an instrument that can detect and measure the gross radiation level of the ocean and identify any structure or distribution in this level and do this <u>in situ</u> on a continuous basis. Although the natural radioisotopes in solution in sea water exceed 10^{10} tons, the concentration (curies/liter) is very small. Detection of this activity is further complicated by the shielding effect of sea water and any pressure vessel that must be used to protect a detector and by the inherent radioactive background of materials used in construction of a complete detector assembly. In terms of doserate, the gamma ray level of the natural radioactivity in sea water is 0.14 μR/h. For comparison, the normal "background" doserate over land in the San Francisco Bay area is 20 to 50 times higher.

The choice of detector to measure this very low specific activity is quickly narrowed to scintillant-photomultiplier tube combinations and further narrowed to only three readily available scintillants: plastic, NaI(Tl), and CsI. Plastic scintillants are extremely rugged and, in this respect, ideal for field measurements; however, they have relatively low light yield, (1/4 th to 1/8 th of NaI), and low stopping power for gamma rays, (1/35th of NaI at 100 keV). Large sections of plastic scintillant are required to attain a reasonable gamma ray cross section, further complicating the light collection problem. The result is that the predominant, low-energy, gamma rays, that result from Compton scatter in water of the primary gamma rays, are difficult to detect above the electronic noise of the system. Barring improvements in photomultipliers or the plastic scintillants, the choice of scintillant for an underwater detector seems to be between NaI and CsI. The following discussion reviews some pertinent characteristics and some recent measurements on these two scintillants and evaluates their relative capabilities for use in an ultrasensitive, underwater detection system.

DISCUSSION

Optimizing a scintillation system for detection of the very low gamma radiation level in the ocean has several aspects: (1) physical dimensions of the detector, (2) mechanical properties of the whole assembly for the rugged field use expected during handling at sea, (3) scintillation efficiency, (4) intrinsic sensitivity of the particular detector, and (5) the radiation background due to radioactive contaminants in a complete detector assembly. Some of these items are of more concern than others in a choice between NaI and CsI, but each has some significance and will be considered in turn.

The physical dimensions of the detector to optimize detection capability are probably the least significant of the items in a choice between NaI and CsI, except to note a comparison based on the most efficient use of a scintillant. Redmond[1], [*] contends that the volume of a scintillant is most efficiently used in small detectors whose dimensions are comparable with a mean free path for absorption of the photon energies being detected. In this context, the most efficient size for a CsI detector for the low energy scattered gamma rays in water is about 2/3 that required for NaI. This kind of consideration can be significant when large sheet detectors are being considered and the area of the edge to which a light pipe is being attached should be the same as the area of the photocathode on the photomultiplier tube.[*]

With respect to mechanical properties of NaI and CsI, there is a choice. Harshaw Chemical Company, in their brochure, have this to say: "Being relatively soft and plastic as compared to thallium activated sodium iodide, cesium iodide can better withstand severe shock, acceleration, and vibration, as well as large temperature gradients or rapid changes in temperature." Considerable difficulty has been

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[*] Redmond, A., and Schlessinger, L., "Detection of a Small Change of Concentration ^{40}K in Seawater," USNRDL-TR- , to be published.

experienced at NRDL with large volume crystals of NaI used in oceanographic measurements. They have to be insulated to protect them from rapid temperature changes and must be carefully shock mounted. Even then a significant percentage is returned to the laboratory broken. From a ruggedness point of view, CsI seems to be a far better choice for oceanographic instrumentation than NaI. There is good possibility that CsI would not have to be protected by a pressure vessel. The soft plastic nature of the crystal suggest s that it can be exposed to the sea pressures and still operate as a scintillant. It has been impossible to locate a reference in the literature on the effects of pressure on luminescence efficiency* of a CsI crystal, however, many crystal transducers, some with rather rigid crystal structures, are exposed to the sea pressures without damage. It is assumed in the subsequent discussion that CsI can be exposed to sea pressures and that part of the value of CsI in oceanographic radioactivity measurements results from the elimination of the shielding and radioactive background of a pressure vessel.

