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**"LESSONS LEARNED FROM A REVIEW OF  
POST-ACCIDENT SAMPLING SYSTEMS, HIGH RANGE EFFLUENT MONITORS AND  
HIGH CONCENTRATION PARTICULATE IODINE SAMPLERS"**

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**ABSTRACT**

Post-accident sampling systems (PASS), high range gaseous effluent monitors and sampling systems for particulates and iodine in high concentrations which were installed to meet the requirements of NUREG-0737 have been reviewed at twenty-one licensee sites in Region I of the U.S. Nuclear Regulatory Commission which includes fifteen BWR's and fourteen PWR's.

Although most of the installed PASS met the NUREG-0737 criteria, the highest operational readiness was found in on-line systems which were also used for routine sampling and analysis. The detectors used in the gaseous effluent monitors included external ion chambers, GM tubes, organic scintillators and Cd-Te solid state crystals. Although all were found acceptable, each had its own inherent limitations in the conversion of detector output to the time varying concentration of a post-accident mixture of noble gases.

None of the installed particulate and iodine samplers fully met all of the criteria of NUREG-0737. Their principal limitations included a lack of documentation showing that they could obtain a representative sample and that many of them would collect of an excessive amount of activity at the design criteria.

**INTRODUCTION**

The accident at Unit-2 of the Three Mile Island Nuclear Power Station (TMI) on March 28, 1979 disclosed numerous deficiencies in the capability to collect and analyze high activity samples of its primary coolant and containment atmosphere, as well as limitations in the dynamic range of its gaseous monitors and in the adequacy of its effluent sampling systems under post-accident conditions. These systems were typical of those employed at U.S. plants at the time of the accident.

Subsequently, the U.S. Nuclear Regulatory Commission (NRC) issued short-term corrective recommendations in report NUREG-0578 (1), which included measures for the improvement of post-accident sampling capability and for the extension of the operational ranges of radiation monitors. These recommendations were finalized for implementation in report, NUREG-0737 (2). Its specific requirement for Post Accident Sampling Capability were set forth in Item II.B.3. Those for High-Range Noble Gas Effluent Monitors were set forth in Item II.F.1, Attachment 1, and those for the Sampling and Analysis of High-Range Radiiodine and Particulate Effluents in Gaseous Streams in Item II.F.1, Attachment 2.

An implementation deadline of January 1, 1982 was specified in NUREG-0737. Responsibility for post-implementation review of these systems was assigned to the NRC's regional offices. In mid-1983, Region I contracted with Brookhaven National Laboratory for technical assistance in their performance.

Starting in late 1983, on-site reviews were conducted at the rate of about one per month for the twenty-one licensee sites in Region I, which currently contain a total of twenty-nine reactors. They are located in five New England states: Maine, Vermont, Massachusetts, New Hampshire, Connecticut and New York, New Jersey, Pennsylvania and Maryland.

#### APPROACH

Prior to the on-site reviews, the individual elements needed to determine the state of operational readiness were identified using a Management Oversight and Risk Tree (MORT) which focused on the integrated ability of personnel, procedures and physical facilities to perform the acquired tasks within the time and dose constraints of Items II.B.3., II.F.1-1 and II.F.1-2. Following this, a specific set of instructions and questions related to each review component were prepared. They included categories such as design, monitoring system, shielding, structures, hardware and support services, readout and recording, staffing and training.

#### FINDINGS

##### A. Post Accident Sampling System (NUREG-0737, Item II.B.3)

The purpose of the requirement for the improved post-accident sampling capability is the prompt provision of information for the assessment and mitigation of the course of an accident. In particular, II.B.3 required chemical and radiological analyses to provide information for the assessment of core damage and reactivity control. The required analyses of containment atmosphere are intended to establish the presence and concentrations of hydrogen and airborne radioactivity.

As shown in a summary in Table 1, the installed PASS systems ranged from relatively simple licensee-designed systems which were intended solely to obtain samples for subsequent laboratory analysis, to elaborate vendor- or architect/engineer-designed systems which were intended to perform most or all of the required analyses on line.

None of the reviewed systems were adjudged perfect in every respect. However, all of those which could be fully tested at the time of the review

met the basic requirements of Item II.B.3 (three could not be tested). These included the ability to collect and to analyze samples within 3 hours or less within the General Design Criteria of 5 rem whole-body dose and 75 rem extremity dose to any individual involved.

