

AN ABSTRACT OF THE THESIS OF

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The 1979 incident at Three Mile Island drew attention to weaknesses in the capability to obtain and analyze reactor coolant samples in an environment where the reactor core incurs significant damage. As a result, the U.S. Nuclear Regulatory Commission promulgated regulations requiring that reactor coolant analytical capabilities be upgraded so they could function in such an environment. The "upgrades" primarily involved the ability to safely obtain a representative sample within the count rate capabilities of the detection system and to deduce useful information from the analysis. Different designs and techniques were developed for fulfilling these regulatory requirements; however, the constraints in an environment of severe core damage make it very difficult to test the feasibility of specific design features and to determine the type of information that would be most useful to operators.

To examine issues associated with post-accident monitoring, the Electric Power Research Institute funded a research project, the scope of which included, among other items, four specific

radiological factors associated with routine and post-accident monitoring. The study of these four factors, evaluations of on-line monitoring versus grab sample analysis, count rate reduction factors obtainable by and spectral effects of variable aperture collimation, use of key radionuclides in assessing fuel pin integrity under both normal and abnormal reactor conditions, and possible spectral interferences from fission product releases at elevated core temperatures, provided the substance for this research report. A description of the four individual studies along with results are reported in four separate chapters. A conclusions chapter is used to summarize the key findings of the research effort.

Routine and Post-Accident Analysis of  
Nuclear Power Plant Reactor Coolant

by

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## TABLE OF CONTENTS

	<u>Page</u>
INTRODUCTION	1
RELATIVE ADVANTAGES AND DISADVANTAGES OF ON-LINE MONITORING AND GRAB SAMPLE ANALYSIS TECHNIQUES	4
Introduction	4
Dual-purpose Systems	7
Data Acquisition	9
Sample Representativeness	9
Spectral Quality	17
Real-time Analysis	19
Flexibility of Analysis	20
Frequency of Analysis	21
High Count Rate Capability	22
Collimation	25
Variable Geometry	28
Dilution	29
Sample Diversion and Holdup	31
Matrix Separation	32
Conclusions for Count Rate Reduction Techniques	33
Ambient Environmental Effects	34
Health Physics Considerations	40
System Maintenance, Repair and Calibration	42
Cost	46
General Conclusions	47
THE TECHNIQUE OF COLLIMATION IN REDUCING HIGH COUNT RATES	50
Introduction	50
Materials and Instrumentation	51
Percent Transmission	59
Percent Peak-to-Total Ratio	60
Conclusions	70
THE ASSESSMENT OF CORE CONDITIONS BY REACTOR COOLANT ISOTOPIC ANALYSIS	73
Introduction	73
Theory	74
Additional Indicators of Core Conditions	75
Source Term	76
Fission Product Chemistry	76

	<u>Page</u>
Release Mechanisms	80
Mass Balance	83
Complicating Factors	84
Multiple Fuel Failures	85
Failure Development with Time	86
Irradiation History	87
Monitoring Techniques	89
Background Radioactivity	90
Data Base	95
Procedures for Assess Core Conditions	96
"Key" Radionuclide Characteristics	98
Specific Radionuclides	100
The Halogens	100
The Noble Gases	102
The Cesiums	103
Computer Models	104
Indicators of Cladding Failures Under	
Normal Operating Conditions	104
Indicators of Elevated Core Temperatures	108
Indicators of Core Melt	114
SPECTRAL INTERFERENCES	117
Introduction	117
Computer Study	121
Conclusions	125
CONCLUSIONS	134
On-line Monitoring versus Grab Sample Analysis	134
The Technique of Collimation in Reducing High	
Count Rates	135
Key Radionuclides	136
Spectral Interferences	139
BIBLIOGRAPHY	140

## LIST OF FIGURES

<u>Figures</u>		<u>Page</u>
III-1	Collimator Set-up	54
III-2	Dependence of % Transmission on Collimator Diameter	62
III-3	Dependence of % Transmission on Energy	63
III-4	Dependence of Reduction Factor on Collimator Diameter	65
III-5	Dependence of % Peak to Total Ratio on Collimator Diameter	68
III-6	Dependence of % Peak to Total Ratio on Energy	69



## LIST OF TABLES

<u>Table</u>	<u>Page</u>
II-1      Summary Comparison of Relative Advantages of Grab Sample Analysis and On-line Monitor- ing Systems	49
III-1     Calculation of Expected $^{152}\text{Eu}$ Activity	52
III-2 $^{152}\text{Eu}$ Decay Characteristics	57
III-3     Percent Transmission	61
III-4     Reduction Factors	64
III-5     Percent-Peak-to-Total Count Rate Ratios	67
V-1       Key Radionuclides Related to Reactor Core Condition	121
V-2       Equilibrium Core Inventory	124
V-3       Gamma Ray Emission Rates of Key Radionuclide Fission Products	126
V-4       Extent of Spectral Interferences for Key Radionuclides	131

## PREFACE

In the aftermath of the Three Mile Island Unit 2 accident in 1979, the U. S. Nuclear Regulatory Commission (NRC) promulgated regulations requiring that post-accident monitoring capability be expanded in nuclear power plants. The short time frame mandated for implementation of the post-accident monitoring requirements did not allow for full investigation of all issues related to the implementation of the new requirements. As a result, the Electric Power Research Institute (EPRI), an organization involved with promoting and financing practical research of interest to electric utilities, funded a project in which issues related to the evaluation, selection and effective utilization of post-accident monitoring systems were examined in detail.

The issues specifically addressed in the EPRI study include the following:<sup>1</sup>

1. Do the NRC requirements adequately cover post-accident information needs, or conversely, are they excessive?
2. Do commercially available post-accident monitoring systems meet NRC requirements?
3. To what extent can post-accident monitoring systems be used in routine monitoring?
4. What level of sophistication should a utility seek to attain in its post-accident monitoring equipment? What

are the relative advantages and disadvantages of the different levels available?

5. What approaches should be explored to improve the usefulness and capabilities of post-accident monitoring systems?

The team participants for the EPRI study included researchers from Sargent & Lundy Engineers and Oregon State University (OSU). Oregon State University's team covered both radiological and chemical aspects of post-accident monitoring.

The chemical research group at OSU analyzed the applicability, merits, and shortcomings of various techniques for analyzing post-accident concentrations of boron, chloride and pH.

The radiological research group at OSU performed several experiments, examined pertinent literature in detail, and developed a prototype of a fully automated (under minicomputer control) post-accident monitoring system. The scope of the project for the radiological group also involved extensive communication with several vendors of post-accident monitoring systems, with utilities, and with national engineering laboratories. The laboratory experiments conducted by the radiological group included collimation techniques for reducing high count rates, determination of the optimal maximum count rate and the lower limit of detection achievable using a conventional germanium detector counting system, and automated chemical separations of fission products. Also evaluated by the radiological group were interferences that may occur in spectra of the primary

reactor coolant following fission product releases from fuel at the onset of conditions leading to core degradation. The literature reviews covered on-line monitoring and grab sample analysis techniques, specific system designs marketed by vendors, systems in use by some research organizations and nuclear power utilities, and techniques for diagnosing the fuel cladding integrity by evaluating specific "key" radionuclide concentrations in the reactor coolant.

This research report examines four topics associated with post-accident reactor coolant monitoring. Each topic is covered in a separate chapter.

# ROUTINE AND POST-ACCIDENT ANALYSIS OF NUCLEAR POWER PLANT REACTOR COOLANT

## CHAPTER I.

### INTRODUCTION

In the aftermath of the Three Mile Island Unit 2 (TMI) accident, the U.S. Nuclear Regulatory Commission organized a "Lessons Learned" Task Force in May 1979, which was charged with identifying and evaluating safety concerns brought to light by the incident and identifying those which should be considered for specific licensing action.<sup>2</sup> The task force concluded that the TMI primary reactor coolant sampling and analysis equipment was inadequate for operation in the environment encountered during the accident. It further concluded that probably most other existing nuclear power plant systems for sampling and analyzing primary coolant had similar deficiencies. It subsequently recommended that reactor coolant monitoring capability be reviewed and upgraded to enable on-site analysis of highly radioactive reactor coolant.

The problems encountered at TMI in the analysis of the reactor coolant involved high radioactivity in the reactor coolant, high background radiation due to direct shine from the core and airborne radioactivity. These factors resulted in radiation dose rates too high to allow personnel to remain on site, and furthermore the

unusually high levels of background and sample radioactivity were too great to allow adequate counting instrumentation performance. As a result, a reactor coolant sample had to be shipped to a Department of Energy facility for analysis, which caused a significant delay of information to the plant operators.

Prior to the accident, no definitive requirements existed for obtaining and analyzing reactor coolant samples under severely degraded core conditions.<sup>3</sup> Based on the Task Force findings and recommendations, the NRC promulgated regulations requiring that reactor coolant monitoring capabilities be upgraded according to specified criteria. The basic guidelines on required system capabilities are contained in two major documents; general requirements are provided in NUREG-0737,<sup>4</sup> and additional guidelines are provided in NRC Regulatory Guide 1.97, Revision 2.5. Where pertinent to this report, specific requirements set forth in these documents will be outlined.

The short time frame for implementation of the post-accident monitoring requirements did not allow for full investigation of all issues related to the new systems before their broad application. This report evaluates several key issues pertinent to the new NRC requirements and the application of post-accident monitoring systems.

After this brief introduction to the current situation surrounding post-accident monitoring, which comprises Chapter I, Chapter II covers an analysis of the relative advantages and disadvantages of on-line monitoring techniques and grab sample analysis. Chapter III

describes an empirical evaluation of the technique of variable collimation in the reduction of high count rates, an analysis of collimator induced spectral effects and the reduction factors obtainable by collimation. Next, Chapter IV discusses how information obtained from reactor coolant isotopic analysis may be used in assessing the integrity of fuel pins and overall core condition under both accident and routine nuclear power plant operations, while Chapter V describes a computer study evaluating possible significant gamma spectral interferences from competing fission products that may be released from a degraded reactor core. The final chapter, Chapter VI, provides a brief summary of the work performed along with pertinent conclusions.

## CHAPTER II

### RELATIVE ADVANTAGES AND DISADVANTAGES OF ON-LINE MONITORING AND GRAB SAMPLE ANALYSIS TECHNIQUES

#### Introduction

Prior to the TMI accident, reactor coolant monitoring systems had used grab sample analysis alone and were intended for routine sampling conditions only. Indeed, prior to the TMI incident no definitive regulatory requirements existed for obtaining and analyzing reactor coolant samples following a core-melt accident, although the NRC staff and the Advisory Committee on Reactor Safeguards had for some years emphasized the need for special features and instrumentation to aid in accident diagnosis and control.<sup>3</sup> Some degree of post-accident sampling capability was available at TMI; however, experience revealed that more was needed. As a result, the NRC promulgated regulations requiring that systems be upgraded according to specified criteria. The new requirements are basically designed to ensure that critical data are obtainable in a safe, timely, correct and unambiguous manner.<sup>6</sup>

Post-accident monitoring systems refer to any type of system designed to have the capability of analyzing reactor coolant under conditions of severe core degradation.

Post-accident conditions refer to circumstances concurrent with and following an accident involving at least partial core melting.



Such an accident scenario is specified in Section II.B.2 of NUREG-0737.<sup>3</sup>

Accident conditions under which post-accident monitoring systems are designed to operate use NRC Regulatory Guide 1.3 or 1.4 releases of fission products resulting from an assumed design basis loss of coolant accident for light water reactors.<sup>7,8</sup> Furthermore, specific guidelines for post-accident monitoring systems are outlined in NRC Regulatory Guide 1.97, Revision 2, and in NUREG-0737. Additionally, current NRC requirements for post-accident monitoring include the capability for chemical analysis; however, this study will evaluate post-accident reactor coolant monitoring systems only with respect to radiological analysis and will reference specific NRC guidance only when pertinent to the topic being discussed.

In the apparent absence of systems designed specifically for post-accident monitoring, several vendors rushed to capitalize on the new NRC requirements. Various types of post-accident monitoring systems were designed, incorporating different means of fulfilling NRC criteria and handling the unique problems associated with post-accident monitoring. A period of rapid development in the technology of post-accident reactor coolant sampling and analysis followed. Until recently, the feasibility of using on-line reactor coolant monitoring techniques had not been favorable. However, significant advances in instrumentation for on-line gamma ray spectrometry facilitated the development of post-accident on-line monitoring systems.<sup>9</sup> Thus, the post-accident monitoring systems designed ranged

from fully automated, computer-controlled, on-line monitoring systems of relatively high complexity to comparatively simple systems for manually obtaining grab samples.<sup>10</sup>

The short time frame for implementation of the post-accident monitoring requirements did not allow for a full investigation of the relative merits of the new monitoring systems being developed and marketed. Therefore, a study was initiated to evaluate the advantages and disadvantages of the two broad types of post-accident monitoring systems, on-line monitoring systems (OLS) and grab sampling systems (GSS), and the results were incorporated into this chapter.

In this study a grab sampling system is defined as a system which collects a grab sample of the primary reactor coolant and subsequently analyzes it at a separate location. A grab sampling system includes the sample acquisition system and all the separately located instrumentation for gamma ray spectrometry. Although a grab sampling system can be designed to automatically acquire reactor coolant samples, the GSS considered here operate manually, as is the case for most available systems.

In this study an on-line monitoring system is defined as an analytical system which views a reactor coolant let-down line directly by using a germanium detector to generate a signal for transmission to a separately located multichannel analyzer. The OLS considered here operate automatically under computer-based control. An on-line monitoring system can theoretically be designed to view the

main reactor coolant line by being located inside of the containment structure; however, such systems have unique problems (and advantages) not applicable to similar systems that only view let-down lines, and will not be considered in this study.

A generic comparison of the advantages and disadvantages of GSS and OLS is not always particularly applicable or conclusive because of specific design variations within these two broad categories which influence each specific system's merit. For example, certain problems and advantages are only applicable to specific designs; however, some clear differences exist between the two broad system types, and these are addressed below. System merits will be viewed in terms of applicability to both routine and post-accident monitoring.

### Dual-purpose Systems

Both GSS and OLS can be designed to function as routine and/or post-accident monitoring systems. Benefits associated with dual-functioning designs generally outweigh stand-alone systems. The concept of incorporating both functions into one system is obviously desirable, since the need for post-accident monitoring is not very likely and a stand-alone post-accident system is essentially nonfunctional except as a contingency item.

The incorporation of both routine and post-accident analysis capability into one system is desirable from several standpoints. A dual-functioning system ensures that the operating personnel are familiar with its operation, thus decreasing the tendency for human

error.<sup>11,12</sup> This consideration is especially important in the collection and transport of highly radioactive grab samples. In addition, familiarity with operating procedures also decreases the frequency of needed refresher training.<sup>13</sup> Dual-functioning systems also provide familiarity with typical routine reactor coolant fission product concentrations. This familiarity will enhance confidence in the validity of abnormal results, thus decreasing the tendency to discount unusually high fission product concentrations as erroneous values.

With dual-functioning systems, proper conditioning processes are also accomplished during routine monitoring. For example, for optimal performance with any monitoring system, valves and seals need to be wetted on a weekly basis. Sample lines also need to be seasoned and conditioned to minimize the possibility of serious crud plateout that can plug lines and block flow.<sup>12</sup>

In somewhat of a contrast to the theory stated above, it was found at the Trojan Nuclear Plant that if the post-accident grab sampling system had also been designed for use in routine sampling, the necessary modifications incorporated to reduce personnel dose during post-accident sampling and analysis would have made routine sampling much more cumbersome.<sup>14</sup> Consequently, separate post-accident and routine monitoring systems were designed with grab sampling points in close proximity and with virtually identical operating procedures. If post-accident sampling is required, a simple valve manipulation diverts the sample flow to the post-

accident monitoring system, with the advantage that the operator is already familiar with the procedures required to sample the reactor coolant.

Whether the incorporation of a stand-alone or dual-functioning system is more desirable in any given nuclear plant depends upon the specific monitoring system design under consideration and the particular policies of the user organization, but, in general, dual-functioning systems seem superior.

### Data Acquisition

The intent of any monitoring system is to provide information that will enable an accurate assessment of fuel pin integrity and general core conditions. In core diagnoses, those factors which affect the collection and evaluation of the data must be taken into account. Such factors include: sample representativeness, spectral quality, delay time between data acquisition and analysis, flexibility of analysis, frequency of analysis and computer interfacing. These factors are discussed below.

### Sample Representativeness

One of the NRC requirements for post-accident monitoring systems is the ability to obtain samples representative of the reactor coolant in the core region.<sup>4</sup> Assuring sample representativeness is one of the more difficult problems in the design of post-accident monitoring systems. Although not defined in the NRC Regulatory

Guides, sample representativeness is defined here as the similarity of radionuclide concentrations and chemical and physical properties between the sample at the time of analysis and the reactor coolant in the core region.

Post-accident sampling undertaken during the TMI accident illustrated the problems that may be encountered in obtaining a representative sample under abnormal circumstances. For example, the TMI reactor coolant sample obtained about 2 hours and 45 minutes after the initial reactor scram was not representative. The "Lessons Learned" Task Force concluded that under the existing circumstances it was impossible to obtain a representative sample.<sup>2</sup> Factors contributing to this situation included inherent properties of the partially uncovered core, a change in the sampling protocol because of high reactor coolant activities, and a large loss of gaseous fission products from the reactor coolant liquid phase. Because of partial uncovering, the reactor core functioned like a still, concentrating volatile elements in the steam and collecting nonvolatile elements in the bottom of the reactor vessel. The volatile elements then condensed out or dissolved in the downflow through the steam generator. The let-down system, which was being operated to provide core cooling and which provided the only available sampling point, had in essence "distilled reactor coolant" flowing through the sample lines. However, even if the internal core conditions had been amenable to representative sampling, deviations from the regular sampling protocol would have virtually guaranteed an

invalid sample because the operators reduced the sample recirculation time from 30 to 8 minutes and also failed to purge stagnant water out of the sample leg. Furthermore, as a result of the sampling process, a tremendous volume of fission gases escaped from the sample.