Of the two scintillants, NaI(Tl) has the highest scintillation efficiency (light yield per unit photon energy absorbed). In the literature only two years ago, NaI reportedly gave four times the light output of CsI(Tl). However, the fabricators have steadily improved the scintillation efficiency of CsI(Tl) and presently publish figures for CsI only half the NaI values. In addition, we have been told informally by one supplier that they are now growing CsI activated with Na rather than Tl and that the scintillation efficiency of this new crystal is very nearly the same as NaI(Tl).

Improved light yield in itself is not an advantage. Low light yield can be compensated for by high photomultiplier tube gain; however, improved light yield is generally accompanied by an improved signal to noise ratio and permits detection of lower energy photons. The net result is that a high scintillation efficiency crystal system more nearly approaches a count rate plateau (e.g., constant count rate with changes in system gain) for the multiply scattered photon spectrum observed from distributed sources in water. NaI has been the detector of choice for most scintillation detector work, including oceanographic measurements, and a good part of the choice has been based on its scintillation

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*Pressure effects on long term (minutes) decay of luminescence centers have been observed(2) but the effect on luminescence efficiency was not reported.

efficiency. In view of the rapidly improving scintillation efficiency figures reported for CsI, this situation is changing; and this change, in part, prompted this review of the relative merits of these two scintillants.

It should be noted at this point that the scintillation efficiency of a crystal is somewhat dependent on its temperature. In both NaI(Tl) and CsI(Tl), there is a broad plateau in the efficiency vs temperature curve near room temperature.[3] Generally the efficiency decreases as the temperature decreases. Of the two, NaI has the least susceptibility and its change is not significant for most purposes compared to the temperature dependence of the photomultiplier tubes. The light output from CsI(Tl) decreases 10 percent from room temperature to 0°C. The new Na activated CsI crystals are reputed to have a temperature dependence very similar to the CsI(Tl).

The final two items, intrinsic sensitivity and radiation background, remain to be evaluated. The remainder of the discussion is devoted to some recent measurements on sensitivity of these two scintillants to ^{40}K distributed in water, the effect of various thicknesses of pressure vessel on this sensitivity and the radiation background associated with completed detector assemblies. These measurements are reviewed in terms of the statistical variations in the mean background compared to the signal count.

Figure 1 is a set of two curves showing the pulse height spectra vs energy for the photons from ^{40}K distributed in water as measured with CsI(Tl) and NaI(Tl) detectors. The measurement was conducted in a large tank facility using dissolved $KHCO_3$ as a source. Both crystals were 4 in. d x 4 in. thick. The background of each crystal as measured in a shielded low-background counting room has been subtracted from the spectral count.

The spectra from the crystals differ only slightly. The CsI has a few more counts in the full energy peak for ^{40}K gamma rays at 1.47 MeV reflecting its higher stopping power. The number of counts at lower energy in the CsI curve is slightly less (more photons were fully absorbed and appear in the 1.47 MeV peak). The net result is that the total integrated count between 30 keV and 2 MeV for either crystal is about the same. The sensitivities expressed in terms of the normal concentration of ^{40}K in sea water (3.63×10^{-11} c/l) were CsI - 10.4 cps and NaI - 9.8 cps.

The background radiation to each crystal, however, was not the same. Figure 2 shows the pulse height spectra measured in the low