The representativeness of the PASS coolant samples and the licensee's radiological analytical capability were tested by a comparison of their analysis with samples from the plant's normal sample sink. The accuracy of the chemical analytical capability was tested by the use of standards.

Even the relatively simple PASS systems were quite complex. Thus detailed and lengthy procedures were required to guide the operators through the sequence of steps necessary to obtain the desired samples and intensive training was required to establish operator proficiency.

The principal deficiencies, as identified during the reviews, are summarized in Table 2. Most of the findings of inadequacy of surveillance were made on the basis of the lack of a suitable schedule or excessive time intervals required to get a system back on line after an identified fault. In many instances the licensee had not verified the stipulated purge times by calculations of the volume of the sample lines.

Midway in the reviews, the NRC's Office of Inspection and Enforcement indicated that the containment atmosphere sample was intended primarily for fission gas measurements, so that sample line losses should not be considered a significant factor unless the licensee intended to use measurements of airborne radiiodines in the assessment of core damage (3). Although their usefulness for core damage assessment may not be significant, in the authors' view, these data could provide important information on the release potential if the containment should leak or fail outright following an accident.

In most cases the shielding provided for PASS systems and sample transport appeared adequate, but several licensees had not conducted a formal study to establish that the GDC-19 criteria could be met during the sequence of sampling, transport and analysis.

The balance of the listed deficiencies and the measures necessary to address them should be self-explanatory.

## B. Noble Gas Effluent Monitors

A summary of the installed high-range noble gas monitors, according to their location (on-line or off-line), type of detector, and fabricator/vendor is shown in Table 3. As indicated, two licensees installed on-line monitors, using ion chambers which were located in or immediately adjacent to stacks or ducts, while eighteen installed off-line monitors. Of the latter, six installed "gas only" high-range monitors as additions to their pre-existing low-range monitors. Twelve licensees installed commercially available monitors with modules for monitoring and for sampling. A view of a typical one (the Kaman KDGm-HR) is shown in Figure 1.

Typically, three overlapping-range detectors are provided to achieve the required full-range sensitivity. In order to achieve an upper limit of  $10^5$   $\mu\text{Ci}/\text{cm}^3$ , most of the vendor designed monitors have high-range detectors which

view a limited volume of gas (as compared to that viewed by their mid- or low-range detectors).

Item II.F.1-1 was not specific on the calibration of noble gas monitors up to the required upper range. However, the NRC provided guidance which recognized the problems of the availability of suitable noble gases, i.e.  $^{133}\text{Xe}$  in sufficient concentrations and of their utilization by licensees. The NRC Staff recommended that a one-time "type" calibration in the laboratory at three values separated by two decades over the intended range be performed and that the transfer procedure of ANSI N323-1978 be utilized in conjunction with solid sources at appropriate energies for on-site calibrations (4).

As indicated in Table 4, most of the vendors appeared to have performed only a "one point" primary calibration, utilizing  $^{133}\text{Xe}$  and/or  $^{85}\text{Kr}$ . They then performed transfer calibrations with solid sources with a range of activities and energies, to establish the energy response and linearity over the full range capability of a given detector.

A summary of the sampling arrangements which were provided to achieve compliance with Item II.F.1-2 is shown in Table 5. Most of the licensees who utilized "gas only" monitors to comply with Item II.F.1-1 installed independent sampling facilities which included additional shielded particulate and iodine sample positions which were connected to an existing low-range sample line. Eleven licensees installed integrated monitor/samplers which included microprocessor controlled sampling modules that provide for the automatic or remote collection of a sample at one of three individual sample positions, (as also depicted in Figure 1). These integrated monitor-samplers typically provide for a much reduced flow of a few hundred  $\text{cm}^3/\text{min}$  for high range sampling, as compared to the 1-2 cfm flow that is typically provided for low- and mid-range sampling. The intent is to thereby limit the total amount of activity that would be collected at concentrations which approach the upper design criterion of  $100 \mu\text{Ci}/\text{cm}^3$  for the stipulated 30-minute sampling period.

At PWRs, the NUREG-0737 requirements also included the monitoring of secondary side steam effluents which might be released through safety and relief valve discharge lines. Externally mounted monitors viewing the main steam line upstream of these valves were acceptable. Of the fifteen PWR's (on eleven licensee sites) which were reviewed, all but two had installed the required steam line monitors. Six utilized ion chambers and seven utilized shielded GM detectors.