The magnitude of the gaseous activity that was released is demonstrated by the much higher exposure rate near the sampling line (over 1000 R/hr at six feet from the sampling line) than would correspond to the measured reactor coolant sample activity of  $1.6 \times 10^3 \mu\text{Ci/ml}$ . The actual reactor coolant activity in the sample line which created the extreme exposure rate at that time was calculated to be about 10 to 30 Ci/ml. It is theorized that because reactor coolant in the sample line was under high pressure and temperature, fission gases were dissolved in the liquid, and when the operator collected the sample, fission gases flashed out of solution and escaped into the atmosphere.<sup>2</sup>

If a sampling system had been in operation at TMI that had the capability of preventing the escape of dissolved fission gases and a valid sampling protocol had been followed, the collected "distilled" reactor coolant sample could have indicated core uncovering by virtue of the fission product profile present in the sample. However, only an adequate data base would have enabled the operators to make this assessment.

In addition to the several specific problems identified at TMI, one general cause of poor sample representativeness is time-induced changes due to the buildup of daughter, and the decay of parent,

radionuclides. Some fission products, which in themselves may be indicative of specific core conditions, are also daughters of other fission products whose releases may indicate different core conditions. A notable example occurs with the cesium radioisotopes. Several cesium radioisotopes are produced by relatively high direct fission yield. In addition, however, certain cesium radionuclides are also formed by xenon parent decay. The release of cesium radioisotopes into the reactor coolant can be indicative of elevated core temperatures, whereas xenon radioisotope releases do not necessarily indicate the same.<sup>3</sup> As a result, it is conceivable that cesium isotopes in reactor coolant originating from xenon decay could be misinterpreted as resulting from elevated core temperatures. Should this occur, it would possibly lead to a false conclusion regarding the condition of the fuel, and in any case illustrates the potential for significant time-induced sample alterations.

With respect to the problem of time-induced changes in samples, OLS have the capability of analyzing on a real (or near real) time basis. This somewhat minimizes time-induced changes in the reactor coolant sample fission product inventory. However, longer time delays are an inherent feature of GSS, since in such systems samples are transported from the sampling point to a separate laboratory for analysis. GSS therefore normally cause greater time-induced sample alterations. To fully compensate for the effect, use of computer codes are necessary to correct for buildup and decay. Knowledge of the precise time delay between collection and analysis is necessary



for accuracy. Such a procedure is quite time consuming and may not be cost-effective. That is, the benefits gained may not be significant enough to warrant computer reduction.

Because of analysis nearer to real-time, OLS view many shorter-lived radionuclides that would otherwise decay to negligible levels before a grab sample could be analyzed. Therefore, OLS definitely analyze more representative samples in terms of time-induced sample changes. However, it is questionable whether actual interpretation of data is improved by a shorter decay time, since interpretation of data also depends on other factors, such as overall radioactivity levels, which radionuclides are used in diagnosing the core condition and length of reactor operating time. On-line monitoring may preclude use of longer-lived radionuclides in core assessment because of gamma spectral interferences from highly radioactive short-lived components. However, this may only be a consideration for post-accident monitoring, since under normal conditions the reactor coolant activity levels are relatively low. Conversely, a grab sample analysis precludes the use of shorter-lived radionuclides in core diagnosis. This may be either unfavorable because of a loss of information or advantageous because of a reduction in gamma spectral interferences.

Sample representativeness can also be lost by physical alterations induced in the sample as a result of specific monitoring techniques with the major effects being phase separation and plateout. Loss in representativeness due to physical alterations is

probably more significant in terms of influencing data interpretation than time-induced changes. Such physical alterations occur to a greater extent in grab sample analysis than with on-line monitoring.

An illustration of the effect of phase separation was described previously for the post-accident sample taken during the TMI incident in which a large fraction of the fission gases escaped. A current NRC requirement for post-accident monitoring is that capability for the quantification of the total dissolved gaseous component in the reactor coolant be provided if analyzing depressurized samples.<sup>3</sup> Most (if not all) post-accident GSS have the capability for stripping and collecting dissolved gases. In the process of collecting a grab sample, the temperature and pressure is significantly altered from that in the reactor coolant system, although some GSS are able to acquire both pressurized and unpressurized samples.<sup>15</sup> A shift in temperature and pressure may alter the liquid/gaseous composition of the sample and induce volume changes which can complicate the determination of fission product concentrations.<sup>1</sup> Temperature and pressure can also induce precipitation of certain radionuclides and may cause dissolved gases to come out of solution.

Without a dissolved gas collecting unit in GSS, a large fraction of the fission gases may escape. However, the removal of dissolved gases from the liquid phase is not 100% efficient and may vary from sample to sample. Thus, for improved data accuracy, a normalizing factor is required to correct for gases remaining dissolved in solution after application of gas stripping procedures. This normalizing

factor may not be appropriate for all cases, since the temperature and pressure may be altered during an accident.

Perhaps an even greater problem than phase separation is the tendency for certain fission elements to plateout onto sample line walls and other system components. Notable examples are iodine and cesium nuclides. The fraction effectively removed from the reactor coolant in this manner is uncertain. Normalizing factors to compensate for removal by plateout developed for normal operational temperatures and pressures may not be appropriate under accident conditions because of probable alterations in environmental conditions which subsequently alter the degree of plateout.

The NRC requires that techniques be incorporated into post-accident monitoring systems to minimize the degree of plateout in sample lines.<sup>3</sup> Such measures include sample line heat tracing, minimizing line lengths, the incorporation of large radius line bends, and the avoidance of dead legs on flow paths and internal crevices in system components.<sup>11,16,17</sup> Another technique requires operating a system at sufficiently high flow rates such that an equilibrium is established between the amount of activity collecting on sample lines and the amount being physically removed.<sup>1</sup>

The problem of plateout is not unique to either OLS or GSS; both require methods of reducing it. However, the magnitude of the problem is more severe with GSS, since OLS can monitor reactor coolant at or near actual reactor coolant temperatures and pressures by including pressure regulators and orifices, and can also monitor

flowing reactor coolant.<sup>1</sup> Moreover, GSS probably use longer sampling lines, which enhances, the plateout problem.

On the other hand, on-line monitoring systems that view static reactor coolant samples must flush the sample lines between sample acquisitions to remove crud, minimize fission product plateout, and prevent blockage by crud deposits.<sup>3</sup> In these systems plateout is of particular importance because it can contribute to background radioactivity. An on-line monitoring system recently installed at the Arkansas One Plant corrects for a background component in the sample lines. A spectrum is acquired prior to sample introduction and is subtracted from the sample data via computer reduction.<sup>18</sup> Such automated computer-controlled operations improve accuracy by correcting for plateout deposited by previous samples.

Another obstacle to achieving a representative sample under accident conditions is that of selecting a suitable sampling point, and selection of a proper sampling location is a problem common to both GSS and OLS. One complication is that a given sampling point may be suitable for some accident sequences, but not appropriate for others. A long sampling line may be required in order to locate the sampling or measuring point out of a high radiation area.

The requirements for sampling lines do not coincide with standard engineering practices, specifically in the area of sample line restrictions.<sup>19</sup> That is, the NRC regulations mandate that upon indication of an accident, the reactor containment must be isolated, thus requiring the isolation of all let-down lines. The NRC only

allows an exception for the grab sampling lines for the purpose of obtaining samples for analysis. In this case, the reactor coolant in the line is allowed to recirculate for about 20 to 30 minutes prior to sample acquisition to improve representativeness. After the sample has been collected, the line must again be isolated.

Because of this regulation, the capacity of post-accident OLS and may be limited, since they could only acquire data if located on the grab sampling line, and under sample line isolation continuous monitoring would not be possible.

### Spectral Quality

The quality of a gamma spectrum is determined by photopeak clarity (resolution), spectral (photopeak) interferences, magnitude of the incident count rate, and the overall quality of the counting system. While primarily influenced by the type and quality of the detector and associated electronics, the resolution is also heavily influenced by the technique used for reducing high count rates (refer to the later section in this chapter entitled High Count Rate Capability).

Spectral interferences frequently arise from different radionuclides that emit gamma rays at the same (or nearly the same) energy, such that their corresponding photopeaks cannot be resolved (or separated for purposes of identification) without special data reduction techniques. Another type of spectral interference is due to the Compton continuum from higher energy gamma rays that mask the

photopeaks of lower energy gamma rays of interest. The degree to which spectral interference develops and the problems it presents depends upon the concentrations and the number of different radionuclides present in a sample. Because OLS detect shorter-lived radionuclides more readily than GSS, the problem of spectral interference is greater for OLS.

The magnitude of the spectral interference problem is enhanced with large fission product releases as would occur during a core melt accident. Recognizing this potential problem, several techniques have been applied to help minimize the resultant degradation of spectral quality. Among these, the technique of gas stripping is used by most post-accident GSS and reduces spectral interferences by a significant amount. Interfering components can also be removed by other fission product separation techniques. Removal of interferences is advantageous because interfering components are separated out, rather than being corrected by computer reduction or ignored. For example, in the case of  $^{135}\text{Xe}$  and  $^{131}\text{I}$ , an improvement in accuracy by a factor of 50 has been found by analyzing the reactor coolant liquid and gas separately.<sup>20</sup> However, in the time required for degassing of the reactor coolant, there may be a loss of short-lived radionuclides valuable in core diagnosis.<sup>21</sup>

The separation of fission products by their chemical characteristics is not restricted to the isolation of gases from liquids. Another technique, ion exchange chromatography, is currently being developed in a prototype of a fully automated on-

line monitoring system.<sup>22</sup> A unique advantage with ion exchange chromatography is that in routine application it can enhance selective analytical sensitivity.<sup>23</sup> Moreover, both eluate and resin can be counted.

Although techniques of fission product group separation are not limited to either GSS or OLS, they are certainly performed more readily in a laboratory. Group separation capability can be incorporated into OLS; however it increases the sophistication requiring fully automated techniques to direct the separations and an extensive network of sample lines, valves and other components. Furthermore, maintenance requirements are more demanding. However, automation of the technique can reduce personnel exposure and analysis time, in addition to improving accuracy and reproducibility.

#### Real-time Analysis

Since prompt analysis of reactor coolant can provide important information for controlling the course of an accident, monitoring reactor coolant radionuclide concentrations, and subsequently assessing fuel condition, on a real-time (or near real-time) basis is valuable.<sup>9</sup> The need for such capability was evidenced during the TMI incident where the availability of real-time reactor coolant radionuclide information could conceivably have indicated to the reactor operators that the abnormal concentrations of fission products in the reactor coolant had resulted from an elevated core temperature. Proper interpretation of the data could possibly have

changed the course of action undertaken by the operators and mitigated the consequences.<sup>3</sup>

The NRC now requires that both post-accident reactor coolant sample acquisition and analysis be completed within three hours.<sup>4</sup> Because OLS can monitor the reactor coolant on a near real-time basis, they enable quicker analytical results than analysis of grab samples, and provide an earlier warning capability of unusual reactor coolant activities. Power plants use beta-gamma detectors (of the ionization chamber variety) to monitor gross radioactivity levels in the reactor coolant. Most OLS use intrinsic germanium detectors. Such OLS can provide an early warning with immediate isotopic analysis and, with computer reduction, can supply a prompt diagnosis. These types of OLS can therefore distinguish between innocuous crud or iodine spikes and elevated temperature releases. On-line gross beta-gamma detectors of the ionization chamber variety also can provide an early warning of elevated reactor coolant activities. However, to determine the cause of the increased count rate, isotopic analysis is required.

#### Flexibility of Analysis

On-line monitoring systems are inherently restricted to their built-in capabilities in the types of analytical techniques they can conduct. On the other hand, once a grab sample has been collected, much more flexibility exists as to what type of analysis can be performed. For example, a given analysis of a grab sample can be



repeated several times on a particular sample, although an OLS can also repeat an analysis, it must be done on successive reactor coolant samples. Also, a specific grab sample can be saved for later re-analysis, although considerable information may be lost due to radionuclide decay. On the other hand, with most OLS only the collected data can be archived and the sample cannot be saved. This greater flexibility in the analysis of grab samples is an important advantage, since it is uncertain what information will be the most useful under specific emergency conditions. Moreover, utility management, government agencies or national laboratories may request an analysis during emergencies which requires some flexibility in analytical capability.

#### Frequency of Analysis

Nuclear plants typically obtain reactor coolant grab samples once per shift (about three times per day), although a few sample less frequently. A low frequency of data collection may be a source of problems because it can result in missing, or delaying the discovery of, some important events. Also, trends in the reactor coolant activity may take longer to recognize because of short term variations throughout the day. In addition, certain results may initially be attributed to short term statistical deviations or spiking, rather than to some other occurrence, making the operators more prone to discount unusual data as anomalous.

Continuous monitoring enables earlier detection of elevated temperature releases and reduces manual sampling efforts.<sup>24</sup> Also, compliance with plant technical specifications can be continuously monitored and certain parameters, such as the reactor coolant  $^{131}\text{I}$  dose-equivalent rate, can be automatically updated.

A recent study compared the applicability of an on-line monitoring system to a grab sampling system for core diagnosis on the basis of continuous monitoring capability.<sup>25</sup> The on-line monitoring system retrieved seven spectra of the reactor coolant, the total of which presented a "picture" adequately depicting what was occurring in the core. However, because of the dynamic conditions in the reactor coolant, the analysis of the one grab sample taken during this time period did not provide enough information to make an accurate evaluation.

Continuous monitoring capability is a definite advantage that OLS have over GSS. It is impossible to achieve such capability with GSS. Moreover, GSS require manual effort, and therefore only relatively infrequent reactor coolant analysis can be performed.

### High Count Rate Capability

The technique for handling high activities is a major post-accident monitoring system design consideration. The practical options include collimation of the gamma rays, sample dilution, sample holdup for decay, special high count rate electronics, variable sample line geometry, separation of fission product groups

based on chemical characteristics, or a combination of these techniques. The feasibility of using any of these specific options varies with OLS and GSS. Because the technique used affects a system's merits, it should be carefully examined in view of the advantages and disadvantages associated with incorporating it into OLS and GSS designs. The mechanisms used can affect sample representativeness, data quality, ease of maintenance, cost, system complexity, ease of operation, and other factors.

New NRC regulations require that post-accident monitoring systems be capable of analyzing samples within an activity range of 10  $\mu\text{Ci/ml}$  to 10  $\text{Ci/ml}$ , a dynamic range of six orders of magnitude.<sup>5</sup> If the post-accident system also functions in a routine monitoring mode, the lower end of the range must be expanded by up to another three or four decades.

It has been calculated that for an accident where the reactor coolant system line does not break, and where 100% of the noble gases, 50% of the halogens and 1% of the other fission products escape from the core into the reactor coolant, the reactor coolant exposure rates one hour after an accident at 1 cm from a sample per  $\text{cm}^3$  of sample will be as follows (sic):<sup>27</sup>

reactor coolant liquid, not degassed	14,110 R/hr
reactor coolant liquid, degassed	10,620 R/hr
reactor coolant fission gases	5,600 R/hr

The various exposure rates and reactor coolant activity levels encountered during the TMI incident provide a case study of the

reactor coolant radioactivity levels a post-accident monitoring system must be able to analyze. For example, at 8:45 a.m., 4 hours and 45 minutes after the initial reactor scram, the exposure rate 6 feet from the sample line exceeded 1000 R/hr. It has since been calculated that the reactor coolant activity levels required to produce such high exposure rates would have had to have been between 10 and 30 Ci/ml.<sup>2</sup> However, the exposure rate at the detector may not have been entirely from the sample line itself, since it could have also included airborne radioactivity and other background radiation components. Nevertheless, the high exposure rates encountered during the accident clearly demonstrate the need for post-accident monitoring systems to include the capability for analyzing high concentrations of radioactive materials.

GSS need to incorporate mechanisms to mitigate high sample activities for personnel protection purposes in addition to preventing detector saturation. Shielding and/or dilution of post-accident samples are usually required to protect personnel involved in sample acquisition, transport, and analysis. Because a grab sample is transported to a separate laboratory for analysis, the capability to handle high activities can be achieved with relatively little difficulty. The factors to consider when determining the specific mechanism to use in reducing high activities are personnel protection, need for accuracy, the key radionuclides to be specifically analyzed, and the desired analysis time. Because the priority of

these considerations varies with each unique accident scenario, grab sample counting in a remote laboratory provides flexibility.

The techniques for handling high activities with OLS are more limited because of space constraints, and the capability must be built into the systems. Because of remote location and automated operations, personnel exposures in analytical operations are reduced. This is especially advantageous in the event of an accident. However, the overall activity levels encountered by an on-line monitoring system in the event of an accident, and even during routine operations, will be higher. If the techniques designed into the system to handle high reactor coolant count rates and to shield against background radiation are found to be inadequate during an accident, the situation cannot be readily remedied by applying additional techniques to reduce radiation levels as would be possible with a grab sampling system. Therefore, high count rate capability is a major design consideration with OLS.

The various techniques that may be used to expand count rate capability will be considered in view of their applicability to OLS and GSS and in view of their effects on data quality and other factors.

### Collimation

Although the concept of collimation is easy to comprehend, the mechanics of its use in post-accident monitoring systems are not without problems. As used in the current application, collimation is

the technique of reducing the intensity of gamma rays incident on a detector by imposing a shield with a built-in aperture between the detector and the radioactive source, so that to a first approximation only those gamma rays passing through the aperture can impinge on the detector. Variable collimation (that is, incorporating different sized apertures into one collimator) allows the effective detector to source geometry to be changed.<sup>20</sup> With a change in aperture comes an accompanying alteration of overall detector efficiency and peak to Compton ratios as a function of energy. As a result of the changing efficiencies, a special calibration procedure is required for each aperture and for each gamma ray energy. This entire topic is discussed more fully in Chapter III.

For practical application in post-accident reactor coolant analysis, variable collimation is one of the easiest ways to extend the ability to count reactor coolant samples with high concentrations of radioactivity. However, it is doubtful that collimation alone can make it possible to analyze reactor coolant activity at the required maximum concentration of 10 Ci/ml because of severe spectral degradation induced by small aperture sizes. Therefore, in order to obtain high quality spectra at extremely high count rates, collimation plus additional techniques for reducing the count rate will normally be required.

Variable collimators typically require massive lead or tungsten shields and subsequently are physically large and heavy. With an on-line monitoring system the collimator assembly has to be controlled

automatically and must employ a driving force to position the assembly remotely.<sup>20</sup> For maximum accuracy and quick response, the use of collimation in OLS requires computer actuated efficiency calibration and aperture selection. With GSS, the collimator aperture selection, changing and positioning are all performed manually and are therefore more cumbersome and a source of possible errors.