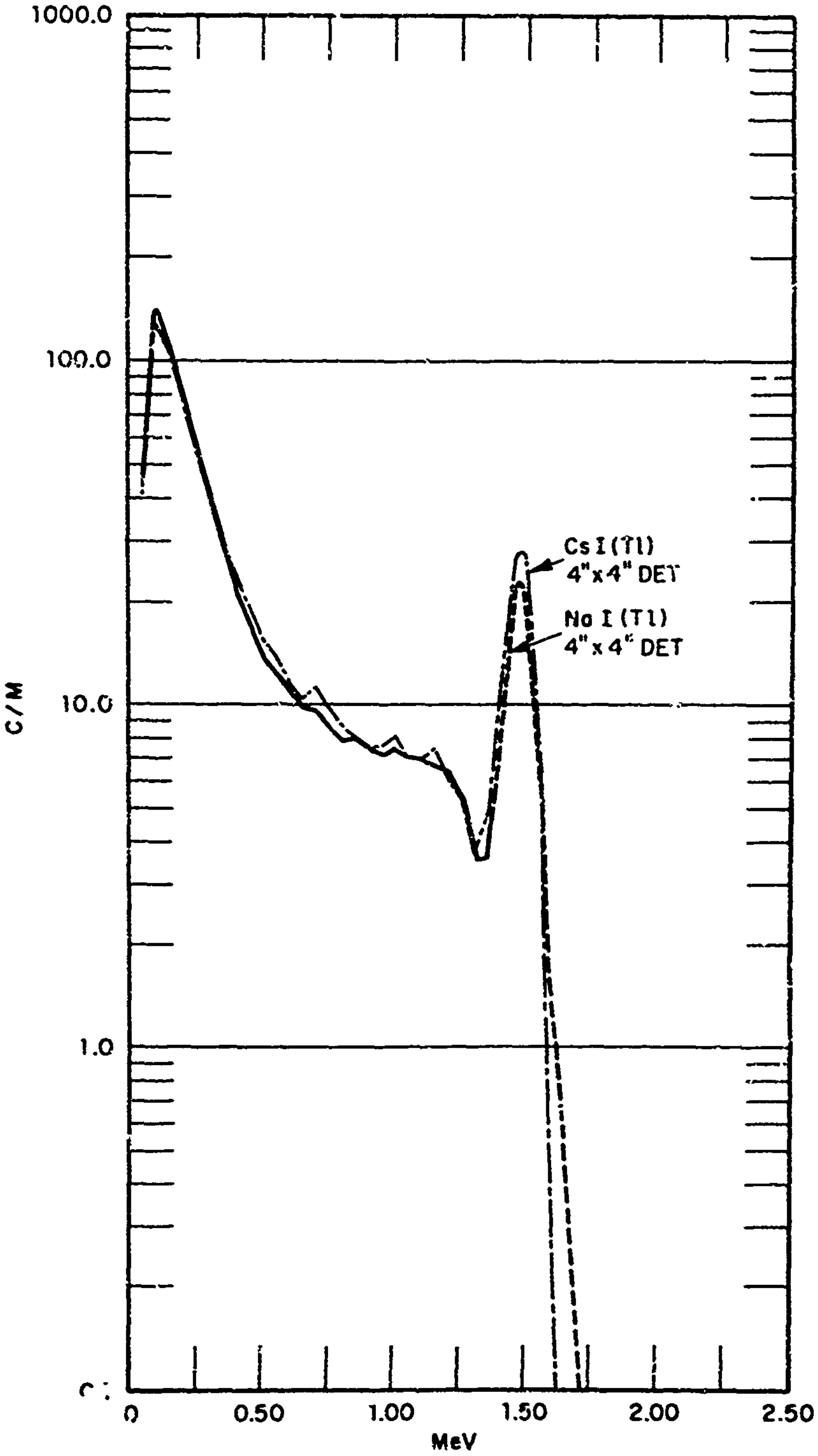

Fig. 1 Pulse height spectra of ^{40}K distributed in water as measured with NaI and CsI detectors.