## LESSONS LEARNED

### A. High-Range Noble Gas Monitors

Oversimplifications in the conversion of the direct indications of the installed gas monitor (typically in cpm or mR/hr) to effluent concentrations and/or rates of release were among the principal shortcomings encountered in the reviews.

Item II.F.1-1 states in part " $^{133}\text{Xe}$  equivalent values for monitors employing gamma radiation detectors" (as most do). This concept has not been fully understood or employed by all vendors and licensees. In several instances, they were found to have directly

employed raw calibration data for  $^{133}\text{Xe}$  or  $^{85}\text{Kr}$  to establish detector response, without a recognition of their limitations. The former emits low energy photons which may be significantly absorbed in the housing or walls of a detector. In contrast,  $^{85}\text{Kr}$  is principally a beta emitter, with accompanying bremsstrahlung gamma radiations and a 0.51 MeV photon with a yield of only 0.4%. Figure 2, illustrates the direct response with distance of Eberline's high-range detector to each nuclide. When corrected respectively for absorption and bremsstrahlung, its true energy response is about midway between the two curves, so that using one point from either could lead to a factor of two error.

Uninterpreted calibration data have also been employed to calculate release rates (in Ci/sec) without regard to the variable energy response characteristic of detectors. This may be close to linear with energy, as shown in Figure 3, for the Kaman KDGM-HR or may be quite non-linear, as shown in Figure 4 for the General Atomics WRGM.

A slight fall-off from linearity of a GM tube detector at the high count rates corresponding to a concentration of  $10^5 \mu\text{Ci}/\text{cm}^3$  was evident. The Kaman high-range detector was modified after a field test with  $^{133}\text{Xe}$  had indicated that an earlier configuration could not meet the requirement (5). A similar fall-off in a field calibration of Eberline's high-range detector (SA-9) was reported by a consultant to a Region I licensee (6). This fall-off appears to be amenable to correction, once it is recognized.

The micro-processor of the Eberline SPING-4 monitor is not radiation hardened. Thus it cannot be assured that it would operate reliably in high post-accident radiation fields. However, Eberline recommends that it be supplemented by other detectors, for which the sensitive components are remotely located (7).

The reviews revealed that several of the licensees with installed micro-processor controlled monitors had experienced failure of their flow sensors which disabled their automatic features. These sensors appear to be sensitive to entrained dust particles and thus need frequent preventative maintenance.

Except for those with installed integrated units which function automatically, few licensees had incorporated provisions or procedures for the isolation and/or purging of their low-level gas monitors, should their range be exceeded. Thus their recovery and availability would be doubtful as effluent concentrations declined to within the low-range region following an accident.

While the steam-line monitors at PWRs were relatively simple and straightforward devices, their detectors were of necessity installed in hostile environments where they were subject to heat and humidity. They therefore require frequent maintenance (especially the GM detector devices). Most of the licensees had accepted the vendor's calibrations and very few had analyzed the effect of the wall thickness of the steam line pipe to account for detector response to low-energy radiation.

## B. Sampling and Analysis of Plant Effluents

The principal deficiency encountered in the review of arrangements for the sampling of radioiodines and particulates was the inability of licensees to document that their sampling systems could collect representative samples.

This is particularly so for those with long sampling lines, in which considerable deposition losses of elemental radioiodines could occur even when installed in accordance with the design guidance of ANSI N13.1-1969. This was recognized by the NRC with the recommendation that licensee's make an empirical determination of sampling line losses (4).

The transmission of elemental iodines through long sampling lines has been measured in the laboratory by Unrein et al (8). Their studies suggest that transmission and collection by the sampler depends upon the relative rates of deposition and resuspension from sampling line walls. Transmission factors greater than 50% were found for 1" OD sampling lines at flow rates of 2-3 cfm, for injection periods of several hours. However, these studies did not indicate how long it took to reach equilibrium after an initial injection. Only a small fraction (<1%) of the injected elemental iodine was transmitted through the 1/4" OD sampling line with a 0.06 cfm flow rate as originally utilized in the General Atomics WRGM.