For either GSS or OLS, a mechanism for determining which aperture size to select based on sample activity must be incorporated. For example, a method which has proved quite successful for an on-line monitoring system located at a research reactor at the Idaho National Engineering Laboratory, which operates on a computer generated signal that directs alignment of the collimator based on the percent busy signal.<sup>28</sup> In this system, a computer controls the variable collimator with aperture selection based on a dead time signal. The correct dead time signal is determined in the live-time corrector by summing the shaping amplifier dead time, the analog to digital converter dead time, and the negative pulse gate time, along with a pile-up rejector signal. A similar, but less elaborate, technique has also been incorporated into an on-line monitoring system installed at Arkansas One.<sup>18</sup>

The technique of collimation for reduction of high count rates can be incorporated into both OLS and GSS. However, incorporation into automatic OLS requires greater sophistication for practical use.

### Variable Geometry

Another effective method of reducing high count rates involves altering the geometric relationship between the detector and the source, either by increasing the distance or by changing the source configuration. This technique is often used in conjunction with other mechanisms for reducing high count rates. For example, one vendor markets an on-line monitoring system with a dual sample line in addition to a variable collimator.<sup>25</sup> In this system, the intrinsic germanium detector views a low level sample line through a slit in a dual aperture collimator. A high activity post-accident line is located at an extended distance with another collimator assembly between the two lines. Computer actuated, solenoid operated valves, with manual valve backup, are used for selection of the sample line. Information for deciding when to change the sample line flow or the collimator alignment is provided by a computer which monitors a gross count rate meter to provide the needed data.<sup>18</sup>

Another system using dual sample lines combines dilution and other count rate reduction techniques with a variable line geometry.<sup>22</sup> A low activity (or routine operation) sample line is coiled about the detector. A gross count rate meter is used by a computer to monitor sample activity. If the activity exceeds a given set point, the flow is diverted into a high activity sample line which passes behind a variable collimator. If this technique does not sufficiently reduce the input count rate, then additional sample count rate reduction techniques are actuated.



The preceding examples illustrate how alteration of sample to detector geometry can be used with OLS to reduce count rates without increasing sample to detector distance. An approach by a CANDU reactor on-line monitoring system employs an automated carriage which can increase the distance between the detector and sample holder over a range of 8 to 100 cm.<sup>29</sup> This approach, however, requires ample space and is not feasible with OLS in most light water reactors.

GSS can generally use any kind of alternate geometry configurations used by OLS, in addition to increasing sample to detector distance, with the only constraint to such configurations is concern regarding personnel exposure. Using variable geometry has the definite advantage of not changing sample representativeness, since the reactor coolant itself is unaltered. In the case of OLS, this also implies no change in sample temperature and pressure. The technique also makes calibration easy whether done automatically under computer control or manually in a laboratory. Because it is easier to introduce variations in geometry with GSS, they are superior to OLS in this regard.

### Dilution

A current NRC criterion with respect to post-accident monitoring systems is that, where necessary and practicable, the ability to dilute samples should be provided in order to improve measurement capability and reduce personnel exposure.<sup>3</sup>

Most post-accident GSS incorporate the capability for sample dilution prior to sample collection. In addition, most have the capability of providing both diluted and undiluted reactor coolant samples. In some systems, the dilution capability is automated and the dilution factor necessary to prevent detector saturation is determined by the initial sample count rate.<sup>11</sup>

Dilution prior to sample acquisition has the advantage of protecting personnel involved in the procedure. Moreover, dilution requires withdrawal of only a small reactor coolant volume from the sample line which also minimizes exposures, but at the same time possibly reduces the sample representativeness.

A definite advantage of dilution is that the count rate reduction factor is unlimited, except by inherent capabilities of a given system and possibly the need to perform certain chemical measurements. With GSS, if the initial dilution factor is inadequate, further dilutions can be made manually in a laboratory. With OLS, the capability to dilute samples until the desired count rate is achieved can be designed into the systems by using computer controlled feedback mechanisms.<sup>22</sup>

A high degree of accuracy in the dilution process for either GSS or OLS must be ensured to minimize error in the results. Automatic dilutions in both types of systems reduce the human error factor and are normally more precise and reproducible.

Dilution can potentially alter the sample pressure, temperature and pH. Such alterations can change the chemical form and

consequently the behavior of the fission products. In the case of OLS, addition of diluent can induce phase separation of the gases from the reactor coolant solution due to physical agitation. Counting samples with separated phases can reduce the accuracy of an analysis. This problem may not occur with post-accident GSS because most GSS collect gases, separately.

#### Sample Diversion and Holdup

Sample holdup reduces count rates by allowing radioactive decay to occur before counting. Because short-lived radionuclides may decay to undetectable limits, this technique also reduces spectral interferences. With both OLS and GSS, the sample is merely set aside for a time period prior to counting. Depending on the initial activity, the sample may be counted prior to holdup. Multiple sample counting can supply additional useful information about the reactor coolant, since both shorter- and longer-lived radionuclides (the latter after initially being hidden by short-lived radionuclides) can be used to assess the core condition.

Although the principle of sample holdup is identical for both OLS and GSS, their methods differ. OLS must have the capability built into the system using additional valves, sample lines, and other components. A mechanism is also required for deciding when to hold up the sample and for what length of time. This feature is not found in presently available commercial OLS, but has been demonstrated in a prototype system.<sup>22</sup> Furthermore, with an on-line

monitoring system the sample cannot usually be saved over an extended time period for re-analysis; only the data can be archived. On the other hand, a grab sample can easily be saved indefinitely, the only requirement being a shielded location in which to store the sample.

Because of the relative ease with which grab samples can be stored, GSS have an advantage over OLS in this regard.

### Matrix Separation

Matrix separation involves the isolation of groups of fission products by their chemical or physical characteristics. The separations can be accomplished in several ways, such as by gas-stripping, ion exchange chromatography, or filtration. Matrix separation has the advantage of reducing both count rates and especially spectral interferences.

Gas-stripping, because of the removal of spectral interferences from noble gases, has been found to improve accuracy in the analysis of certain radionuclides. For example, the accuracy of  $^{135}\text{Xe}$  and  $^{131}\text{I}$  analysis has been improved by a factor of up to 50.<sup>20</sup> Gas-stripping also can eliminate interferences caused by escape peaks of high energy  $^{88}\text{Kr}$  gamma rays. For instance, at Arkansas One it was found that the 1174 keV double escape peak of  $^{88}\text{Kr}$  interfered with the analysis of  $^{60}\text{Co}$ ; likewise the 1370 keV double escape peak of  $^{88}\text{Kr}$  interfered with  $^{24}\text{Na}$  analysis.<sup>13</sup>

Matrix separation by ion exchange chromatography can increase the sensitivity in the analysis of certain radionuclides typically

found in low concentrations in the reactor coolant. In one performance test of an automated ion exchange chromatography system analyzing reactor coolant, an improvement by a factor of ten was demonstrated over other techniques.<sup>23</sup> Therefore, for routine analysis, ion exchange chromatography can prove to be a definite advantage. In addition both the eluate and resin can be counted separately.

Filtration can separate iodine isotopes and/or particulates from the reactor coolant sample.<sup>30</sup> The filter can be counted, thereby increasing sensitivity in routine analyses.

Drawbacks of matrix separation when conducted manually in a laboratory include time consumption, personnel exposure, increased work load, and the degradation of information associated with short-lived radionuclides due to the time required to separate the fission products.<sup>21</sup> Moreover, for accuracy in assessing concentrations, separation efficiency factors must be established and, these factors may be altered under abnormal circumstances. For example, a change in temperature, pH or pressure can alter ion exchange properties.

Matrix separation can be accomplished with both GSS and OLS. However, such separations require more elaborate system designs in OLS.

#### Conclusions for Count Rate Reduction Techniques

Grab sampling systems allow flexibility with respect to the method of handling high count rates because the analysis is conducted

in a separate laboratory. The analytical limits are those imposed by the techniques available in the laboratory and the capability of the personnel performing the analyses. However, personnel exposure considerations may make certain time consuming techniques unfeasible for analyzing highly radioactive post-accident samples.

The techniques incorporated in the specific on-line monitoring system restrict the techniques that can be used to handle high count rates. If the design proves inadequate in reducing count rates in an emergency, it can not be remedied, whereas with a grab sample, other count rate reduction techniques can be used. However, with OLS personnel exposure during analysis of highly radioactive post-accident samples will not be as great a problem.

The advantages and disadvantages of any particular technique for reducing high count rates must be assessed relative to alternative techniques available, the design of the system, and the overall effects on data quality.

#### Ambient Environmental Effects

Post-accident monitoring systems must be designed to operate in conditions much more severe than the normal environment.<sup>1</sup> Environmental effects listed by the American National Standards Institute that can adversely effect the performance of a counting system include extreme temperature and pressure, shock, vibration, harsh chemicals, ambient ionizing radiation, humidity, high current contact closures, electrical power transients, magnetic and radio frequency

fields, and atmospheric dust.<sup>31</sup> However, with respect to post-accident and routine monitoring systems, only temperature, pressure, humidity, and ambient ionizing radiation fields are of any practical concern. The severity of any of these environmental factors will be greater on an on-line monitoring system because of its location. Furthermore, the insult will be even greater during an accident and the magnitude of the effect will vary depending on system proximity to the core region. A harsh environment affects all system components; however, certain components are more sensitive and more important than others. With both OLS and GSS high performance gamma ray detectors are critical for optimal data acquisition. Germanium detectors have typically not been optimized for nuclear power plant application. They are usually purchased on the basis of relative efficiency, energy resolution, and peak to Compton ratio. Consequently, these are the parameters that germanium detector vendors have tended to optimize, ignoring other factors such as stability against changes in ambient temperature, effect of high count rates on detector performance (such as resolution and gain shift), microphony effects, and effects of other environmental conditions (such as relative humidity, dust, chemical fumes, etc.).<sup>32</sup>

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The greatest detrimental effects on system components during normal conditions occur from high temperatures and ionizing radiation fields. However, harsh chemicals may also be a problem under accident conditions, since a large concentration of corrosive chemicals in addition to fission products may be present in the reactor coolant.

Elevated temperatures affect both the detector and electronics. The influence ranges from minor calibration shifts to severe degradation of performance and possibly permanent detector damage. In designing post-accident monitoring systems, anticipated ambient thermal operating extremes and other possible heat loads must be considered, especially with OLS.<sup>31</sup>

An exposure to extreme elevated temperatures can significantly damage germanium detectors, and in order to function they must be cooled by liquid nitrogen to prevent thermally induced signals from being generated. However, the relatively new intrinsic germanium



detectors have the advantage of not being damaged by loss of cryogenic conditions, although they will become nonfunctional.

The germanium detectors must be recharged with liquid nitrogen at least every month. This is relatively easy to do under laboratory conditions with GSS but much more difficult with OLS although remote filling capability can be built into OLS. If in either case high radiation levels should preclude the recharging of the detector, it may be rendered inoperable when needed.

Ambient temperature can also significantly affect the amplifier gain.<sup>32</sup> One group at the Idaho National Engineering Laboratory solved this problem by thermally insulating their amplifier.<sup>27</sup> In their situation it was especially important, since the amplifier is part of an on-line monitoring system which monitors the reactor coolant of an experimental reactor where elevated temperatures and high radiation fields are encountered. This measure is not as important with an amplifier located in the relatively mild temperatures of a laboratory but may be an absolute requirement with any on-line monitoring system.

The effects of elevated ambient radiation fields include radiation damage to system components in addition to possibly degraded spectral information or even total malfunctioning of a system due to detector count rate saturation.

As mentioned previously, the background radiation field can become extreme during an accident scenario. The background radiation field components include direct shine from the core and sample lines

and from airborne radioactivity. An NRC requirement with respect to post-accident monitoring is that background levels of radiation in the analytical facility be restricted such that sample analysis will provide results with an acceptably small error (approximately a factor of 2).<sup>3</sup> It is further suggested that shielding around the sample be used in conjunction with a separate ventilation system to sufficiently reduce the airborne radiation component.

The ambient radiation field will be significantly lower in a laboratory where a grab sample will be analyzed, than at the location of an on-line monitoring system. However, during the TMI incident, even the elevated background activities in the laboratory where post-accident samples were to be analyzed prevented the counting system from being operational.<sup>2</sup> The laboratory experienced an exposure rate of 25 R/hr within five hours after the initial reactor scram.<sup>2</sup> Because of the high exposure rates personnel were not allowed to enter the counting area. Such a potential problem with GSS can be remedied by locating and equipping an emergency analysis station separately from the routine analytical laboratory. At the emergency analytical laboratory, special measures can and must be taken to reduce the ambient background radiation levels. Such measures include incorporation of ventilation systems, massive shielding and remote location from the containment building. However, such measures are costly, take up extra space, and require that extra instrumentation be committed to the emergency laboratory.

Portable intrinsic germanium detectors and battery-powered

portable multichannel analyzers are available which can remedy this problem to some extent by allowing sample counting to be done elsewhere if needed, assuming a grab sample is obtainable.

OLS must incorporate shielding which will enable them to operate in extreme background levels. The amount of shielding necessary under accident conditions may be considerable, since a detector should not experience greater than  $\mu\text{R/hr}$  background levels if it is to avoid having an excessive interfering count rate.<sup>33</sup>

The on-line monitoring system recently installed at Arkansas One is claimed to have sufficient shielding (calculated by extrapolation from lower ambient levels) to adequately ensure capability for reactor coolant analysis in an ambient background exposure rate exceeding 100 rad/hr (sic).<sup>18</sup> That amount of shielding, however, would be totally inadequate against the exposure rates encountered in a core-melt accident. For example, in such a situation, the resulting contact dose rate from a one gram ( $\sim 1 \text{ cm}^3$ ) post-accident reactor coolant sample (assuming a non-line break design basis accident) was calculated to be  $9.7 \times 10^5 \text{ rads/hr}$ .<sup>1</sup> Thus, installation of the amount of shielding an on-line monitoring system would require to ensure proper detector performance in the presence of a core-melt accident with an on-line monitoring system may be impossible because of space constraints.

In addition to the gamma radiation component, the background neutron field should not be overlooked. While a detector located in a laboratory will not likely experience a neutron field of sufficient

intensity to be of concern, a detector in an on-line monitoring system will be exposed to a higher neutron field. The magnitude of the neutron field will be a function of the detector distance from the core, since greater distances allow for quicker decay of short-lived neutron emitting fission and activation products and a reduced direct fission neutron component.

Concerns about radiation damage to equipment and the problem of ambient background radiation is greatly reduced in the grab sample approach compared with on-line monitoring. The main reasons for this evolve around the fact that because the analytical equipment is only exposed to radioactive reactor coolant occasionally, the sample is likely to be diluted immediately if gross activity exceeds a certain value, the sample volumes are significantly less, and the background radiation field is lower.<sup>1</sup> The reduction in damage to system components will result in reduced frequency of repair and possibly greater system reliability.

### Health Physics Considerations

The current design basis for post-accident reactor coolant monitoring systems from a health physics standpoint requires a capability to obtain and analyze a sample without having radiation exposures to any individual exceed the criteria in 10 CFR Part 50, Appendix A.<sup>34,35</sup> Measures must be taken to protect personnel from exposures originating from the reactor core, sampling lines, the sample itself, and airborne radioactivity. Although the hazards are magnified under

severe accident conditions, protective efforts are required for routine monitoring purposes as well. The primary techniques for minimizing personnel doses include reducing sample activity, incorporating shielding, reducing exposure time, and increasing distance between samples and personnel.

The high exposure rates experienced during the TMI accident illustrate how difficult it is to limit personnel doses to NRC requirements under conditions of significant core degradation. As a consequence of dissolved gases coming out of solution during the sampling process at TMI, high airborne radioactivity resulted which forced the evacuation of site personnel.<sup>2</sup> Several of the personnel at TMI incurred relatively high radiation exposures while obtaining reactor coolant samples.<sup>10</sup>

Measures taken to either prevent separation of or collect dissolved gases would minimize personnel doses from airborne radioactivity. Indeed, post-accident GSS incorporate the capability to strip and collect dissolved gases. Since OLS monitor flowing reactor coolant, the gases remain contained and the problem of requiring a gas collecting unit is circumvented.

Since a post-accident monitoring system may have to analyze many sources of significant activity, the radiation levels alone indicate a need for substantial shielding to keep personnel doses below the regulatory limit over the course of an accident.

Because OLS can be operated automatically at remote locations, they are definitely superior from a radiological health standpoint.

OLS eliminate the need for personnel to spend time in the vicinity of the reactor coolant and keep personnel distances at a maximum. On the other hand, the acquisition and analysis of grab samples results in greater personnel exposures, even though several features can be incorporated which somewhat minimize doses, such as reducing sample activity by dilution and obtaining small sample volumes. Post-accident GSS are extensively shielded, and shielded containers are provided for transporting samples to the laboratory. Distance can be increased by the use of reach rods with which to operate system valves and handle the sample. However, because GSS are manually operated, human error factors must be considered both in sample acquisition and analytical operations, especially in the event of an accident. The extreme stress that may be induced from an accident can increase the possibility of mistakes. Also, new procedures may be required for accident sampling which could further enhance a tendency for errors.

Reduction of personnel dose is a very significant advantage of on-line monitoring systems and should be assigned high priority when selecting post-accident reactor coolant monitoring systems.

#### System Maintenance, Repair and Calibration

Because OLS are located in a harsher chemical, physical and radiological environment than GSS, and because of their greater complexity and sophistication it can be expected that their components will fail at a greater frequency than similar parts on GSS.

Therefore, repair of OLS will probably be more frequent and, because of their location, more difficult to accomplish.

The sample acquisition portion of GSS consist basically of durable stainless steel mechanical parts. Less radiation resistant parts (such as some electronic components) can readily be shielded and, if necessary, replaced without much difficulty. Most importantly, the more sensitive counting equipment is sheltered in the relatively milder environment of a counting laboratory. Thus, grab sampling systems have the advantage of avoiding the extreme environment in the first place, rather than increasing system complexity to protect components. Similarly GSS are easier to calibrate because they are not as complex and are not remotely located.

The NRC requires that all equipment and procedures which are used for post-accident sampling and analysis be calibrated or tested at a frequency which will ensure to a high degree of reliability that the system will be available if required.<sup>3</sup> Two types of calibration must be performed, energy resolution and counting efficiency. The energy calibration is the determination of the relationship between gamma ray energy and output data channel number. This calibration is typically performed at setup and also frequently during periods of system use. It provides a simple and regular way of checking for system malfunctions. The efficiency calibration is the determination of the relationship between detection efficiency and gamma ray energy. This calibration is typically performed at setup but not

repeated frequently. It is affected by source to detector geometry, so that an alteration, such as occurs by altering the collimator aperture, requires performance of a new efficiency calibration.