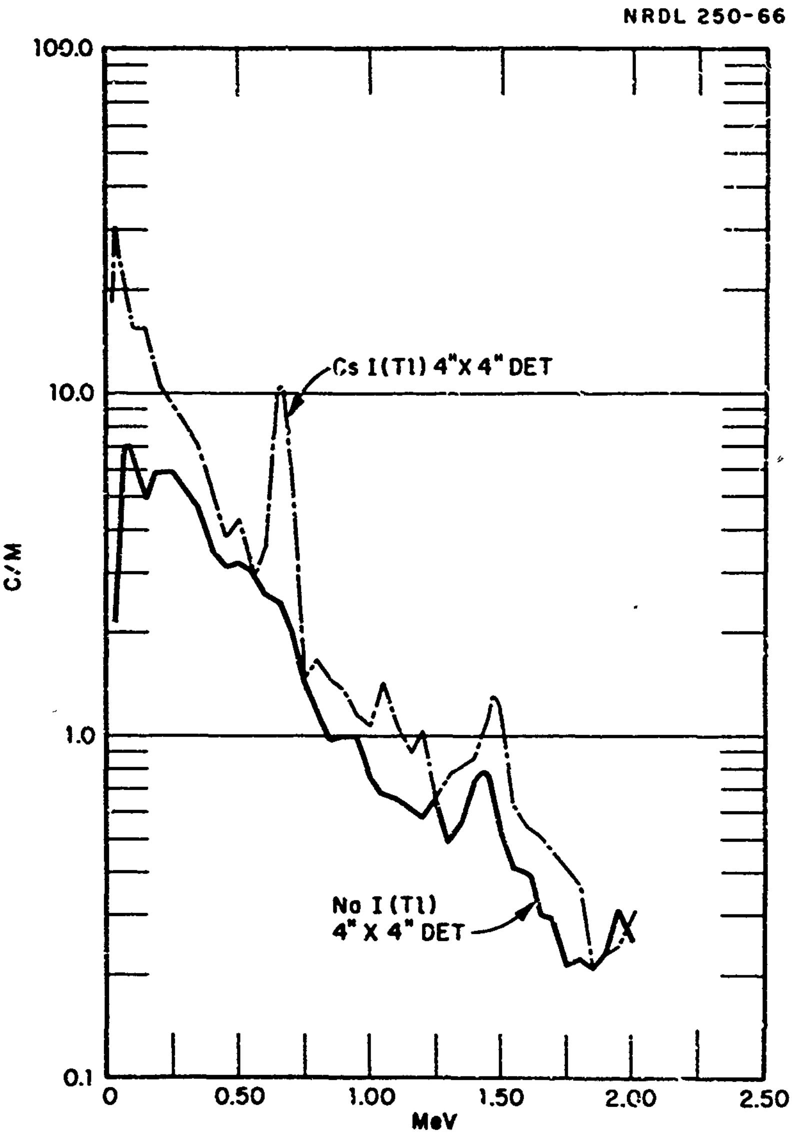

Fig. 2 Radiation background pulse height spectra measured in the NRDL low background room with NaI and CsI detectors.

background room. There is a cosmic ray component in both spectra and a slight count from the low background room itself; however, most of the difference that is evident results from radioisotopic contaminant in the CsI crystal itself. The peak at 661 keV is characteristic of ^{137}Cs, one of the persistent fallout products in our atmosphere as a result of past nuclear tests.

The integral count in the background, 30 keV to 2 MeV, for NaI was 8.3 cps and for CsI was 11.5 cps. Most of the difference is attributed to internal contamination of the CsI crystal. From previous measurements in the low background room, it is estimated that the room and cosmic rays contribute approximately 5 cps to a 4 x 4 in. crystal. Contaminants in the NaI assembly, crystal and photomultiplier tube, therefore contribute 3.3 cps to the background count and those in the CsI assembly about 6.5 cps.

Comparing the two crystals at this stage, the unshielded NaI assembly has about the same intrinsic sensitivity as CsI but has about half the background radiation count. This is not the whole story, however. First, NaI crystals are sensitive to thermal changes and mechanical shock, and in all the known underwater detection systems, they are housed in a pressure vessel. The housing attenuates the flux from radioactivity in the sea and contains radioisotopes as contaminants that add to the unwanted background count. Secondly, the manufacturer of the CsI detectors is aware of the radioactive Cs contaminants in his crystals and has made arrangements to obtain Cs ores directly from a mine and process the ores himself to eliminate the fallout component. He is confident that the next generation of CsI crystals will have a background count rate comparable to present day NaI.