The NRC's proposed guidance suggests that the closest approximation to representativeness may be achieved at equilibrium, when deposition and re-entrainment or re-suspension are equal. This could be expected to occur most rapidly in a continuously operated system, rather than one in which flow is initiated only upon the occurrence of high-range concentrations. The Kaman and the Eberline AXM-1 monitors incorporate this feature. Upon an indication of abnormal gas concentrations, they isokinetically obtain a small local side-stream flow of a few hundred  $\text{cm}^3/\text{min}$  from the low-range monitoring/sampling line, in which the normal flow of 1-2 cfm is maintained. Recently installed WRGM's have included isokinetic flow-splitters installed in the high-flow sampling line close to the low-flow accident range sampling module.

All licensees were apparently aware of the need for the heat tracing of sampling lines when exposed to "outdoor" conditions. However, some have not recognized a similar need for the heat tracing of long indoor horizontal sampling lines in which condensation could occur, especially under the high moisture loads or loss of building climate control under some accident sequences.

Although II.F.1-2 calls for capability for continuous sampling, the procedures of five licensees called only for the analysis of a grab sample to be collected post-accident over a short period of time (to limit the amount collected to the capability of their laboratory GeLi analysis systems), with no indication of how they would evaluate the preceding sample to establish the total amount released from the onset of accident conditions.

In several instances, which included the three SPING-4s, the three RAGEMS and one licensee devised installation, the filter assembly for the collection of particulates and iodines was either unshielded or inadequately shielded. None had conducted an analysis to assure samples could be collected, retained and transported within the GDC-19 dose limits. It should be noted that the SPING-4 was designed with a nominal sampling rate of 1-2 cfm, so as to achieve sensitivity for routine monitoring. By two successive 1/200 dilutions, the RAGEMS should collect only relatively low activity samples.

Only a few licensees had developed adequate procedures for the analysis of high activity samples which might considerably exceed the upper limit which

could be analyzed by their GeLi counting and analysis systems. Several had established procedures for counting samples in a geometry distant from the detector, but would still be unable to cope with samples approaching the 85-170 Ci of radioiodines which would be collected at a concentration of 100  $\mu\text{Ci}/\text{cm}^3$  at normal flow rates of 1-2 cfm for the stipulated 30-minute sampling period.

#### COMMENTS AND RECOMMENDATIONS

Most of the PASS systems which were encountered required frequent and considerable attention to keep them fully operational. All required frequent retraining to maintain operator proficiency. The example of the one in-line system which is also used for routine sampling suggested that the readiness of the other systems could be enhanced if they are periodically used for routine sampling.

If the NUREG-0737 requirements were solely for noble gases, ion chambers would be the most straightforward detectors, in view of their simplicity, wide range capability, and linear energy response characteristics. Unfortunately, they are relatively insensitive and require a large volume of contained gas which is difficult to shield from extraneous radiations.

The integrated monitoring/sampling devices that incorporate microprocessor data handling and control accomplish the full range requirements of Item II.F-1.1 by routing the flow to more than one detector, each of which is designed to be sensitive to portions of the full range requirement. This facilitates the isolation of low-range detectors during periods of high concentrations and the routing of flow successively to selected filters to limit the collected activity. The capability to store and to provide a history of release rates over time makes them attractive for routine and accident monitoring.

Much of the confusion over the use of the " $^{133}\text{Xe}$  equivalent" concept in the calibration of high-range noble gas monitors could be eliminated by the adoption of the "Ci-Mev" concept as has been described by Mourad (8). A simplified version of the same concept was described by Lahti et al at the 1986 Annual Meeting of the Health Physics Society (9).

To minimize ambient post-accident radiation fields in their vicinity, most post-accident monitors and/or samplers were located at considerable distances from the points of effluent release, thus necessitating long sampling lines (typically 1" OD x 100-250'). This creates a dilemma between the desirability of maintaining a high flow rate in the sample line to minimize deposition losses and the desirability of minimizing the amount of collected radioactivity on the sampler. Several vendor designed monitors provide a second stage of isokinetic sampling, with a probe situated within the high-flow line close to the sampling head and with a much smaller flow (a few hundred  $\text{cm}^3/\text{min}$ ) through the "high-concentration" sampler. This seems desirable on the grounds of both convenience in handling and analysis and of ALARA considerations.

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5. D. McClure, "Accident Range Monitor Response" HPS Newsletter, 18:6 pg. 7 (1985).
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10. G.P. Lahti et al., "Effluent Monitor Corrections for Use in Emergency Planning", Presented at 31st Annual Meeting of the Health Physics Society, Pittsburgh, PA, June 1986. Author's address: Sargent & Lundy, 55 East Munroe, Chicago, IL 60603.