Equipment in a laboratory can readily be calibrated by conventional methods for efficiency and energy prior to counting a grab sample. However, calibration of remote OLS requires the incorporation of more elaborate methods. For example, automatic calibration schemes are included in the on-line monitoring system in use at the Power Burst Facility.<sup>27</sup> This system provides automatic energy calibration information for each collected spectrum by means of a precision pulser. By alternately gating two stable current sources through fixed resistances, two square wave pulses are produced with amplitudes corresponding to gamma energies of 260 and 2600 keV. The square wave pulses are fed into the preamplifier through a charge injection capacitor and processed along with gamma ray pulses. This enables an accurate and uniform energy calibration to be performed for every spectrum that is collected in the presence of temperature and electronic drifts and it also provides dead time information. The analog to digital circuitry can identify and sort out the pulser events so that they will not appear in the spectrum. This is clearly a superior calibration method, enabling great accuracy in results and can be conducted prior to each data acquisition.

A remotely located on-line monitoring system installed at the Arkansas One power plant uses a different automated calibration scheme.<sup>13</sup> A multiple nuclide gamma ray source is fixed on a



collimator assembly. Prior to data collection, the source is remotely positioned in front of the detector and the system is automatically calibrated for efficiency and energy under computer control.

An on-line monitoring system requires remote calibration, making the incorporation of automatic calibration procedures more likely. On the other hand, it is improbable that such sophisticated measures would be implemented in a laboratory system.

However accomplished, accurate calibration is essential for accurate data collection, particularly during an accident, and automatic calibration procedures will provide an advantage here in return for increased cost and complexity. Automatic calibration schemes are more consistent, prompt, have less opportunity for human error and can recalibrate immediately prior to data collection. Besides providing better data in all situations, the last consideration may also compensate for possible peak shifting and other electronic changes that may plague OLS during an accident due to the higher radiation fields.<sup>36</sup> On the other hand, automatic calibration is inherently more complex requiring more maintenance and greater cost, and a failure at an inopportune time may not be easily correctable.

Grab sampling systems have the advantage of requiring less sophisticated measures for calibration and maintenance, in addition to being located in an environment less harsh on system components. GSS will therefore not require as much system repair as an OLS and will probably last considerably longer.

### Cost

OLS are more expensive than GSS for several reasons. The cost of OLS still includes considerable research and development expenses, because isotopic on-line monitoring is a relatively new technique since the technology making it feasible has only recently been developed.<sup>9</sup> Conversely, the collection of grab samples has been a widespread practice in routine monitoring for several years in nuclear power plants. OLS are inherently more complex and for that reason also more expensive. The NRC requires that backup post-accident grab sampling capability be supplied if a post-accident on-line monitoring system is purchased.<sup>3</sup> Thus, included in the cost of an on-line monitoring system is the additional backup grab sampling system.

Overall cost should be taken into account (such as operational costs for maintenance, repair, etc.), not just the initial expenditure. For instance, although OLS are initially more expensive, full automation may effectively reduce man-hours previously committed in sample acquisition and analysis.

The initial capital expense should also be viewed in terms of which type of system will provide data of greater value to the utility. A primary consideration is which system is superior in its ability to avert possibly deleterious consequences of a severe accident, that is, which type of system is better able to alert operators at the onset of an accident to plant conditions so that measures can

be taken to avert a possible core melt.

### General Conclusions

The determination of which type of post-accident monitoring system is superior is not clear-cut. There is no proven track record for any type of post-accident monitoring system in nuclear power plants, and hopefully never will be. Furthermore, because of the extreme radiation levels that will be encountered under conditions of extreme core degradation, it is difficult to adequately test any specific system under simulated accident conditions, although one highly sophisticated on-line monitoring system in a special test facility has been proven successful and practical under extreme radiation levels in a high temperature and high humidity environment.<sup>27</sup>

Because on-line monitoring is a fairly new technology and GSS have a history of use in routine monitoring applications at power plants, utilities have tended to choose the latter for post-accident monitoring.

Table II-1 gives a brief summary of the relative merits of each system. The relative advantages of various features are not of equal weight, making the comparison of OLS and GSS even less straight-forward. Also, superiority must be considered in view of the specific design features.

There is no universal answer as to which type of post-accident monitoring system is more desirable for power plant application.

Each utility must evaluate the merits of a given analytical system in light of the organization's operating conditions and physical design features, considering the value of the data obtainable from the system under both post-accident and routine conditions.

Table II-1

SUMMARY COMPARISON OF RELATIVE ADVANTAGES OF GRAB  
SAMPLE ANALYSIS AND ON-LINE MONITORING SYSTEMS

Feature	Grab Sample	On-line Monitoring
Sample representativeness	-	Superior, since analyzes reactor coolant close to actual conditions
Spectral Quality	Superior, because of a reduced possibility of spectral interferences since greater decay of short-lived elements and easier performance of matrix separations	-
Real-time data	-	Superior, since analytical results are obtained quicker
Flexibility in types and techniques of analysis	Superior, since capability is not limited to that built into system	-
High count rate capability	Superior, because of greater flexibility in count rate reduction techniques and reduced background levels	-
Ambient environmental effects	Superior, because of location in a less harsh environment	-
Health physics considerations	-	Superior, because of remote, automatic operation
Maintenance, calibration, repair	Superior, because more accessible location and less frequent malfunctions	-
Cost	Superior, because of reduced cost	

## CHAPTER III

### THE TECHNIQUE OF COLLIMATION IN REDUCING HIGH COUNT RATES

#### Introduction

Collimation is a procedure commonly used in many applications for reducing radiation dose rates or count rates to acceptable levels. Collimation has been incorporated into some post-accident monitoring system designs used in nuclear power plants. Collimation involves interjecting a shield which has a built-in aperture between a radioactive source and a detector so that only those gamma rays which traverse the aperture can impinge on the detector. The collimator thickness and material must be sufficient to reduce the uncollided gamma ray flux incident upon the detector after traversing the collimator thickness to a negligible value. The degree of count rate reduction depends on the size of the aperture, the source geometry, the gamma ray energy and the collimator thickness.

In addition to reducing the count rate, the use of a collimator will degrade the gamma spectral quality due to a proportionately increased background in the resultant gamma ray spectrum. The increased background consists of annihilation photons, Compton scattered gamma rays, and x-rays produced in the collimator, which manage to escape and impinge on the detector. A certain amount of useful information in the photopeak is obscured by the elevated background, and the degree of information lost is greater at lower energies.

Previous collimation studies have used a variety of collimator geometries such as cylindrical orifices (of varying diameter), wedge shaped collimators, and collimators lined with tungsten and/or cadmium.<sup>37,38</sup> In the experiments reported here, collimation using cylindrical apertures of various diameters was analyzed. The intent was to document the effect of collimation on spectral quality and count rate reduction, both as a function of aperture size and gamma ray energy.

#### Materials and Instrumentation

A radioisotope of europium ( $^{152}\text{Eu}$ ), a broad range multi-energy gamma ray source, was chosen so that the energy dependence of the reduction factor and spectral degradation could be investigated.  $^{152}\text{Eu}$  is easily produced, has a sufficiently long half life (13.4 years), and stock solutions of  $^{151}\text{Eu}$  were readily available along with  $^{152}\text{Eu}$  standards traceable to the National Bureau of Standards.

A small volume of  $^{152}\text{Eu}$  was produced by irradiating three polyvials, each containing one ml of a 1 mg/ml  $^{151}\text{Eu}$  stock solution, in the rotating rack facility of the Oregon State University Triga Mark II research reactor. The irradiation was for six hours at a power level of one megawatt, which corresponds to a thermal neutron flux density of  $3.0 \times 10^{12}$  neutrons  $\text{cm}^{-2} \text{ s}^{-1}$  at the sample location. The activity from  $^{152}\text{Eu}$  resulting from the irradiation was calculated to be 104  $\mu\text{Ci}$  in each polyvial (refer to Table III-1). Counts were not collected until after the  $^{152\text{m}}\text{Eu}$  component (9.3 hour half life) had

Table III-1

CALCULATION OF EXPECTED  $^{152}\text{Eu}$  ACTIVITY

$$A = \phi \times N \times S \times (1 - \exp [-\lambda t])$$

$A$  = activity, disintegrations per second  
 $\phi$  = thermal neutron flux, neutrons per  $\text{cm}^2$  per second  
 $N$  = total number of atoms of Eu-151  
 $S$  = thermal neutron capture cross section,  $\text{cm}^2$   
 $\lambda$  = decay constant, per hour  
 $t$  = irradiation time, hours

$$\phi = 3 \times 10^{12} \text{ n/cm}^2/\text{s}, \text{ corresponding to a power of 1 Megawatt}$$

$$N = [(6.02 \times 10^{23} \text{ atoms/mole}) \times (1 \times 10^{-3} \text{ g}/150.91 \text{ g/mole})] \\ = 3.99 \times 10^{18} \text{ atoms}$$

$$S = \pi/2 \times 5800 \times 10^{-24} \text{ cm}^2 = 9111 \times 10^{-24} \text{ cm}^2$$

$$\lambda = (\ln 2/13.4 \text{ years}) \times (\text{year}/365.25 \text{ days}) \times (\text{day}/24 \text{ hours}) \\ = 5.9 \times 10^{-6}/\text{hour}$$

$$t = 6 \text{ hours}$$

$$A = 3 \times 10^{12} \text{ n/cm}^2/\text{s} \times 3.99 \times 10^{18} \text{ atoms} \times 9111 \times 10^{-24} \text{ cm}^2 \\ \times (1 - \exp[-5.9 \times 10^{-6}/\text{hr} \times 6 \text{ hrs}]) \\ = 3.86 \times 10^6 \text{ disintegrations/second} = 104 \text{ } \mu\text{Ci}$$

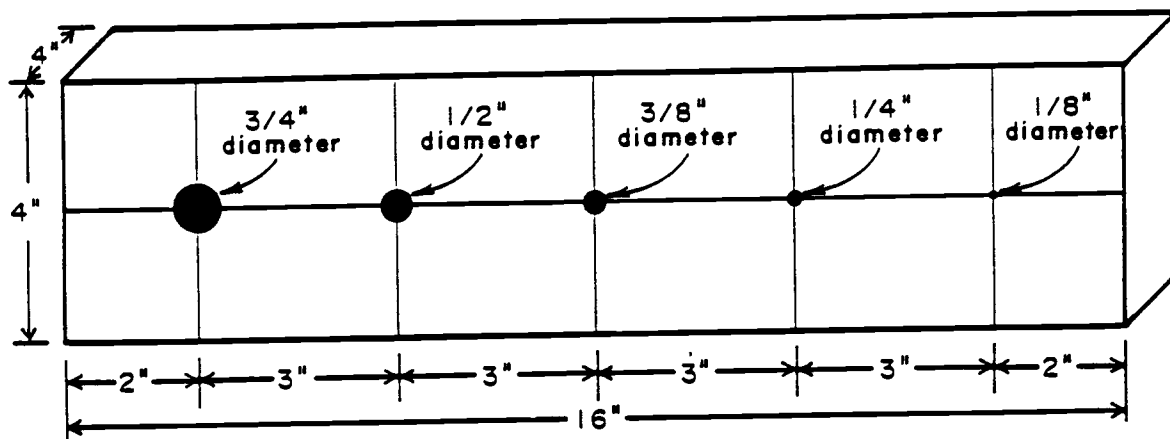


decayed to negligible levels, i.e., more than 90 hours after the end of the irradiation. The actual activity involved at the time the counts were collected was determined by comparison with a National Bureau of Standards traceable  $^{152}\text{Eu}$  standard.

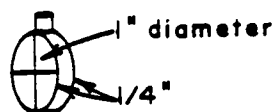
A disk source container was constructed by soldering together two steel planchets, sized such that it adequately covered the largest aperture diameter. The dimensions were 1" diameter and 1/4" depth with an inner volume of  $2.04\text{ cm}^3$  (refer to Figure III-1). A steel tube was built into the top of the disk through which solution could be added by hyperdermic needle injection. To prevent possible leakage of the contents, the projecting steel tube was plugged with "super-glue" after filling the source container and was subsequently covered again with a rubber stopper for added protection. The container was prepared for insertion of the  $^{152}\text{Eu}$  source by several washings with warm water, which were accomplished by repeated hyperdermic needle injections and aspirations.

Health physics protocol was followed throughout the transferring procedure. The transfer was conducted in a hood lined with absorbent paper, protective clothing was worn, and the hyperdermic needle, the unused source solution and the associated materials were discarded into specially marked low-level steel radioactive waste containers.

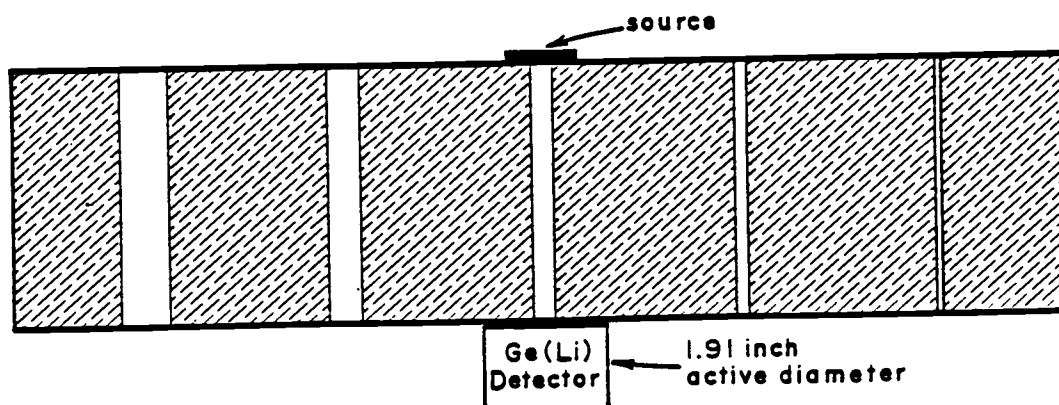
A right circular cylinder (4.85 cm active diameter), lithium-drifted germanium ( $\text{Ge(Li)}$ ) detector, drifted coaxially with one open end, was used in this experiment. The detector had a 14.5% relative efficiency at 1332 keV.



Variable Aperture Lead Collimator



Source Holder



Collimator Cross Sectional Top View

Figure III-1. Collimator set-up

A block-shaped, 4" by 4" by 18" lead collimator, with five cylindrical orifices of varying diameter (1/8", 1/4", 3/8", 1/2", 3/4") was constructed at Oregon State University (refer to Figure III-1). The centers of the apertures were three inches apart so that the detector could only "view" one aperture at a time. Four inches of lead was calculated to sufficiently attenuate an uncollided gamma ray flux traversing the entire collimator thickness:

$$\begin{aligned}\mu/\rho \text{ Pb (2 MeV)} &= 0.0455 \text{ cm}^2/\text{g} \\ \text{Density of Pb} &= 11.34 \text{ g/cm}^3 \\ \text{Initial Flux} &= I_i \\ \text{Final Flux} &= I_f\end{aligned}$$

$$I_f = I_i \times e^{-(0.0455 \text{ cm}^2/\text{g} \times 11.34 \text{ g/cm}^3 \times 4 \text{ in} \times 2.54 \text{ cm/in})}$$

$$I_f/I_i = 0.00517$$

Therefore, only about 0.5% of the uncollided gamma rays at 2 MeV could possibly impinge on a detector after traversing four inches of lead. This calculation is conservative for this experiment since the maximum gamma ray energy analyzed was 1.4 MeV. Thus the error incurred from uncollided gamma rays passing through the lead is insignificant.

Measures were taken to ensure that the source and detector were centered on each aperture. Vertical and horizontal lines were inscribed into the lead, centered on each orifice. Similarly, perpendicular lines dissecting the disk midpoint were inscribed onto the source container. Prior to the collection of each count, the source container was secured by tape over the orifice with the perpendicular lines aligned with those inscribed on the lead surrounding each opening.

A steel constraint was fastened to the counter top so that it rested against the lead shielding the detector. The side of the collimator adjacent to the constraint was positioned so that its entire length rested against the steel constraint, ensuring proper alignment along the collimator axis normal to the detector axis.

Since both the source and detector diameters were significantly larger than the largest aperture diameter and because of the measures taken, precise alignment was not a major source of error. The alignment error was calculated to be negligible for source disk misalignment up to  $5/64$ " and detector misalignment up to  $9/16$ ".

#### Experimental Method

The effect of aperture size and gamma ray energy on the percent transmission of uncollided gamma rays and degradation of spectral quality was investigated. Eight prominent gamma rays of  $^{152}\text{Eu}$ , covering a broad energy range, were selected for use in the analysis of the observed effects (refer to Table III-2). Spectral quality was determined by analyzing collimator-induced alterations in the photo-peak-to-total count rate ratios. The percent transmission resulting from each aperture was also presented in terms of the reduction factor to aid in clarifying the observed effects. Because the effects were normalized to the "no collimator" case, other energy dependent factors, such as photofraction and detector efficiency, cancelled out.

Table III-2

 $^{152}\text{Eu}$  DECAY CHARACTERISTICS

Half life = 13.4 years

ENERGY (keV)	PHOTOFRACTION (%)	$A_0$	$A_1$
		GAMMA/SEC/G*	GAMMA/SEC/G**
121.8	28.4	$3.13 \times 10^3$	$2.25 \times 10^3$
244.7	7.5	$8.54 \times 10^2$	$6.14 \times 10^2$
344.2	26.5	$2.94 \times 10^3$	$2.12 \times 10^3$
443.8	3.1	$3.04 \times 10^2$	$2.19 \times 10^2$
779.1	12.7	$1.33 \times 10^3$	$9.58 \times 10^2$
964.4	14.5	$1.42 \times 10^3$	$1.02 \times 10^3$
1112.2	13.3	$1.35 \times 10^3$	$9.72 \times 10^2$
1408.1	20.7	$2.10 \times 10^3$	$1.51 \times 10^3$

\* $A_0$  = gamma per second per gram europium at 0900 Pacific Standard Time January 1, 1977, Eu-1 standard, 0.3397 grams of 99.2 ppm europium solution

\*\* $A_1$  = gamma per second per gram europium solution (99.2 ppm Eu) at 0900 Pacific Standard Time March 1, 1983; stock solution decay corrected by the relationship

$$A_1 = A_0 e^{-\lambda t}$$

$$t = 6.25 \text{ yrs.}$$

$$\lambda = \frac{\ln 2}{13.4 \text{ yr}} = 5.17 \times 10^{-2} / \text{yr}$$

$$A_1 = A_0 e^{-(5.17 \times 10^{-2} / \text{yr} \times 6.25 \text{ yrs})}$$

$$A_1 = A_0 \times 0.72$$

Source: reference 39

The individual spectral acquisition times were selected so that the standard deviation of the count rate for any given photopeak analyzed did not exceed 1% (i.e., total counts exceeded 10,000, since background was relatively negligible). The count time varied with aperture size and virtually no dead time losses occurred, except in the no collimator case. Because of the low count rates incident on the detector when using small aperture openings, long count times (10 to 14 hours) were required. For greater accuracy, the ambient background component was minimized. This was accomplished by counting only while the reactor was not operating, by extensive shielding of the detector, and removal of all other sources located in the laboratory.