Pressure vessels to house the NaI detector are commonly made of aluminum or steel in the form of cylinders. For the Trieste operations,(4) the cylinder walls were 1 in. thick aluminum and the flat end plate 2 in. thick. A steel pressure vessel for the equivalent depth would be approximately 1/2 in. thick. The effect of this mass of absorber between the crystal and the distributed activity in sea water can be calculated easily for the primary radiation, but the attenuation of the scattered component is less obvious. Figure 3 shows a set of three curves of the pulse height spectra observed with a CsI detector immersed in a solution containing ^{40}K when the detector is unshielded, shielded by 0.864 in. of aluminum, and shielded by 0.228 in. of steel. The full energy peak at 1.47 MeV is attenuated by the aluminum and

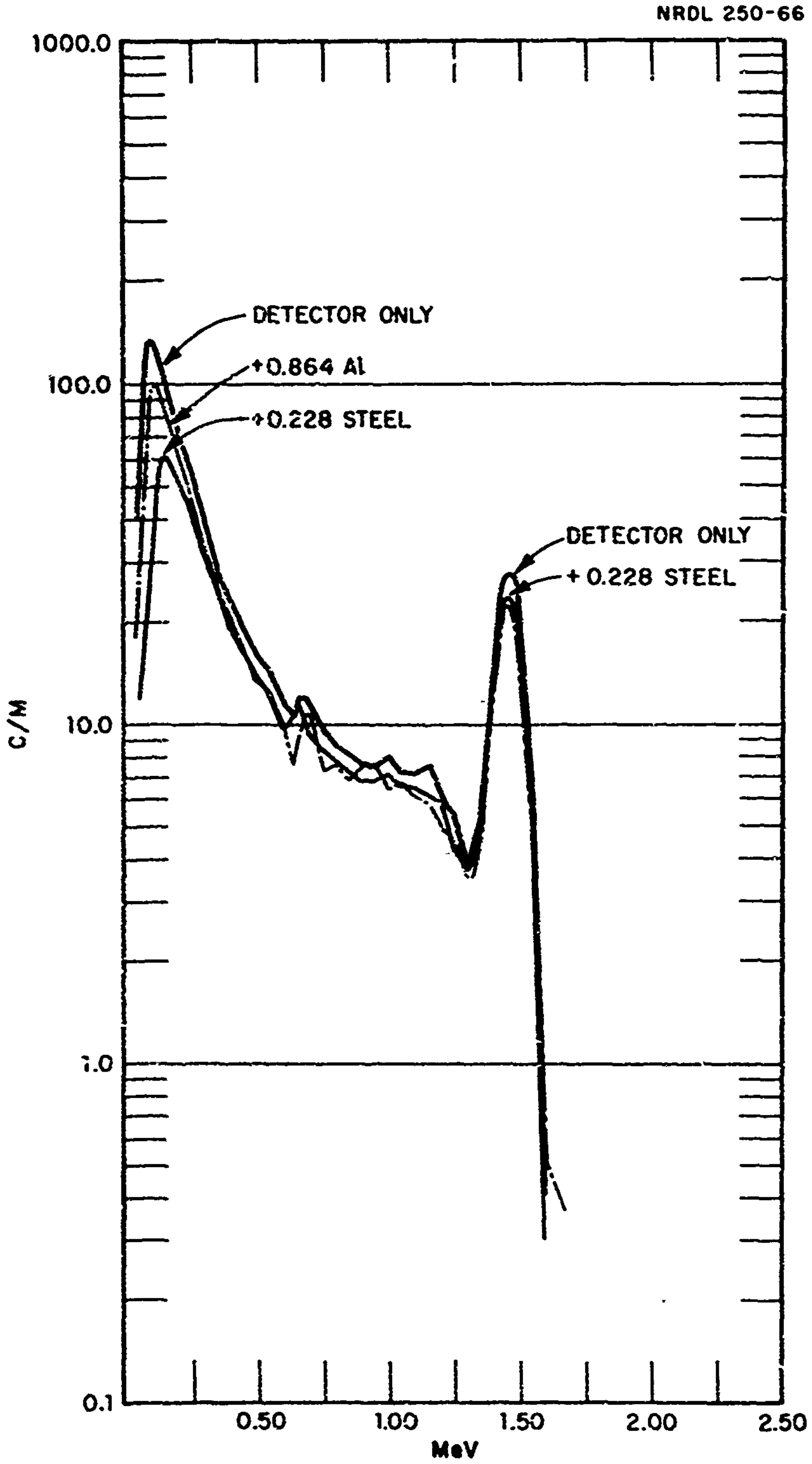

Fig. 3 Pulse height spectra observed with CsI (4 in. t X 4 In. d) detector immersed in a solution containing ^{40}K when the detector is unshielded; shielded by 0.864 in. of aluminum; and shielded by 0.228 in. of steel.

steel as depicted by $I/I_O = e^{-\mu x}$ where μ is the total attenuation coefficient for the 1.47 MeV gamma rays and x is the thickness of the material (14 percent for steel and 18.5 percent for aluminum). Attenuation of the scattered radiation at 80 to 100 keV is considerably different. The curve for aluminum is down 26 percent - the curve for steel down 55 percent.