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Appreciation is also appropriate for the typist Marie Cooney.

Table 1 Summary of Installed Post-Accident  
Sampling Systems

Sample Collection (no on-line analysis capability)

<u>Design</u>	<u>No. Sites (Reactors)</u>	
Licensee	4	(4)
G.E.	9	(12)

Sample collection (limited on-line analysis capability)

<u>Design</u>	<u>No. Sites (Reactors)</u>		<u>Remarks</u>
General Dynamics	2	(4)	In-line-pH, Cond.
Quadrex	1	(2)	In-line-pH, B, Cl
Sentry	1	(2)	In-line-pH, Cond, Dis O <sub>2</sub> , Dis H
	1	(1)	In-line-pH, Cond, DO <sub>2</sub> , Dis H, Cl
Stone & Webster	1	(1)	In-line-isotopic analysis pH, B, Cl

Full in-line analysis capability (including isotopic)

<u>Design</u>	<u>No. Sites (Reactors)</u>	
Sentry	1	(1)
Combustion Eng.	1	(2)

Table 2 Principal Deficiencies Identified in Review of  
Post-Accident Sampling Systems

<u>Frequency</u>	<u>Deficiency</u>
13	Inadequate surveillance and maintenance program
11	Inadequate purge times
11	Non-representativeness of radioiodines in containment air sample
11	Inadequate time and motion studies to document that shielding in sample room and/or of sample during transport sufficient to enable operation within GDC-19 criteria
8	Improper pressure and/or temperature corrections
8	Procedures inadequate or in need of revision to conform to actual operation of PASS
5	Dilution beyond the range capability of the analytical procedure
6	Moisture carry over into gas chamber during gas stripping
4	Inadequate assurance of sample flow (no flow meter installed)
4	Insufficient or no backup for one or more in-line analyses
4	Inadequate test of all features of system by licensee prior to on-site review
4	Volume delivered by ball valve (for dilution) not established by actual measurement
3	Inadequate training or insufficient number of trained personnel to assure ability to operate system during post-accident conditions
3	Inadequate assurance that sample could be obtained when reactor depressurized (no pump in PASS)
3	Needle bent during attempt to perforate septum of sample collection vial
3	Improper interpretation of flow produced by critical orifice (or of pressure required to maintain design flow)
3	Unsuitable cask/shield vial for sample transport
3	Chemical analysis procedure not adequately tested for possible interferences
2	Sample not returned to containment

Table 3 Summary of Installed Mid- and High-Range Noble Gas Monitors

<u>No.</u>	<u>Range</u>	<u>Detector</u>	<u>Vendor</u>	<u>Model</u>	<u>Operating Mode</u>	<u>Data Processor</u>	<u>Background Subtraction</u>
<b>Sites (Reactors)</b>							
<b>On-Line</b>							
2(2)	Mid/High	Ion Chamber	(1) GA (1) Victoreen	RD-2A 847	Continuous	No	No
<b>Off-Line</b>							
<u>Gas Only</u>							
1(2)	Mid/High	Plastic	NMC	GA-270	High Alarm	No	No
1(1)	Mid High	GM Ion Chamber	Victoreen Victoreen	847	Continuous	No	No
3(3)	Mid/High	Ion Chamber	Victoreen	847	Continuous	No	No
1(1)	Mid/High	Ion Chamber	Reuter-Stokes	C4-2510-101	High alarm	No	No
<u>Integrated Gas Monitors and Particulate-Iodine Samplers</u>							
6(8)	Mid High	Cd-Te Cd-Te	GA	WRCM	High Alarm	Yes	No
3(4)	Mid High	GM GM	Eberline	SPING-4	Continuous	Yes	Yes
2(4)	Mid High	GM GM	Kaman	KGM-HRH	High Alarm	Yes	No
1(2)	Mid/High	Ge-Li	SAI	RAGEMS	Continuous	Yes	NA
1(2)	Mid High	GM GM	Eberline	AXM-1	High Alarm	Yes	Yes