The following counts were obtained:

- 1) 15 hour background count
- 2) 15 hour count, 1/8" aperture diameter, source 4" from detector
- 3) 15 hour count, 1/4" aperture diameter, source 4" from detector
- 4) 15 hour count, 3/8" aperture diameter, source 4" from detector
- 5) 10 hour count, 1/2" aperture diameter, source 4" from detector
- 6) 1 hour count, 3/4" aperture diameter, source 4" from detector
- 7) 0.28 hour (1000s) count, with no collimation, source 4" from detector
- 8) 10 hour background count
- 9)  $^{152}\text{Eu}$  standard count, standard 12" from detector

- 10)  $^{152}\text{Eu}$  source count 12 inches from the detector

Since the background count rates, obtained separately towards the beginning and end of the experiment (31.3 cps and 30.6 cps) varied by less than 3%, it was assumed that the background count rate remained consistent throughout the experiment. The  $^{152}\text{Eu}$  source and standard were counted at the same distance to calibrate the source activity. Although the geometrical configuration of the two differed somewhat (the source was in a disk while the standard was in a cylindrically-shaped polyvial), both approximated point sources because of the distance and hence the calibration procedure could be accurately applied.

The output was coupled through standard modular nuclear counting instrumentation to a Nuclear Data ND660 2048 multichannel analyzer. The data was stored on floppy disks and analyzed using peak search algorithms (program PEAK) generated by Nuclear Data, Inc.

#### Percent Transmission

Percent transmission is defined as the ratio of uncollided gamma rays counted at the detector with collimation to those counted in the no collimator case, expressed as a percentage:

$$\frac{\text{Photopeak count rate with collimation}}{\text{Photopeak count rate without collimation}} \times 100 \%$$

Reduction factor is the inverse of percent transmission times 100.

Reduction in count rate is quite energy dependent and relatively greater at smaller apertures. (refer to Table III-3 and Figures III-2 and III-3). The reduction factors observed also illustrate this effect. With a 1/8" orifice diameter, 1408 keV and 122 keV photopeak count rates were reduced by two to nearly four orders of magnitude, respectively. Conversely, for the 3/4" diameter, reduction factors ranged from about two to five (refer to Table III-4 and Figure III-4). These specific gamma ray energies lie at the extremes of the energies examined. With one exception, the other gamma ray reduction factors lie intermediate between these two. The percent transmission for the 1/8" aperture diameter did not exceed 0.6% for any gamma ray energy. Conversely, for the 3/4" opening no values were lower than 20%. Thus, the 1/8" aperture was highly effective in reducing the count rates. Indeed, in this experiment with 1/8" aperture collimation the count rate was only at a fraction of a second, so that an extreme count time was required for an accurate analysis.

For the 1/8" aperture only, the percent transmission tended to fluctuate somewhat as a function of energy; however, it did show a general trend of increasing percent transmission with increasing energy which was consistent with the other apertures.

#### Percent Peak-to-Total Ratio

The normalized percent peak-to-total ratio (%P/T) is defined as the net photopeak count rate divided by total count rate in the



Table III- 3

## PERCENT TRANSMISSION

$E_{\gamma}$ (keV)	APERTURE DIAMETER (inches)				
	3/4	1/2	3/8	1/4	1/8
121.8	21.3	2.13	1.55	0.318	0.021
244.7	23.6	5.03	1.53	0.321	0.011
344.2	25.9	5.57	1.80	0.354	0.023
443.8	29.3	6.43	2.14	0.414	0.371
779.1	37.4	9.59	3.24	0.743	0.061
964.4	38.8	10.49	3.82	0.903	0.125
1112.2	41.7	11.91	4.61	1.217	0.252
1408.1	43.7	13.80	5.70	1.831	0.556
Total	25.5	6.48	2.69	0.891	0.405

$$\text{Percent Transmission} = \frac{\text{Photopeak count rate with collimation}}{\text{Photopeak count rate without collimation}} \times 100\%$$

$$\text{Photopeak count rate} = (\text{counts per second at } E_{\gamma} - \text{background counts per second at } E_{\gamma})$$

Figure III-2. Dependence of % transmission on collimator diameter

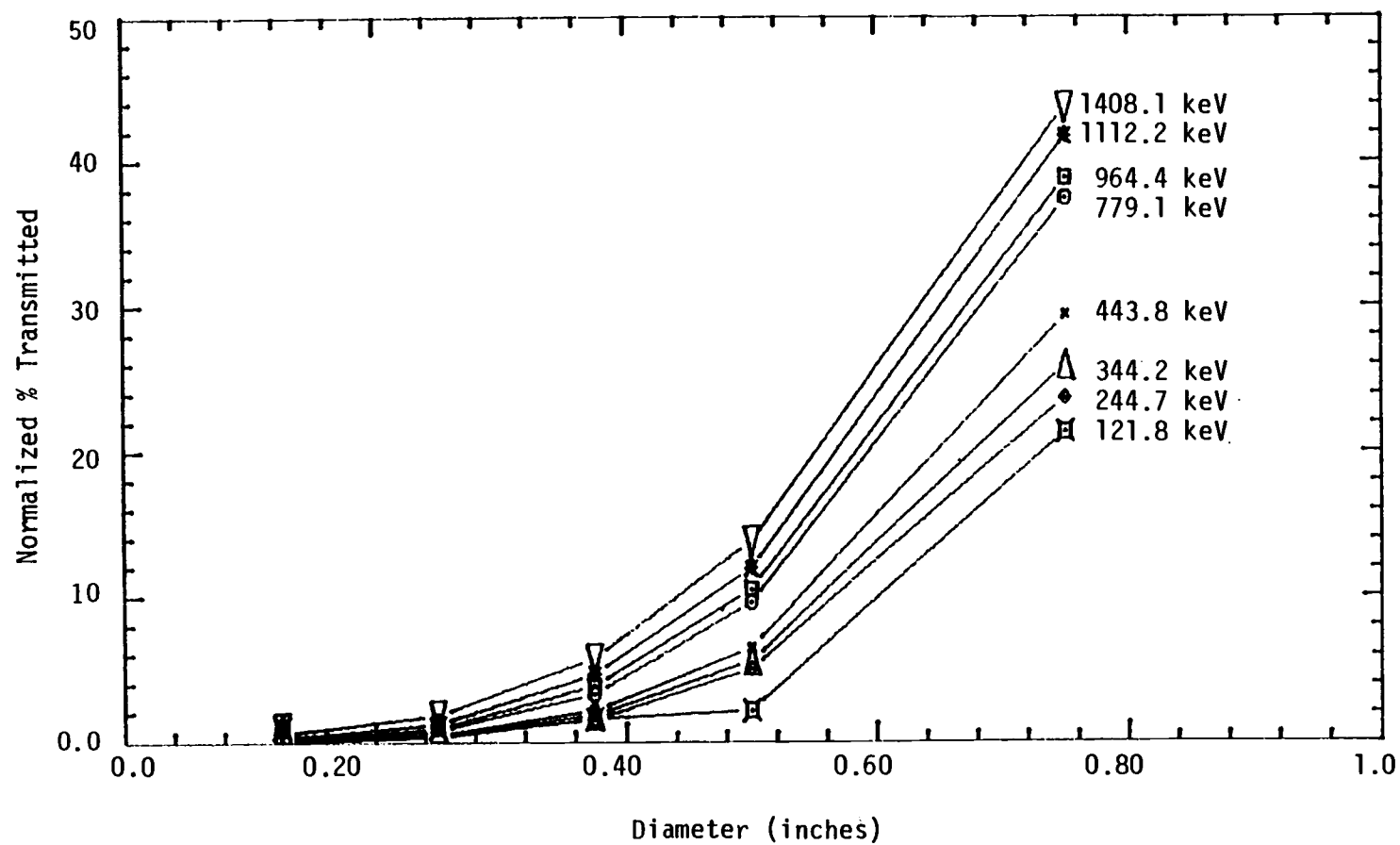


Figure III-3. Dependence of % transmission on energy

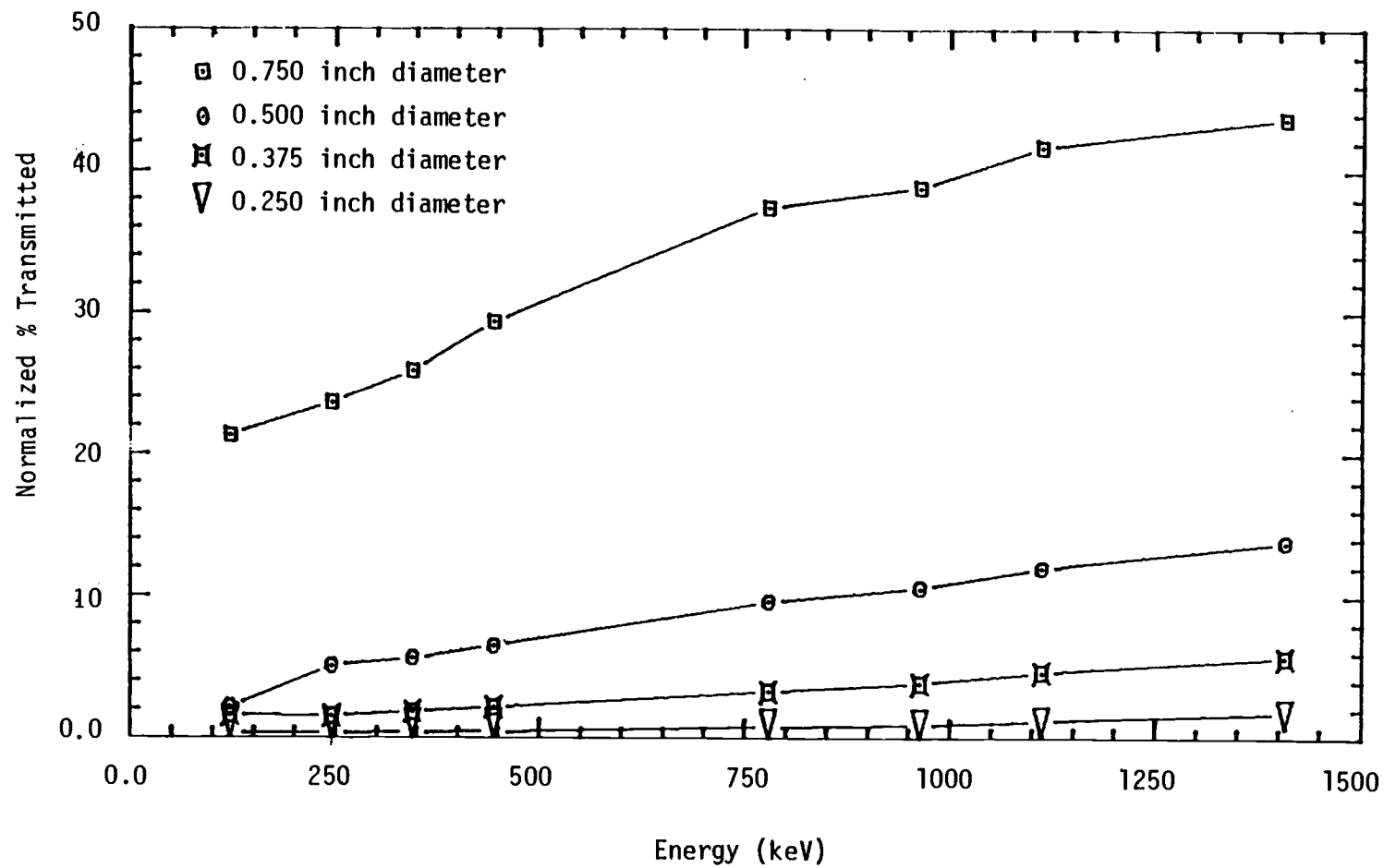


Table III-4

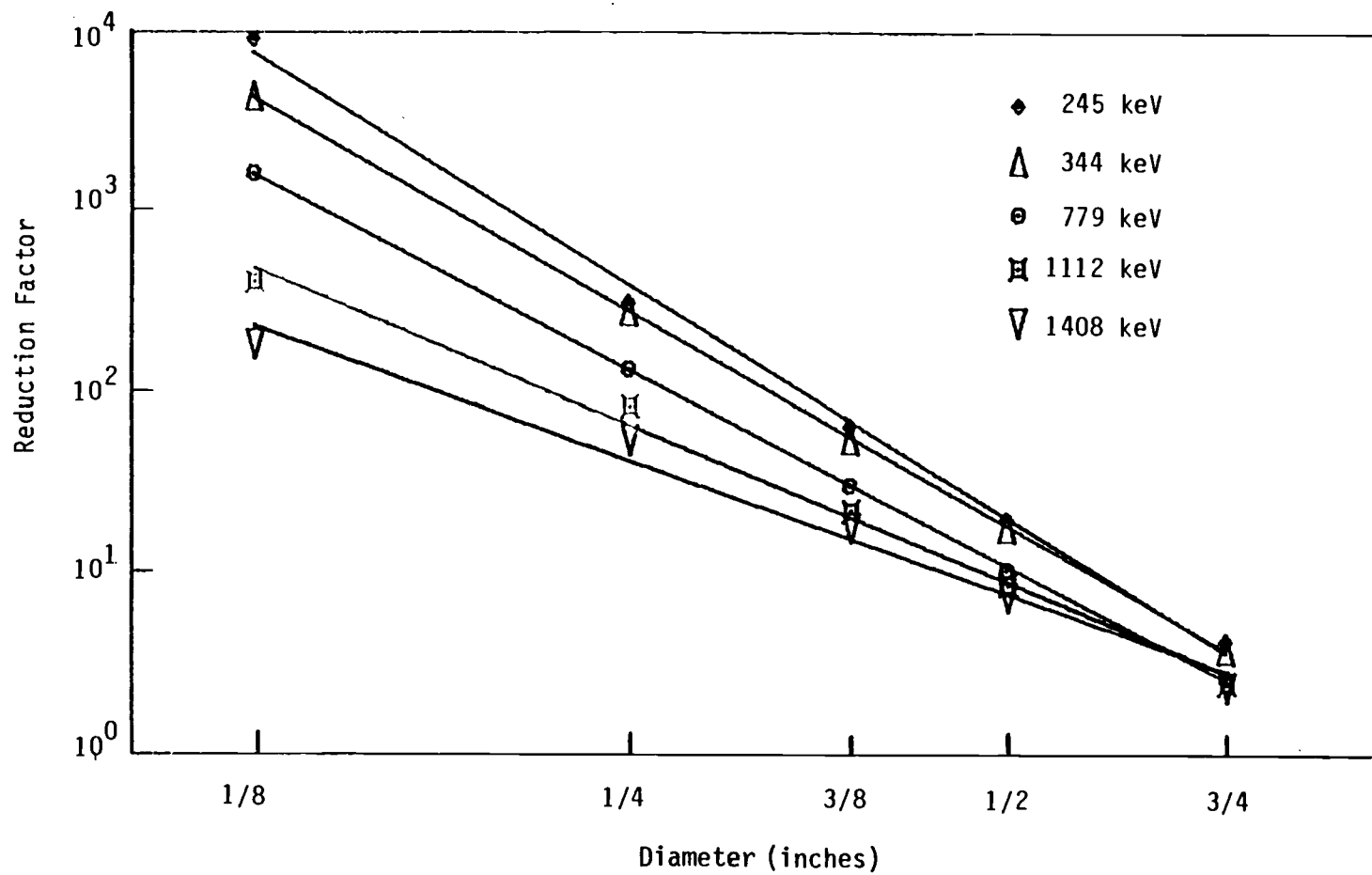
## REDUCTION FACTORS

$E_{\gamma}$ (keV)	APERTURE DIAMETER (inches)				
	3/4	1/2	3/8	1/4	1/8
121.8	4.70	47.0	64.5	315	4762
244.7	4.24	19.9	65.3	312	9091
344.2	3.86	18.0	55.6	283	4348
443.8	3.41	15.6	46.7	242	270
779.1	2.67	10.4	30.9	135	1639
964.4	2.58	9.5	26.2	111	800
1112.2	2.40	8.4	21.7	82	397
1408.1	2.29	7.2	17.5	55	180

$$\text{Reduction Factor} = \frac{\text{Photopeak count rate without collimation}}{\text{Photopeak count rate with collimation}}$$

$$\text{Photopeak Count Rate} = (\text{Counts per second in photopeak at } E_{\gamma} - \text{background counts per second at } E_{\gamma})$$

Figure III-4. Dependence of reduction factor on collimator diameter



spectrum for a given aperture divided by the same quantity for the case of no collimator:

$$\%P/T = \frac{\frac{\text{Count rate in photopeak with collimation}}{\text{Count rate in total spectrum with collimation}}}{\frac{\text{Count rate in photopeak without collimation}}{\text{Count rate in total spectrum without collimation}}} \times \%100$$

It was selected as a parameter that would indicate the effect of collimation on spectral quality.