A series of curves of the type in Figure 3 were made on ^{40}K with varying thicknesses of absorber around the crystal. These were integrated between 30 keV and 2 MeV and the integrated count compared to an unshielded crystal and plotted against absorber thickness in Figure 4. The sensitivity of a detector inside a pressure vessel can be calculated from these percentage transmission curves. For the two examples of deep submergence pressure vessels, the 1 in. aluminum attenuates the ^{40}K signal 28 percent and the 1/2 in. steel housing attenuates it more than 70 percent.

The background count attributed to the NaI crystal and radioactive contaminants within the 1 in. thick housing was 15 cps.(4) In other measurements on a 3/8 in. pressure vessel,(5) the total background count rate was 12 cps. No figures are available for the background count rate to a 4 x 4 in. crystal enclosed in a 1/2 in. steel pressure vessel. It is assumed in the subsequent discussions that the background count rate is the same as an unshielded NaI crystal, namely, 3.3 cps.

A figure of merit that can be used to evaluate a particular detector package in terms of the signal count and background count is the fractional standard deviation FSD(6) when a standard deviation of the total count $\sigma(N_B + N_S)$ of the background N_B plus signal N_S in a measurement period is compared to the signal count for the same period as follows:

$$\mathrm{FSD} = \frac{\sigma(N_B+N_S)}{N_S} = \frac{\sqrt{N_S+N_B}}{N_S}$$

Expressing the formula in terms of a signal count rate S, a background count rate B, and measurement time t, it becomes

$$\mathrm{FSD} = \frac{1}{\sqrt{t}}\sqrt{\frac{B+S}{S^2}} = \frac{1}{\sqrt{t}}\sqrt{\frac{B+S}{S^2}}$$