Table 4 Concentrations for Vendor Calibrations  
of II F.1-1 High Range Monitors

	$^{133}\text{Xe}$ Concentrations $\mu\text{Ci}/\text{cm}^3$	$^{85}\text{Kr}$ Concentrations $\mu\text{Ci}/\text{cm}^3$
<u>Eberline</u>		
Mid-Range SPING NGD-1 (SA-13)	0.13*	1.47
High-Range SPING AXM-1(SA-14)	0.26	1.47
SA-15, SA-9	2.06*	9.98
<u>General Atomics</u>		
Mid/High Range-WRCM	$\sim 1.0^{**}$	$9.6^{**}$
<u>Kaman</u>		
High-Range-HRH	$\sim 1 \times 10^2^{**}$	

\* Peak concentration of successive calibrations

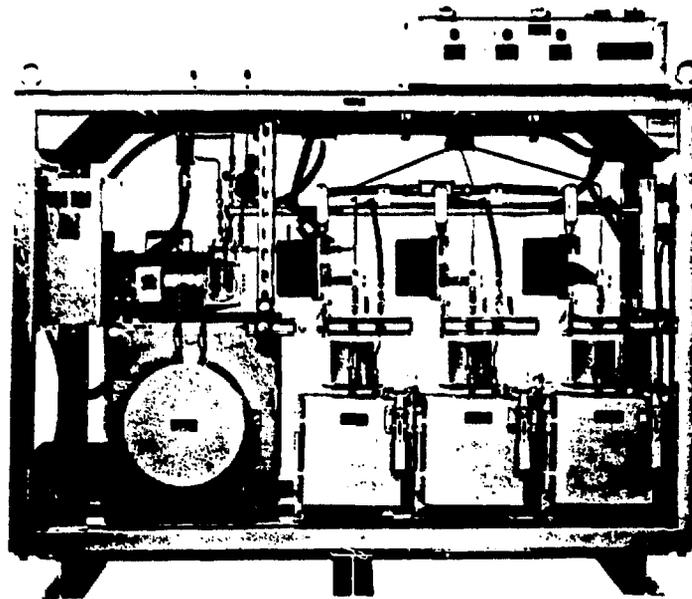
\*\* Based on calibration data supplied by vendor and as inferred from NBS Reference Date.

Table 5 Sampling and Analysis of Plant Effluents, II.F.1-2

Independent Utility Design

<u>No.</u>	<u>Range</u>	<u>Vendor</u>	<u>Model</u>	<u>Sample Positions</u>	<u>Shielded</u>	<u>Filter Selection</u>	<u>Remarks</u>
5 (5)	-	-	-	1	Yes		(In each instance)
<u>Vendor Design</u>							
1 (1)	-	NRC Ind.	MAP-5	3	Yes	Local/remote control	Timed sample
1 (2)	-	Kaman	HRH	1	Yes		
<u>Integrated Units</u>							
5 (7)	Mid/High	GA	WRGM	3	Yes	Local/remote control	Timed sample
3 (4)	Mid/High	Eberline	SPING-4	1	No	Fixed	
2 (4)	Mid/High	Kaman	KGM-HRH	3	Yes	Automatic (GM Monitor)	Automatically timed sample
3 (4)*	All	SAI	RAGEMS	1*	Yes	Automatic	Note 1*
1 (2)	Mid/High	Eberline	AXM-1	1	Yes	Fixed (GM Monitor)	

\* One licensee had installed this system, but did not utilize its Ge-Li detection feature.



HIGH RANGE

Figure 1. Kaman HRH High-Range noble gas monitor and sampler.