Both elevated (above 100%) and depressed (less than 100%) ratio resulted. The effect was highly dependent on gamma ray energy and aperture size. A generated series of curves of %P/T versus aperture diameter shows severe energy dependence, with photopeaks especially depressed at lower energies and smaller orifice sizes (refer to Table III-5 and Figures III-5 and III-6). At low energies (nominally 300 keV and less) the curve continually rises with increasing diameter but never attains 100%. At intermediate gamma ray energies (nominally 400 to 1000 keV) the curve rises, reaches a plateau and subsequently levels out at 100 to 150%. With high gamma ray energies (nominally greater than 1100 keV) the curve initially rises, levels off, and subsequently drops. At the highest gamma ray energy, the %P/T is greater than 100% for all aperture sizes and achieved an apparent enhancement of up to about 200%. However, there is no real increase in photopeak information at higher energies since the denominator of the ratio is reduced, rather than the numerator being

Table III- 5

## PERCENT-PEAK-TO-TOTAL COUNT RATE RATIOS

$E_{\gamma}$ (keV)	APERTURE DIAMETER (inches)				
	3/4	1/2	3/8	1/4	1/8
121.8	83	33	58	36	5
244.7	93	78	57	36	3
344.2	102	86	67	40	6
443.8	115	99	80	47	92
779.1	147	148	121	84	15
964.4	152	162	142	101	31
1112.2	163	184	171	137	62
1408.1	171	213	212	206	137

$$\% \text{ Peak to Total Ratio} = \frac{\text{P/T with collimation}}{\text{P/T without collimation}} \times 100\%$$

$$\text{P/T} = \frac{\text{Count rate in photopeak}}{\text{Count rate in total spectrum}}$$

$$\text{Photopeak count rate} = (\text{Counts per second in photopeak at } E_{\gamma} - \text{background counts per second at } E_{\gamma})$$

Figure III-5. Dependence of % peak to total ratio on collimator diameter

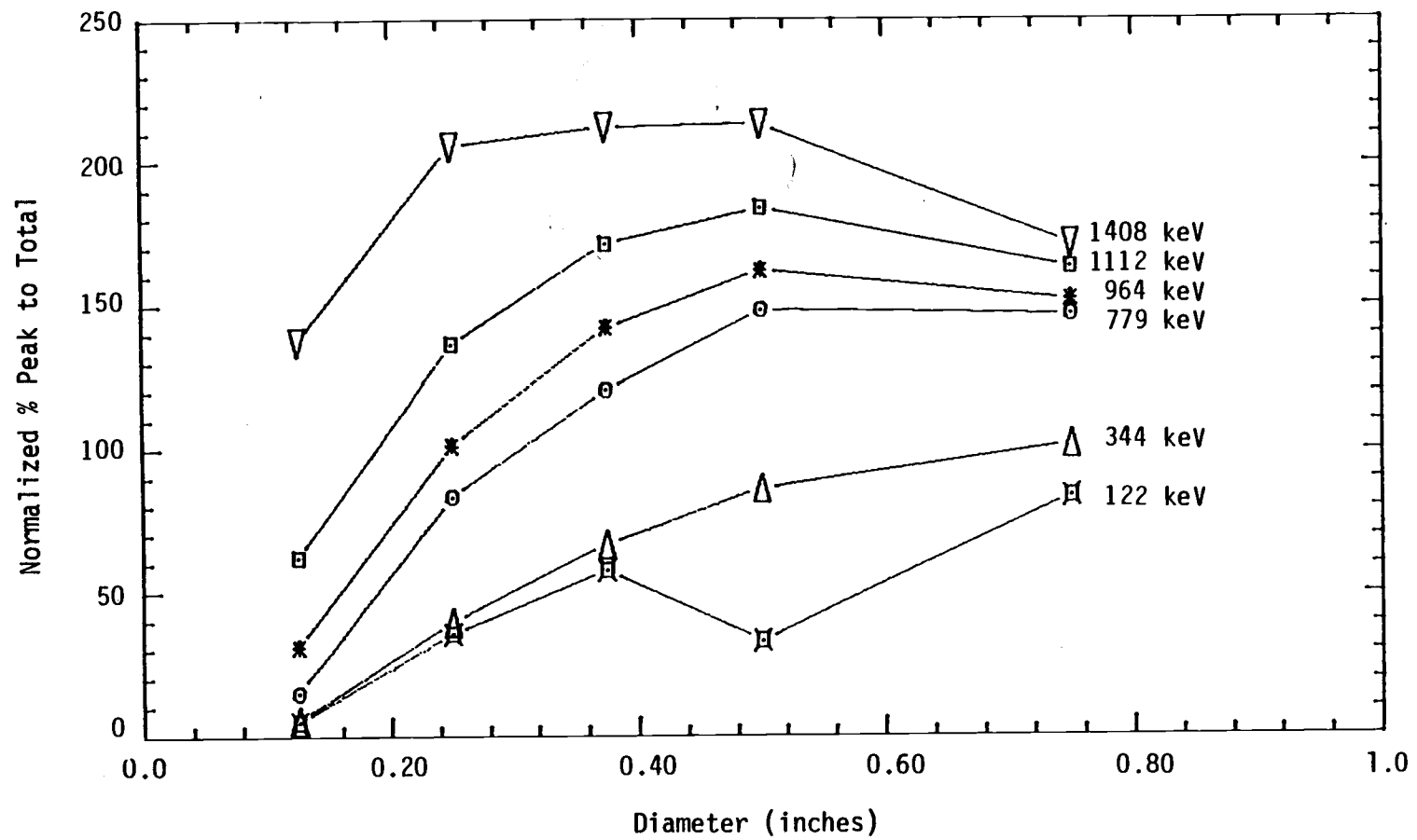
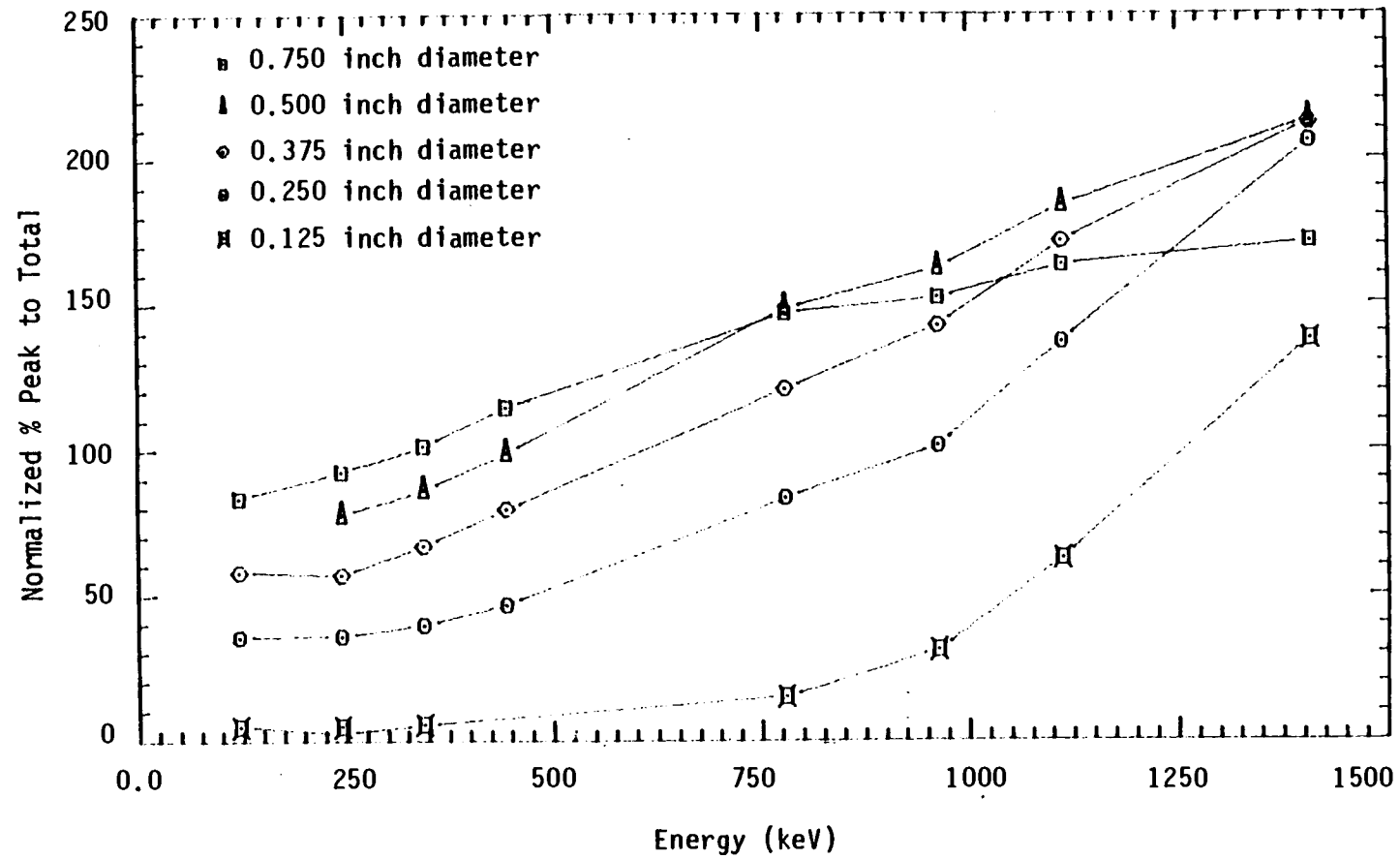




Figure III-6. Dependence of % peak to total ratio on energy



increased. This is a result of greater attenuation of lower energy gamma rays than those of higher energy.

### Conclusions

The different energy dependences observed in the peak-to-total ratios and in the percent transmission can be explained by two effects. The higher energy gamma rays experience less attenuation through a given thickness of lead than lower energy gamma rays. Therefore, a greater probability exists for higher energy gamma rays to reach the detector than those of lower energy. The probability of any gamma ray emitted by the  $^{152}\text{Eu}$  source traversing the four inch thickness of lead unattenuated is minimal.

The second effect is due to Compton scattering. Relatively more Compton scattering will occur for smaller aperture openings, having the effect of producing a higher Compton continuum at lower energies, thus tending to obscure lower energy photopeaks to a greater extent than higher energy photopeaks.

In selecting an optimal aperture diameter, two separate collimation-induced effects must be considered, namely the effect of collimation on spectral quality and the reduction in count rate. From the data presented, it appears that collimator diameters below about 1 cm result in sufficiently reduced spectral quality so as to no longer justify the decrease in count rate obtainable for these smaller apertures. For example, although the reduction in count rate achieved by reducing the aperture diameter from 1/4 to 1/8" was

considerable, the reduction resulted in a significant degradation of spectral quality, as evidenced by a reduced %P/T ratio for all energies relative to other diameters. On the other extreme, a significant reduction in percent transmission occurred by reducing the aperture diameter from 3/4" to 1/2", without an accompanying severe degradation in spectral quality, except at very low energies.

The theoretical upper limit for aperture size is dependent only on the count rate reduction factor desired. However, spectral degradation for small apertures represents a practical limitation to the amount of count rate reduction obtainable by the collimation techniques. The practical range is nominally from one to two centimeters in diameter if using orifice collimator geometries.

With one exception, the experimental values observed with the various aperture sizes varied smoothly. However, the values obtained with the 1/8" aperture tended to fluctuate more, revealing an unpredictability with extreme collimation. The degree of fluctuation observed was greater than that attributable to counting statistics at extremely low count rates. Such unpredictability would make calibration schemes at extreme collimation less reliable.

Because of the strong and differing energy dependence for each aperture size and source geometry, efficiency calibrations must be performed independently at each aperture for a series of gamma ray energies in order to generate adequate calibration curves.

Although the data presented in this chapter represent the spectral composition of the particular radionuclide used ( $^{152}\text{Eu}$ ), a

similar effect can be expected to result for other complex gamma ray emitters.

For post-accident monitoring the count rate range obtainable by collimation alone does not meet the current NRC requirement of six orders of magnitude. The practical dynamic range achievable by using collimation, where spectral degradation is insignificant, is only about two orders of magnitude (refer to Table III-4). Therefore, other techniques in conjunction with collimation must be used to sufficiently extend the dynamic counting range.

## CHAPTER IV.

### THE ASSESSMENT OF CORE CONDITIONS BY REACTOR COOLANT ISOTOPIC ANALYSIS

#### Introduction

The NRC requires that a "group" of key radionuclides which may be indicative of the extent of core damage be analyzed under abnormal core conditions, although the specific radionuclides comprising the group is left to the discretion of each licensee.<sup>5</sup> The NRC further requires that procedures for the analysis of these radionuclides be prepared. By using the isotopic analysis technique as part of the post-accident monitoring program, it is hoped that the onset of conditions leading to core melting will be indicated by alterations in the reactor coolant isotopic profile and operators will thereby be alerted to take necessary corrective actions.

In addition to a primary role in post-accident monitoring, reactor coolant isotopic analysis can to some degree indicate the extent, size, batch, and location of fuel failures in a core under normal operating conditions.<sup>40</sup>

In spite of the rather broad guidance presently given by the NRC for this type of post-accident monitoring, coolant isotopic analysis remains as one of the potential techniques for gathering information on the integrity of the reactor fuel. As a result, information has been compiled in this chapter which identifies a group of

radionuclides that can serve as indicators of core condition, describes methods by which reactor coolant isotopic analysis can be used to diagnose core conditions under both accident and routine operating environments, and identifies problems associated with such methods of analysis.

### Theory

Core diagnosis by reactor coolant isotopic analysis is based on the premise that certain types of fuel failures and core conditions will result in unique patterns of fission products being released into the reactor coolant. Factors commonly affecting fission product concentrations ultimately observed in the reactor coolant include:<sup>40</sup>

1. Fission product source term in the fuel;
2. Chemical interactions within the uranium dioxide ( $\text{UO}_2$ ) fuel matrix;
3. Migration patterns within the  $\text{UO}_2$  fuel matrix;
4. Release rates and release mechanisms from the  $\text{UO}_2$  fuel matrix;
5. Chemical interactions within the fuel pins;
6. Release rates from cladding breaches;
7. Mass balance in the reactor coolant.

Each of these factors is further influenced by parameters such as temperature, pressure, oxygen potential, etc. Even though the variables affecting the concentrations of fission products in the reactor coolant are multiple and complex, a careful selection and

analysis of "key" radionuclides can give an indication of the condition of the core if the factors determining their relative abundance are adequately understood. However, this latter consideration is the most difficult hurdle to overcome in the application of the technique.

### Additional Indicators of Core Conditions

With the exception of gross beta-gamma radioactivity measurements, none of the other methods for obtaining information about core conditions, such as indicators for containment radioactivity (airborne and general), core and reactor coolant temperature, containment hydrogen ion concentration, and core water level, are comparable to reactor coolant isotopic analysis in providing information about the nature and timing of fuel damage under conditions of severe core degradation as well as under normal operating conditions.<sup>1</sup> The type of information supplied by the other measuring systems is general, indicating whether conditions are normal, and only providing a rough approximation of the amount of deviation if abnormal.

A gross beta-gamma radioactivity measurement indicates overall reactor coolant concentration changes and provides a fast, unambiguous signal, without requiring a sophisticated system. However, increases in the gross reactor coolant activity (e.g., iodine spikes) can occur without severe fuel damage. In that case, only isotopic

analysis can provide information useful in determining the cause of elevated activity.

### Source Term

In evaluating the significance of a particular fission product concentration in the reactor coolant, the core inventory must be taken into account. The fission product source term in irradiated fuel is a function of various formation and removal processes: formation by direct fission yield, decay of precursor nuclides, neutron capture of isotopes one atomic number lower, and losses by decay, neutron capture and escape from the fuel pin. Formation of fission products by direct yield is greatest for those elements having atomic mass numbers around 95 and 139, including the noble gases (krypton and xenon) and several iodine radioisotopes. In addition, the fission product inventory is not homogeneous within irradiated fuel because of the neutron flux and temperature gradients.

### Fission Product Chemistry

Since fission product chemistry influences release mechanisms from the  $\text{UO}_2$  fuel matrix, affects the release rates from cladding breaches, and determines transport behavior in the reactor coolant, it plays a large role in determining reactor coolant fission product profiles. The chemical behavior of fission products within the fuel pellet is poorly understood, especially under conditions of elevated



core temperatures.<sup>41</sup> Because the chemical form and reaction rates are dictated largely by the ambient physical and chemical environment, alterations in temperature, pH, oxygen potential, pressure, etc. change fission product chemical behavior and therefore release patterns. Models should account for this effect, since the physiochemical environment can be considerably altered under abnormal core conditions.

An example of the effect of fission product chemistry is illustrated by the reactivity of different forms of iodine under various core conditions. Thermodynamic studies, in addition to conclusions from experiments, indicate that the major form of iodine in  $\text{UO}_2$  fuel and the fuel gap region under normal core conditions is  $\text{CsI}$ .<sup>42,43</sup> Another important form is elemental iodine ( $\text{I}_2$ ), although lower in concentration. The different chemical behavior of  $\text{CsI}$  and  $\text{I}_2$  result in dissimilar release rates. Since  $\text{CsI}$  is more reactive and tends to plateout on inner fuel and cladding surfaces, it is held up and appears in the reactor coolant in lower concentrations than if the chemical form were  $\text{I}_2$ , which is more volatile and not nearly as reactive. The oxidation state also plays a significant role in dictating the chemical form of iodine; this effect is illustrated by tests in which the major form of iodine released was  $\text{I}_2$  rather than  $\text{CsI}$  when fuel was melted under dry air conditions.<sup>44</sup> Conversely, fuel melted in environments of steam and/or water resulted in  $\text{CsI}$  being the predominant form released. Abnormal core conditions that occur in an

accident can result in highly altered physiochemical conditions and consequently variable reactor coolant iodine concentrations.

Chemistry of parent fission products can also play an important role in the release rates and migration patterns of daughter radionuclides. For example, all important iodine isotopes (except  $^{135}\text{I}$ ) were originally in the core for a considerable time as tellurium; therefore tellurium mobility and chemistry play an important role in the overall picture of iodine release, especially since tellurium is relatively volatile and displays a chemical affinity for cesium. Likewise, the noble gases, being more readily released and mobile than their daughters, can increase the reactor coolant concentrations of their daughter elements as well. This may be especially significant if these daughters also serve as indicators of specific core conditions.

Fission product release rates from cladding defects are not only dependent on gap inventories, but also on chemical reactivity with internal cladding and pellet surfaces. Reactivity effectively reduces the apparent gap inventory available for release. The degree of reactivity varies with the specific fission product, being the lowest with the noble gases. Because fission product chemistry may dictate release rates out of given defects, an analysis of the ratios between certain elements of diverse chemical reactivity can supply some useful information. As an illustration, iodine fission products are released into the gap spaces from fuel pellets at about the same rate as the noble gases; however their release rates out of cladding

failures are significantly less because the predominant form of iodine (CsI) reacts readily whereas the noble gases do not. Therefore, an analysis of ratios of iodine and noble gas fission products may provide an indication of cladding defect size.