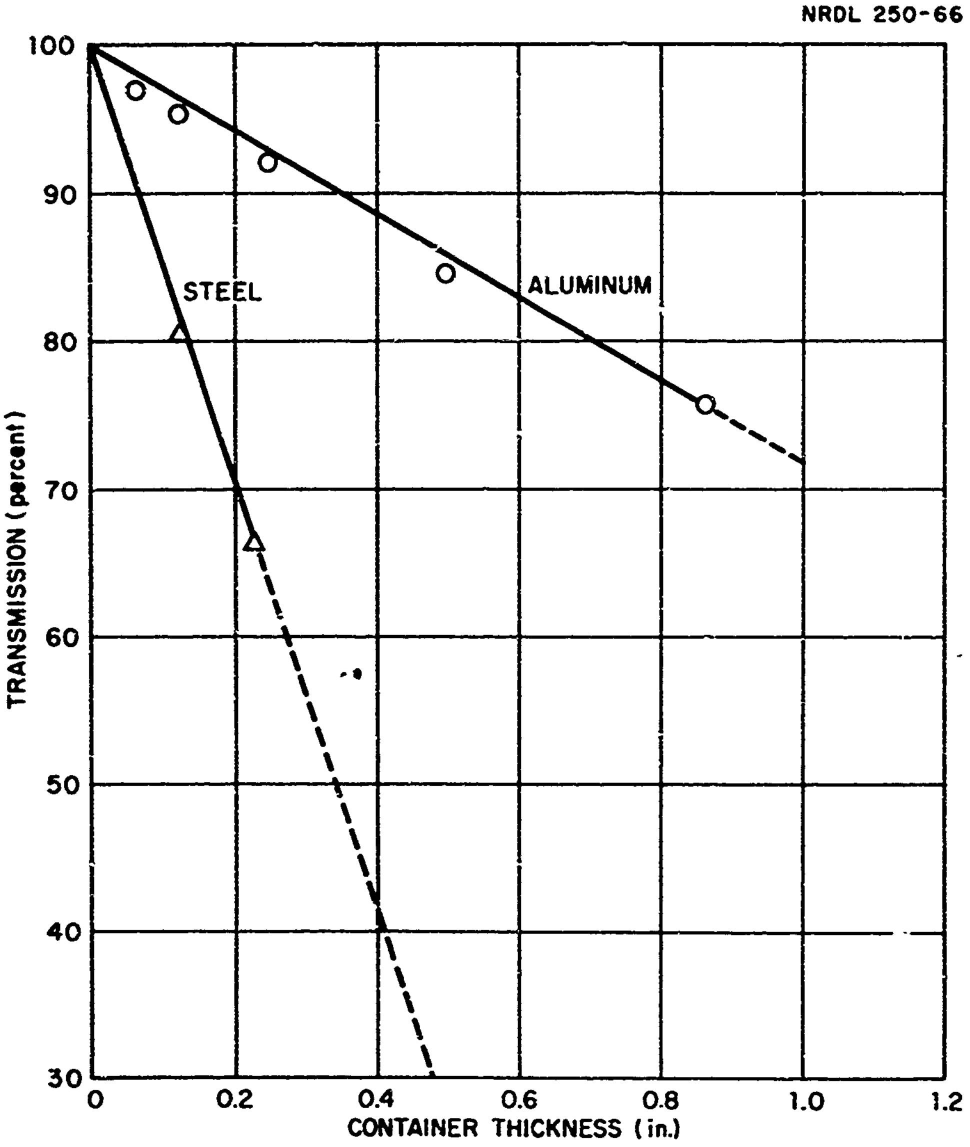

Fig. 4 Percentage transmission of integrated count from ^{40}K solution versus the thickness of shield surrounding the detector.

Using this formula, the background and sensitivity count rates and percentage transmission figures just discussed, Table 1 has been prepared.

TABLE 1

Typical Values for Fractional Standard Deviation
Assuming a One Hundred Second Count (t = 100)

Item	Detector	Housing	B (cps)	S* (cps)	$\frac{B + S}{S^2}$	FSD (%)	$\frac{\text{FSD}}{\text{FSD (CsI)}}$
1	CsI(Tl)	none	6.5	10.4	0.156	4.0	1.0
2	NaI(Tl)	3/8" Al	12	8.8	0.27	5.2	1.3
3	"	1" Al	15	7.	0.45	6.7	1.7
4	"	1/2"Steel	3.3 est	3.	0.7	8.3	2.1
5	CsI(Na)	none	3.3 est	10.4	0.126	3.6	0.9