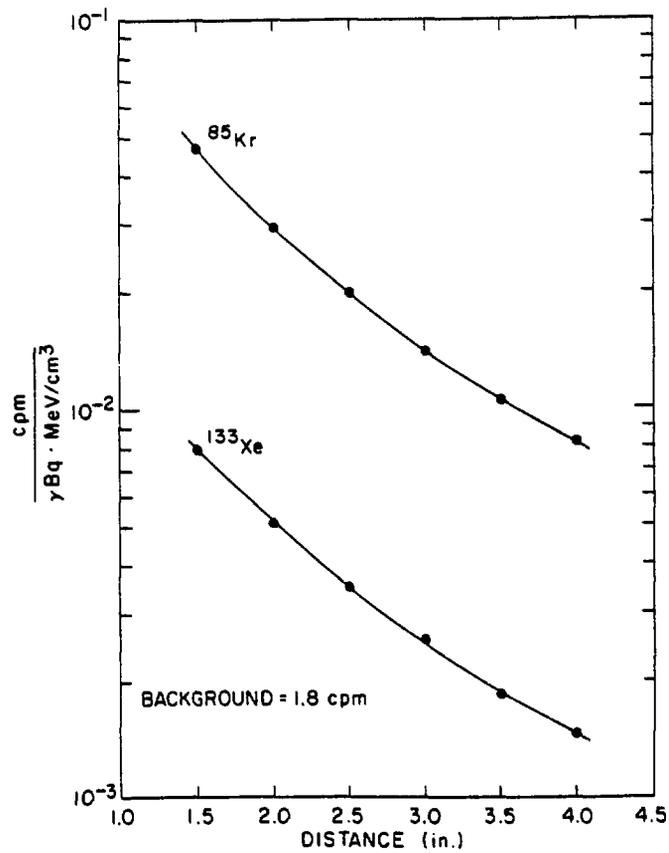


Figure 2. Response of Eberline SA-9 high-range detector to  $^{85}\text{Kr}$  and  $^{133}\text{Xe}$

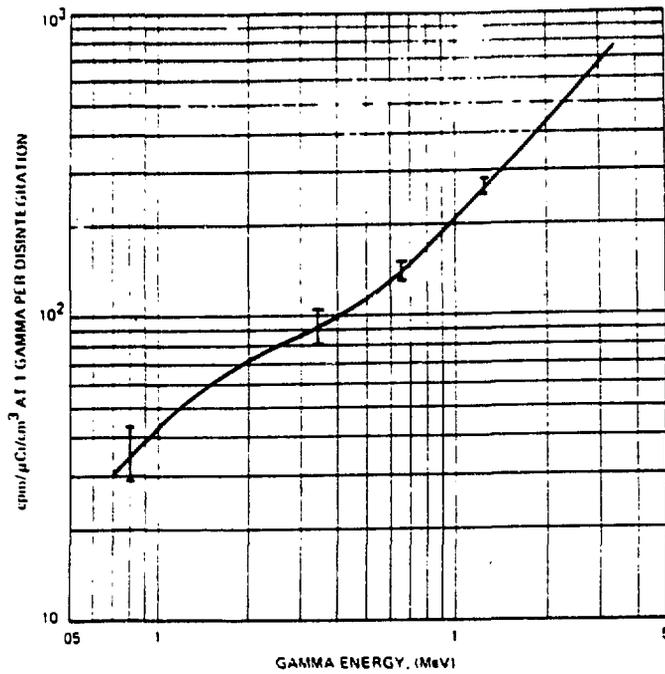


Figure 3. KDGM-HR enhanced detector in KSG-HRH sampler, enhanced high-range energy dependence characteristic.

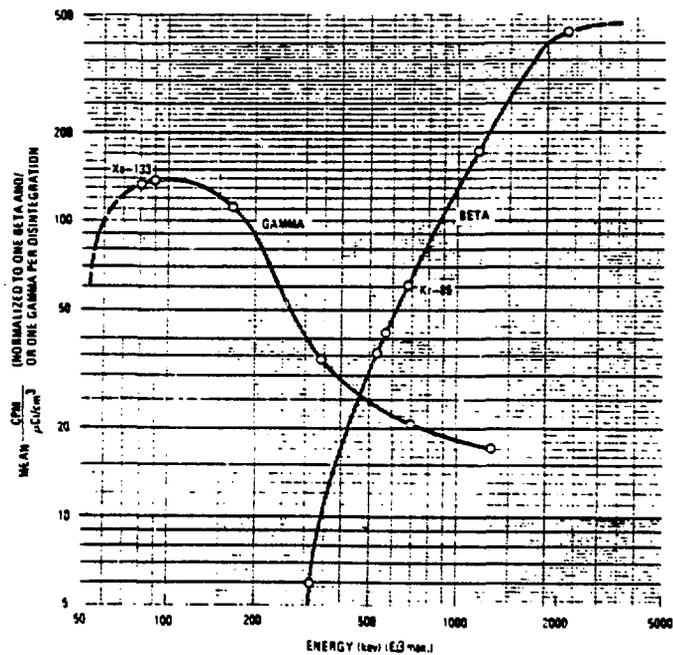


Figure 4. General Atomics wide-range gas monitor RD-72 high-range detector energy response curve.