Transport behavior of fission products in the reactor coolant is determined primarily by their chemical characteristics. Ideally "key" indicators should be readily soluble in the reactor coolant; however most fission products are capable of either separating out of phase or plating out onto adjacent surfaces. Since those that plateout have effectively lower concentrations, an assessment of core conditions based on assumed release fractions will be in error unless the removal by plateout is properly allowed for. However, it may be difficult to quantify the fractional plateout of various elements, and even then, the rate of plating out can be altered by changes in the physiochemical environment during abnormal conditions. The tracking of trends in the relative abundance may reduce this source of error. Also, if it is assumed that all isotopes of the same element have identical chemical forms, then plateout would not be a problem in the analysis of ratios of the same element, since plateout presumably will occur in equal proportions. Some fission product gases (notably the noble gases) readily separate out of aqueous media, with the tendency being greatest when the physical and/or chemical environment is altered. Unless careful analytical procedures are followed, separated gases can be lost from a grab sample resulting in the analysis of an unrepresentative sample.

### Release Mechanisms

The release of fission products from fuel may occur by three different routes:<sup>40,45</sup>

1. Recoil: The direct recoil of a fission product after the fission event leading to release from the fuel at birth. This mode of release is available only for fissions occurring at a distance from the fuel surface which is less than the average range of the fission fragment.
2. Knockout: The release of the fission product inventory from a small volume of fuel that has been vaporized by the intense energy dissipation from a fission fragment.
3. Diffusion: The migration of fission products through the  $UO_2$  lattice to trapping sites often at fuel grain boundaries.

The principal results of several studies indicate that, at reactor steady-state fuel temperatures, knockout is the predominant release mechanism for fission products, while recoil and thermal diffusion processes play minor roles. However, release rates by diffusion increase with temperature and above 1200 °C become significant.<sup>40</sup> The diffusion of fission products through the grain of  $UO_2$  is a very complex process which cannot be readily modeled. Fission product release rates and mechanisms vary greatly depending on half life, parent-daughter relationships, chemistry, fuel centerline

temperature, fuel burnup, fuel oxidation, and position of a failure along a fuel pin.<sup>1,40,45</sup>

Noble gas behavior in the reactor core is primarily characterized by the limited capacity of the  $UO_2$  fuel to retain these gases. Their release is aided by grain boundary tunnels. Thus the noble gases tend to collect in the fuel-to-cladding and plenum spaces from which they can be released into the reactor coolant by cladding defects and serve as initial indicators of the same. It may be possible to provide more information on the batch in which a given failure is occurring due to the fact that the network of grain boundary tunnels is increased with fuel burnup.<sup>46</sup>

Iodine isotopes have no chemical affinity for uranium dioxide and, unlike the noble gases, their fractional release is independent of the level of burnup.<sup>42</sup> However, iodine tends to move into the gas phase with the noble gases. Iodine release rates are comparable to those of the noble gases if the thermal power is cycled or when the cladding defect is close to the fuel pin centerline; otherwise the rates are lower as a consequence of chemical interactions with cladding inner surfaces.<sup>47</sup> Both iodine and cesium migrate radially within the fuel matrix, moving down the temperature gradient and collecting in the cooler fuel pin portions.<sup>41</sup>

The speed of various release mechanisms will affect the release rates of fission products, depending upon their half lives. Models indicate that short-lived fission products are released mainly by recoil and knockout, which are both temperature independent

phenomena, whereas nuclides with long half lives are also released by temperature induced migration, where elevated temperatures increase the rates of diffusion.<sup>40</sup> This distinction can serve as a basis for indicating high temperature releases and for locating failures within the reactor core because of the dependence of fuel centerline temperature on neutron flux and therefore core location. This can be accomplished by analyzing certain short- to long-lived fission product ratios. Using radionuclides of the same element would be the most useful since the main variable in that case would be half life with chemical effects cancelling out.

A nonhomogeneous distribution of fission products along a fuel pin due to migration along temperature gradients and power differences may result in variable fission product releases from defects of similar size depending on the location of the failure along the pin. This can complicate an analysis and should be recognized as a variable that may not be readily taken into account.

Only those elements that are volatile or gaseous at normal operating temperatures are free to migrate throughout the ceramic fuel matrix.<sup>26</sup> However, the majority of fission products are refractory, being retained by either trapping or chemical reactions with fuel constituents. The migration of radioisotopes to cooler fuel regions results in higher concentrations at the fuel pin end and peripheral areas; this causes the effective fuel inventory available for release (especially if defects are located in fuel pin periphery) to be higher than indicated by total fuel inventory.

Release rates out of cladding failures are dependent on three main variables: fission product gap inventories, half lives, and chemical interactions with inner fuel pin and cladding surfaces. The gap inventory is a function of the fuel source term and release mechanisms of either precursor radionuclides or the specific fission product itself. The half life dictates the average lifetime of a given radionuclide in addition to the buildup of daughter elements. Short-lived fission products may not have sufficient time to migrate in large quantity into the gap spaces (dependent on burnup and temperature) and may undergo considerable decay before release out of cladding breaches. Certain fission products are more reactive with inner fuel pin and cladding surfaces than others and therefore plateout. Chemically inert fission products are released in greater quantity than those that are highly reactive; however physiochemical alterations can change fission product chemical reactivity. Moreover, steam bathing or reactor coolant water on the inner portions of the fuel pin can wash out those radionuclides that have plated out onto the inner fuel pin and cladding surfaces, resulting in elevated release fractions.

#### Mass Balance

The mass balance of fission products in the reactor coolant is a function of several processes affecting fission product escape into and removal from the reactor coolant. It must take into account leaks from cladding breaches, removal by reactor purification

systems, leaks from the reactor coolant system, radioactive decay and altered physiochemical reactor coolant conditions such as might be induced by an accident.<sup>40</sup> The escape and removal rates vary with radioisotope, so that mass balance equations are unique for each fission product. For instance, iodine tends to be removed at a faster rate by purification processes than other fission products as was demonstrated by some tests on failed fuel conducted at the Power Burst Facility at the Idaho National Engineering Laboratory. It was found that iodine concentrations in the reactor coolant declined much faster than expected from decay alone. The observed behavior, which also was unlike that of other closely monitored fission products, was determined to result from changing reactor coolant conditions which effectively induced a quicker plateout rate than under typical reactor coolant conditions.<sup>48</sup>

Mass balance relationships will be somewhat in equilibrium under steady-state reactor operations, whereas upon reactor shutdown and startup they will be in a continually dynamic state, making the proper interpretation of fuel conditions based on reactor coolant fission product profile largely dependent on recent power history.

### Complicating Factors

Because of the number and complexity of the variables involved in generating reactor coolant fission product activities, correlation with specific defects in the reactor fuel is often imprecise.<sup>49</sup> These variables are listed below and are discussed in the following



subsections:

1. Multiple fuel failures
2. Failure development with time
3. Irradiation history
4. Defect location
5. Monitoring techniques
6. Background radioactivity
7. Inadequate data base

#### Multiple Fuel Failures

Most operating power reactors have at least a few fuel pins in the core which have small defects in the cladding. Fission product releases from all fuel cladding failures accumulate in the reactor coolant, making a correlation of fission product activity with any specific type of failure complex and imprecise. Multiple defects most likely will not be similar in size, fuel burnup, core location, etc. Also, varying amounts of fission product activity may be released from a single defect.<sup>49</sup> The presence of multiple defects alone makes an analysis of reactor coolant fission product concentrations a poor source of information for determining defect size, fuel batch, location, etc. For example, it may not be readily apparent whether the reactor coolant isotopic pattern is a result of one large defect from a relatively new fuel element or several smaller defects from older fuel elements.<sup>50</sup> Long-term (coupled with short-term) tracking of trends in fission product activities may

minimize this problem by supplying information on individual defects as they develop. However, more than one defect may develop simultaneously, complicating this analytical technique as well.

#### Failure Development with Time

The time frame over which failures develop and subsequently produce changes in the reactor coolant fission product profile must be considered in assessing the core condition. Studies conducted on defective fuel elements at the Atucha Nuclear Power Station in Argentina indicate that it is not possible to determine the precise time of fuel failure onset.<sup>51</sup> The rate of onset and development of cladding defects can vary greatly depending on the circumstances and causes of the failure, where failure development may occur rapidly in a few days or slowly over many months.<sup>52</sup>

Usually minor cladding failures remain quite innocuous; however for reasons not yet clear, a few propagate rapidly into large defects.<sup>53</sup> The onset of minor fuel failures may produce unrecognizable changes in the reactor coolant fission product concentrations while comparing day to day spectra. Long-term tracking of reactor coolant fission product concentrations over an extended time period (taking into account expected changes in reactor coolant background components over the same time period) can facilitate the detection of such minor defects.

Unlike in the development of minor defects, an instantaneous major fuel failure where no defect previously existed may produce a

sudden increase in reactor coolant activity.<sup>52</sup> If an instantaneous large failure occurs simultaneously with power reductions, it could be mistaken for spiking however.

More severe conditions leading to severe core degradation can be anticipated to occur rapidly over a period of several hours, with subsequent massive fuel fracturing. In such a situation an instantaneously large increase in coolant concentrations will be observed.

### Irradiation History

Fuel burnup affects both the magnitude and diversity of fission products that are released into the reactor coolant out of cladding failures so that the fuel fission product inventory and subsequently the gap inventory, continually changes with fuel burnup. The effect is also in part due to neutron irradiation progressively producing physical changes within the fuel pellet matrices, affecting certain release rates. For example, the release rates of fission gases from the  $UO_2$  matrix are enhanced due to a buildup of grain edge and grain boundary porosity.

Certain fission product ratios, predominately  $^{134}Cs$  and  $^{137}Cs$ , are evaluated in order to determine the degree of burnup of rods in which failures are located.<sup>58</sup> Since the concentrations of  $^{134}Cs$  and  $^{137}Cs$  will depend on their relative formation and decay rates integrated over time, and since their half lives are relatively long (about 2 and 30 years, respectively), their concentrations will

always be increasing (although asymptotically) under normal operating conditions. Thus an indication of the degree of fuel burnup can be obtained because the principal variable affecting cesium concentrations is then the integrated formation rates, which are in turn directly related to the degree of burnup. Taking the ratio of the two isotopes serves to eliminate some variables that may affect the ultimate measurement of their concentrations in the reactor coolant, if it is assumed that the two isotopes behave chemically very similarly. Also, if a simplified model assumes the diffusion rates are large compared to the cesium decay constants, then the ratio will be the same in the reactor coolant as it is in the fuel.<sup>40</sup> Problems associated with interpreting cesium ratios include the influence of crud deposit phenomena occurring in the reactor coolant system,<sup>54</sup> nonlinearity with irradiation, increasingly smaller alterations in the ratio as fuel burnup progresses, differentiating between simultaneous defects in the fuel pins as well as between simultaneous fuel failure occurrences, and the difficulty of detecting reactor coolant cesium concentrations because of low activities, requiring a time delay (2 to 5 days) for the decay of shorter-lived radionuclides.

If cladding failures are large enough to allow the release of fuel pellet material, analysis of the ratios between the actinides (mainly uranium and plutonium) can also be used to assess fuel rod burnup, since the ratios also vary with the intensity of irradiation.<sup>40</sup> An advantage to using transuranic ratios is that the

rate of change in the ratios continues to be relatively large with increasing irradiation.

Cesium ratios (or any other) used in assessing burnup should be analyzed in view of other conditions which affect their reactor coolant concentrations. Rather than relying on just the cesium ratio on which to make the assessment, other fission product activities and/or ratios should also be considered to improve the accuracy of the analysis.

#### Monitoring Techniques

The sample acquisition and analysis techniques used can affect a core diagnosis by changing reactor coolant sample fission product concentrations and should be taken into account in the final analysis. For instance, noble gases can readily come out of solution in the process of acquiring a grab sample because of changes in temperature and pressure; hence the separated gases should be collected independently. This problem was illustrated by reactor coolant sample collection during the TMI incident where noble gases were unquantifiably lost out of post-accident reactor coolant samples during the process of reactor coolant sample acquisition, producing an unrepresentative sample and consequently a questionable indication of actual core conditions. For a more thorough discussion on the possible effects of specific analytical and sample acquisition techniques on core diagnosis, refer to Chapter II.

### Background Radioactivity

Activity increases in the reactor coolant resulting from fission product releases out of failed fuel can be obscured by high background activity levels, and high backgrounds (particularly spikes) can increase the difficulty and accuracy of any desired measurement. The major steady-state background components originate from activated corrosion products, such as  $^{58}\text{Co}$ ,  $^{60}\text{Co}$ ,  $^{59}\text{Fe}$ , and  $^{54}\text{Mn}$ , with additional amounts from fission products of tramp uranium and water activation products, such as  $^{16}\text{N}$  and  $^{17}\text{N}$ .<sup>23</sup> Background reactor coolant fission product concentrations for the conditions of each specific power plant should be assessed to establish a baseline from which to evaluate releases from fuel failures. Some background components cause more problems than others, being dependent on specific circumstances.

Activation of gases dissolved in the reactor coolant results in short-lived nitrogen radioisotopes, which can be a special problem because their activities are extremely large and they emit a profusion of gamma rays, many of high energy. Since they have short half lives (about 4 and 7 seconds), they pose no special problem under most monitoring conditions. However, with some on-line monitoring systems, because of a close proximity to the core, they can interfere considerably with the detection of fission product concentrations. The problem can be remedied, however, by incorporating a delay line which allows the nitrogen activation products to decay to negligible levels.

Of the activated corrosion components,  $^{60}\text{Co}$  is usually found in highest concentration in the reactor coolant. It can be significant since cobalt especially tends to plateout onto hot surfaces such as are found in the core region and consequently become activated en masse. Cobalt emits two major gamma rays of intermediate energies, making it easily distinguished from other radioisotopes. However, it contributes to the overall Compton continuum at lower energies.

Although steady-state background radiation can be somewhat difficult to deal with, the major problem occurs in differentiating sudden reactor coolant activity spikes generated during normal operating conditions from sudden reactor coolant activity increases due to abnormal core conditions or simultaneously occurring fuel failures. These activity spikes can be categorized as tramp uranium, corrosion products, or specific radionuclides such as iodine, noble gases or cesium. To eliminate the possibility of missing the occurrence of significant events by mistaking them for typical spikes, a means of determining the source of sudden activity increases must be implemented. An illustration of how this can be a problem occurred during the TMI incident. At about two hours after the initial reactor scram, the reactor coolant gross activity level was about 4 Ci/ml, which was one order of magnitude greater than the activity of a sample obtained before the reactor scrambled. However, this was not high enough to be of real concern to the operators since increases in activity of this magnitude are commonly observed following reactor shutdowns, although at that time the core was in an abnormal condi-

tion being partially uncovered.<sup>2,26</sup> Perhaps if means of distinguishing activity spikes from other causes of elevated activity had been implemented, the operators would have been alerted to the serious condition of the core.

Tramp uranium spikes can arise spontaneously and can increase the reactor coolant activity by a factor of up to 100. Tramp uranium increases the reactor coolant concentrations of a broad range of fission products and the release to birth ratios of the fission products found are independent of their radioactive decay constants.<sup>40</sup> This release pattern can serve as a means of distinguishing tramp uranium spikes from other sources, since otherwise many of these fission products would not be detectable in the reactor coolant unless large fuel failures occurred exposing  $UO_2$  directly to the reactor coolant. As a result, certain computer models which evaluate fuel failures compensate for tramp uranium contributions by correcting for the recoil components.<sup>40</sup> With massive fuel failures, subsequent extreme activity levels would be much greater than those from tramp uranium so that corrections would only be necessary while evaluating fuel defects during normal operating conditions.

Activated corrosion product (crud) spikes are generated by demineralizer changeouts, addition of hydrazine, or power fluctuations with the accompanying temperature changes which tend to cause accumulated crud to slough off from internal surfaces. The main crud components are cobalt, manganese, iron, chromium, and zinc activation



products in the form of complex ions. These species complex with stainless steel and tend to dissolve in water. Because the radionuclides in activated corrosion products are not formed by direct fission in significant quantities, crud spikes can be readily distinguished from other sources. However, the problem of spectral interferences from both an increase in the Compton continuum and similar energy photopeaks may exist.

The problem of spiking is the most significant with iodine radioisotopes, which can increase reactor coolant concentrations by a factor of up to 30 and persist up to three days.<sup>1,55</sup> Iodine spiking results from power changes altering the pressure inside fuel pins relative to pressure in the core. This allows water to enter a defective fuel pin and drive out readily soluble CsI, which has been previously plated out on internal cladding surfaces.<sup>54</sup> Iodine spiking behavior can serve as a technique for evaluating fuel failures, since it occurs in the reactor coolant only if there are one or more defective fuel pins in the core.<sup>55</sup> Additionally, spike characteristics are dependent on the number, size, and location of defects in the core. An improved data base and a greater overall understanding of spiking behavior could possibly enable the development of improved models for correlating spiking behavior (of all radioisotopes) with fuel defects in light water reactors. The feasibility of using this technique has been demonstrated at a CANDU power reactor in India.<sup>56</sup>

Although spiking occurs with other specific fission product ele-

ments, such as the noble gases and cesiums, the magnitudes are relatively minor compared to iodine spiking and their characteristics differ. Noble gas spikes only occur upon reactor startups, unlike iodine spikes which are generated by all power fluctuations. Also, iodine spikes always precede noble gas spikes, whereas spiking of long-lived cesium isotopes is identical in time behavior with iodine spikes but lower in magnitude.<sup>46</sup> The identical time behavior probably is a result of the major form of cesium being CsI, while the other major form of cesium (cesium uranate) is not readily released.

The ratio of  $^{134}\text{Cs}$  to  $^{137}\text{Cs}$  is often evaluated to assess the fuel burnup in pins where failures are found. However, their relatively low activities (because of long half lives and plateout) make detection difficult. Since cesium spiking increases reactor coolant concentrations of cesium radioisotopes, analysis of the spike may prove useful in the diagnosis of the fuel failure batch by making detection easier.