FSD (%) is the percentage change in the signal that would increase the total count rate one σ above the total count rate before the change. It is a measure of the smallest signal that can be detected. In Table 1, using the CsI(Tl) crystal, the signal from ^{40}K would have to change 4 percent to cause the average total count rate to increase as much as one σ variation. All the other assemblies would require larger changes in ^{40}K concentration to give an equivalent increase. Using the base CsI assembly as a reference, the increase in activity to attain the same confidence level with each of the other assemblies is shown as a ratio in the last column, FSD/FSD (CsI). In the case of the 1 in. aluminum pressure vessel, the increase would have to be 70 per cent (1.7/1) greater than with the unshielded CsI. With a 1/2 in. steel case, the signal change would have to be over twice as large to give the one σ indication. The last item, CsI(Na), is added with an estimated background of 3.3 cps in anticipation of its availability. This unit would show an improvement of 10 percent over the CsI(Tl) crystal.

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* Calculated from measured sensitivity to a normal concentration of ^{40}K in sea water and the attenuation curves in Figure 4.

For completeness, alternate materials for pressure vessel for NaI might be considered. Two materials are worthy of attention-fiberglass and beryllium. In preliminary estimates of wall thickness using fiberglass, the wall would have to be 2.to 2.5 times thicker* than aluminum in designing for the same pressure depth. As a consequence the attenuation through the wall would be approximately 1.5 times that of an aluminum pressure vessel. The background from contaminates in the fiberglass would be very dependent on the glass used in construction and the contamination in glasses varies widely among the various types. Assuming no contamination, which is unlikely, the background count would be the 3.3 minimum estimate. The $(B+S)/S^2$ ratio would be 0.29 and the FSD ratio 1.35. A NaI crystal in a fiberglass vessel equivalent to the 1 in.aluminum pressure vessel would require a 35% greater increase in the activity compared to a bare CsI crystal to be detected at the same confidence level. Conversely, it would require 1.8 (0.29/0.156) times as many detector assemblies (NaI in a fiberglass pressure vessel) with no change in B/S ratio to get the equivalent sensitivity. The beryllium pressure vessel is considered to be another problem. A berllium cyclinder should not attenuate the radiation flux significantly and probably will not add to the background, however the cost of a beryllium pressure vessel is significant. Only one estimate has been obtained and that was for a hemisphere a foot in diameter for operation to 1,000 ft depths. The unit was to cost about $2,000. It is anticipated that a deep submergence pressure vessel of beryllium would be a significant cost item for each crystal.

NaI costs less than CsI. At present, a CsI crystal costs twice as much as NaI of the same size. However based on the comparisons in Table 1 between a bare CsI detector and the NaI in a 1 in. pressure housing, three times (0.45/0.156) as many NaI crystal assemblies would be required to get the same sensitivity. In this context to get a specific sensitivity, CsI is the least expensive approach.

Most of the foregoing discussion has shown that CsI is better than NaI as a scintillant for an underwater radiation detection system where a relatively thick pressure vessel is required for NaI. It is assumed throughout the discussion that CsI can be operated as a scintillant in a deep submergence system without a pressure vessel** thus eliminating

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*Present state of commercial art.

**The photomultiplier tube will be in a pressure vessel and coupled to the scintillant through an appropriate pressure tight window.

attenuation of the signal by the walls of the vessel and minimizing the background count from contaminants in materials surrounding the crystal. Based on the measured background count and response to ^{40}K distributed in water, it is shown that a detection system using CsI is significantly more sensitive than a NaI system of equivalent size surrounded by a pressure vessel. Improvement is particularly significant for deep submergence systems. In addition, CsI is more rugged than NaI and less sensitive to thermal and mechanical shock. In two areas, cost and scintillation efficiency, NaI is the better choice; however, the scintillation efficiency of CsI is being steadily improved and the cost figures should decrease as its scintillation characteristic and use increase. It remains only to try a CsI crystal under hydrostatic pressure, e.g., scintillation efficiency vs pressure, to complete the evaluation.

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