Large power fluctuations typically result in a normal spike activity, but concurrently the propensity for fuel failure (and resultant changes in reactor coolant isotopic profile) is also greatest during power transients. Normally occurring spikes can hide elevated temperature or fuel failure releases. Moreover, specific spike fission product activity ratios can interfere with the analysis of other ratios, reducing the accuracy of the analysis. For instance, iodine spikes predominantly contain longer-lived radioisotopes, such as  $^{131}\text{I}$  and  $^{133}\text{I}$ . This can be a problem since iodine ratios are frequently

used in core diagnosis.

Means of distinguishing spikes from other sources of elevated reactor coolant activity must be developed. This is possible since extensive investigations of spiking in various PWR's have shown that in all of the observed cases, the spike could be described by a certain set of characteristics.<sup>57</sup> For instance, in iodine spiking the peak value was reached 60 to 90 minutes after the beginning of the spike, the slope of the  $^{131}\text{I}$  activity increase exhibited a doubling time of about 10 minutes, and the  $^{131}\text{I}$  activity was higher than those for other short-lived iodine isotopes. Such information can enable identification of spikes, as well as possibly, with a thorough understanding of all processes, enable an assessment of fuel defects based on spike behavior.

#### Data Base

The data base for correlation of core condition to reactor coolant isotopic profile is small especially for abnormal conditions. The data base comes from three main sources:

1. Data gathered from operating light-water-cooled power reactors, where fuel defects and operating conditions are "real", but the size and complexity of the system make correlations with any specific defect difficult.
2. Data obtained from loop testing of fuel pins where defects have been introduced, and where fission product releases can be easily correlated with the type of de-

fect. However, results are only applicable to the specific environmental conditions of the test.

3. Data obtained from actual reactor accidents that have occurred.

The degree to which the data obtained from these three main sources can be applied to assessing core conditions in currently operating plants is yet uncertain. The data base is still incomplete, since the correlation of observed reactor coolant isotopic profiles with core conditions (under normal conditions) requires knowledge of the extent and types of existing fuel defects, which can only be assessed after refueling. Such examinations are rarely done and also require that certain assumptions be made to arrive at an experimentally determined defect level.<sup>58</sup> Obviously then, the data base is far less complete for accident conditions because of the few cases that have actually occurred, although experiments where fuel pins are overheated provide some information even if it is not precisely applicable to operating plants. Information obtained from most experiments suffers from an inability to accurately simulate reactor operating conditions, and studies of purposely defected fuel pins are applicable only to the specific defect type and conditions under consideration.<sup>58</sup> Therefore, prospects for rapidly improving the useful data base do not appear particularly promising.

#### Procedures for Assessing Core Conditions

The primary purpose of preparing procedures for analyzing a

group of reactor coolant samples containing varying concentrations of certain "key" radionuclides is to establish the ability to provide a realistic estimate of core conditions. The NRC has developed a guide to aid in the development of procedures to estimate core damage. In their guide, four major fuel conditions are identified, but there are further breakdowns within each broad categorization. The four main categories are:

1. No damage
2. Cladding failures
3. Fuel overheating
4. Core melt

In preparing procedures for assessing the core condition by analysis of the reactor coolant "key" radionuclide profile, three factors should be considered:

1. The percentage of the total available "key" radionuclide inventory (in the core) released (often expressed by release to total inventory ratios);
2. The "key" radionuclides not present in the reactor coolant;
3. The relationship of various "key" radionuclides in the reactor coolant to each other.

The NRC suggests that to make estimates of core damage as realistic as possible, assessment procedures should not only include the above-mentioned types of analyses, but also a weighted evaluation of their meaning based on all available plant indicators. Furthermore,

the operating conditions of the reactor (power, flow patterns, temperatures, volumes of the primary coolant components, etc.) are required for a systematic evaluation of the fuel pin failures.<sup>40</sup>

### "Key" Radionuclide Characteristics

Since an accurate core diagnosis depends on the ready detection and identification of a group of "key" radionuclides, they should possess certain characteristics such as:

1. Ability to represent a specific core condition;
2. Emission of high energy gamma rays;
3. Emission of more than one gamma ray in high yield;
4. Emission of gamma rays with unique energies distinct from other nuclides;
5. Half lives compatible with the monitoring system capability and time of analysis after sample acquisition and/or reactor shutdown; and
6. High core inventories.

Of the above considerations, "key" radionuclides must first of all be indicative of a specific core condition by their reactor coolant concentrations (individually or when compared to other fission product concentrations) or by their absence. For instance, the absence of rubidium in the reactor coolant indicates with high probability that core melting has not occurred, although its presence does not necessarily indicate the opposite.

The "key" radionuclides should have decay characteristics amen-

able to ready detection where extremely complex reactor coolant spectra occur. Indicators should also emit relatively high energy gamma rays to facilitate the detection of lower concentrations. Ideally, more than one prominent gamma ray should be emitted to allow computer validation of the radionuclide identities. The gamma rays selected for identification of "key" radionuclides should also have unique energies so that the possibility of spectral interferences will be reduced; however, the occurrence of spectral interferences is somewhat hard to predict (especially under conditions leading to severe core degradation), since the quantity of radionuclides present in a reactor coolant sample could be quite varied. See Chapter V for a more thorough discussion of spectral interferences.

Half lives of the selected radionuclide indicators should be compatible with the capability of the monitoring system. If possible, a wide range of half lives should be represented by the group of "key" radionuclides to cover a broad contingency of occurrences and delays in analysis after sample acquisition or reactor scram.

In addition to the above considerations, the key radionuclides should have core inventories, release rates out of  $\text{UO}_2$  fuel and cladding breaches, and absolute photopeak intensities sufficiently high to facilitate ready detection. Furthermore, chemical behavior should allow transport in the reactor coolant, and not cause plateout, precipitation, or phase separation to the point where their detection is meaningless.

### Specific Radionuclides

There is a small select group of fission product elements that have characteristics which result in relatively high release rates from cladding defects so that they are useful primary indicators of cladding breaches under normal operating conditions. In addition, they experience the greatest releases at elevated core temperatures and therefore may also be of value for assessing abnormal core conditions. This primary group consists of:

1. The halogens;
2. The noble gases; and
3. The cesiums

The characteristics which make this group unique are high core inventories, high concentrations in fuel pin cladding gaps because of rapid release rates from the  $UO_2$  fuel, rapid release rates out of cladding defects, suitable chemical behavior, and a number of useful radioisotopes covering a broad range of half lives and gamma ray energies. With some radionuclides, these features apply to precursor (parent) elements in addition to the specific fission products directly.

#### The Halogens

Because of their high yield, relatively rapid diffusion, fairly predictable behavior, and wide range of half lives, the iodine fission products have been the predominant industry choice as basic



indicators of fuel defects.<sup>58</sup> Ratios among different iodine radioisotopes are also frequently analyzed to assess defect size. Since the NRC also requires that iodine be specifically analyzed because of the public health hazard it poses, monitoring this group of radionuclides serves a dual function. The predominant form of iodine that is present within the fuel pellets and gap spaces is CsI, with elemental iodine ( $I_2$ ) also present but in lower concentration (<0.5%).<sup>1</sup> Elemental iodine is more volatile and mobile than CsI, since CsI tends to react more with inner pin and reactor coolant system components. Although iodine release rates from fuel pellets are comparable to the noble gases so that gap concentrations are equivalent, iodines have effectively lower concentrations in the reactor coolant due to reduced release rates out of cladding failures. However, iodine release rates out of cladding failures are comparable to noble gas releases if the thermal power is cycled or when the cladding defect is near the fuel pin centerline.<sup>47</sup> Once in the reactor coolant, iodine concentrations are suppressed to a high degree by plant purification systems so that removal rates are greater than for many other elements.<sup>6</sup>

Although bromine fission products have characteristics that could make them useful as "key" radionuclides, they have not been widely used in the analysis of core conditions. Bromine has many characteristics similar to iodine. Bromine tends to plateout like iodine, although at a higher rate reflecting its more reactive

nature. It also has a smaller core inventory, which is partially offset by a diffusion coefficient from  $\text{UO}_2$  fuel that is greater than iodines by a factor of about 200.<sup>52</sup> Bromine isotopes come in a wide range of half lives, although overall they tend to be shorter-lived than iodine radioisotopes. Because of their similar behavior, analysis of both bromine and iodine radioisotopes may be redundant, since they would indicate similar core conditions.

One bromine radioisotope which is specifically used by industry and serves a unique "indicator" function is  $^{87}\text{Br}$ . It is monitored as an indicator of large defects in CANDU reactors. Because it is highly water solubility (requiring leaching by the reactor coolant before significant quantities can be released) and since it is short-lived (55 second half life), detectable releases occur only from large cladding defects.  $^{87}\text{Br}$  is uniquely detected by monitoring for the delayed neutrons it emits. However, monitoring for delayed neutrons requires the use of an on-line neutron detector system.<sup>53</sup> Although the value of monitoring for  $^{87}\text{Br}$  has been demonstrated at CANDU reactors, this technique may not be feasible in light-water-cooled reactors because of major differences in plant design.

### The Noble Gases

The basic characteristics that make the noble gases highly useful as initial indicators of cladding failures include chemical inertness, rapid mobility, and high core inventories because of high direct and precursor fission yields. Monitoring for the noble gases

is not complicated by plateout or precipitation, although they readily separate out of reactor coolant samples. With the exception of  $^{87}\text{Kr}$  and  $^{88}\text{Kr}$ , noble gases also tend to emit low energy gamma rays making detection more difficult, although this is partially offset by high core inventories and release rates. Because of precursor decay,  $^{135}\text{Xe}$  and  $^{133}\text{Xe}$  concentrations remain relatively static for up to 24 hours after reactor shutdown, and the  $^{135}\text{Xe}$  activity eventually builds up to a maximum.

### The Cesiums

Cesium radioisotopes are frequently used as indicators of elevated temperatures and in the assessment of fuel burnup. Some of the cesium radioisotopes are formed only by direct fission, whereas others are formed by decay of parent xenon radioisotopes as well. The cesium radioisotopes that do not have xenon precursors are probably the most useful indicators of elevated core temperatures since xenon contributions do not have to be taken into account and xenon releases will be great with elevated core temperatures. The major chemical form of cesium under normal operating conditions appears to be cesium uranate,<sup>42</sup> although it also readily complexes with iodine to form  $\text{CsI}$ .<sup>44</sup> The two major forms behave differently. Cesium uranate is essentially bound within the fuel matrix, where  $\text{CsI}$  is more mobile.

### Computer Models

Extensive computer codes will be necessary to adequately take into account the complicating factors and power plant operating conditions which can affect the diagnosis of the core by analysis of key radionuclides in the coolant. The use of computer reduction for measuring data could be quick, reliable, probably more accurate, make alarm generation quicker, and speed the routing of information to the appropriate places. Ideally, such codes will also correlate all plant variables and supply error values for the final core evaluation.

Areas of greatest uncertainty that will require quantifying and incorporating into computer models to improve accuracy are:<sup>1</sup>

1. Fuel oxidation and its influence on fission product release and fuel centerline temperature;
2. Migration processes from fuel to cladding gap spaces; and
3. Defect development with time

Some computer models such as COFF (Babcock & Wilcox) and PROFIP (French Atomic Energy Commission) have already been developed for evaluating core conditions; they rely heavily on iodine concentrations. However, current computer models may not be highly accurate since several gross assumptions are made which, although greatly simplifying analyses, can introduce significant error.

### Indicators of Cladding Failures Under Normal Operating Conditions

A fuel failure is defined as a perforation in the cladding which consequently leaks fission products into the reactor coolant.<sup>60</sup>

Only those fission products which have previously collected in the fuel-to-cladding gap and plenum spaces will be available for release from cladding breaches. The fission products that collect in these spaces depend on fuel inventory of the fission product, parent-daughter relationships, half life of the fission product, release mechanisms from the  $UO_2$  fuel matrix, and fuel burnup. Therefore the profile of fission products found in the gap spaces will not resemble the profile found in the reactor fuel. Although several studies have been conducted evaluating the gap concentrations of different radionuclides, the actual inventories are not readily known since they are dependent on the degree of fuel burnup and thus continually changing.<sup>44</sup>

Models have revealed that short-lived fission products are released mainly by recoil and knockout which are both temperature independent phenomena, whereas longer-lived fission products are also released by temperature dependent migration. This effect, coupled with their longer lifetimes, accounts for the fact that the gap contents consist mainly of longer-lived fission products. Thus, for short-lived fission products to be detected in the reactor coolant, they must escape from both the  $UO_2$  fuel and cladding defects within a time period comparable to their radiological half lives.<sup>10</sup>

Ratios of iodines and noble gases (specifically  $^{131}I/^{133}I$ ,  $^{87}Kr/^{88}Kr$ ,  $^{135}Xe/^{138}Xe$ ) are frequently used to determine defect sizes relying on differences in half lives for information. Gap content or background reactor coolant ratios should be known, however, to serve

as a baseline for comparison. Unfortunately, this may be difficult to determine because of an incomplete knowledge of release dynamics and the additional influence of various parameters on the gap inventory, such as the degree of fuel burnup.

Because a small cladding defect provides an effective time delay between fission product birth and subsequent release, and because the pin provides many surfaces for chemical interactions, only the most volatile, inert, and long-lived fission products that have previously accumulated in the gap and plenum spaces can be released in significant quantity. Basically this only includes long-lived noble gases, halogens, and their daughters. The noble gases will be released in greatest quantity since they are chemically inert, extremely mobile, produced in high direct fission yield in addition to having high yield precursors, and are released by a unique mechanism from  $\text{UO}_2$  which facilitates their collection in the gap and plenum spaces.

Since larger cladding defects permit higher release rates of the gap contents, a wider range of noble gases and iodines will escape into the reactor coolant and a relative increase in the ratio of shorter- to longer-lived fission products will result.<sup>52</sup>

Defects large enough to expose fuel pellets directly to the reactor coolant can result in the loss of fuel pellet material by knockout and abrasive action of the reactor coolant, with the  $\text{UO}_2$  being lost either as individual grains or large particles broken out

of the matrix by grain boundary oxidation.<sup>46,61</sup> The amount of pellet material lost is proportional to the defect size and burnup; the degree of burnup also determines which specific fission products are ultimately found in abundance in the reactor coolant.<sup>61</sup> Defects large enough to produce gross fuel exposure will release a broad spectrum of fission product activities in the reactor coolant, either by the direct ejection of fuel particles or by leaching action of the reactor coolant. This will facilitate the release of otherwise immobile fission products. Since refractory radionuclides (e.g.,  $^{139}\text{Cs}$ ,  $^{125}\text{Sb}$ ,  $^{106}\text{Ru}$ ,  $^{106}\text{Rh}$ ,  $^{144}\text{Ce}$ ,  $^{154}\text{Eu}$  and the transuranics) are not volatile or readily mobile at normal operating temperatures, they can be used as indicators of gross fuel contamination and therefore large cladding defects.<sup>62</sup>

There are several problems associated with the detection of most refractory fission products and transuranics, namely, they tend to plateout readily and thus not travel far once released into the reactor coolant.<sup>63</sup> In addition, transuranics are also difficult to detect since they tend to emit soft gamma rays. One transuranic specifically analyzed is  $^{239}\text{Np}$ , since some data have shown sudden increases in  $^{239}\text{Np}$  activity after the first indications of defective fuel.<sup>58</sup>

The occurrence of certain fission product ratios can also indicate gross fuel contamination. One analysis involves a comparison of the  $^{131}\text{I}/^{133}\text{I}$  and  $^{131}\text{I}/^{135}\text{I}$  ratios found in the reactor coolant with their expected ratios in the fuel.<sup>23</sup> If the ratios are abnormally

low, the concentrations probably include a considerable recoil release component, indicating little decay of short-lived fission products by delay within the fuel pin prior to release. Such ratios are only found in the reactor coolant with direct fuel exposure.

Reactor coolant isotopic analyses may be much less reliable for evaluating fuel pin integrity once gross amounts of fuel has escaped, even after the removal of the defective fuel pins. The problem can arise from fuel becoming ground up and continually circulating or plating out onto surfaces, such as in the core region. This results in a "memory" effect, which produces a constantly high fission product background activity for a broad range of fission products, making the detection of other fission products released from fuel pin defects much more difficult.

#### Indicators Of Elevated Core Temperatures

Elevated temperatures are those above 800 to 1800°C for the fuel centerline and 650 to 800°C for the overall core. It is critical that operators be immediately alerted to conditions of elevated core temperatures, so that remedial actions can promptly be applied. Even though temperature increases can be indicated by thermocouple readings in addition to resultant increases in the gross reactor coolant activity levels, isotopic analysis is superior in that it can supply information about the extent of core damage in addition to providing a warning of an increase in core temperature.

At highly elevated core temperatures, two major effects occur in



the reactor coolant isotopic profile, namely there are tremendous increases in both the diversity of fission products represented and the gross radioactivity levels. At the onset of rising core temperatures, increases in reactor coolant activity are primarily a result of volatile fission products experiencing a more rapid diffusion out of  $UO_2$  fuel pellets and cladding breaches. Also, some previously non-volatile fission products become volatilized and therefore available for release.<sup>26</sup> Eventually with sustained temperature increases, massive fuel pin failures will result, exposing large amounts of irradiated fuel directly to the reactor coolant.

Large scale rupturing of cladding can occur by two separate mechanisms: heating until the cladding melting point is attained or weakening the cladding by oxidation (as a result of high temperatures) until the cladding is prone to fracture upon sudden stress.<sup>42,26</sup> During the TMI incident, fuel pins fractured en masse because of a sudden quenching of oxidized cladding with cold water.<sup>26</sup> This type of quenching action can lead to considerable fuel fracturing, or powdering as well, and the measured release fractions of the refractory elements may increase two orders of magnitude higher than expected.<sup>48</sup>

Fission products which can best serve as indicators of elevated core temperatures are those which are not readily volatile or mobile at normal operating temperatures, but which quickly become so as the temperature begins to rise. Such nuclides must not, however, be

highly chemically reactive. Since it is critical that operators be immediately warned of the onset of excessive core heating, a careful selection of these key radionuclides is very important, although information on which to base such a choice is limited.

The data base on fission product releases as a function of elevated core temperature comes predominantly from experiments conducted at testing facilities where simulated, or actual, irradiated fuel is heated. Also the incident at TMI provided some additional information. However, the available information may only apply to certain precise conditions.

The major indicators of initially rising core temperatures are:

1. Alteration in the ratios of longer-lived fission products to other fission products;
2. Increases in gross reactor coolant radioactivity levels;  
and
3. Increases in the number of noble gases, halogens, and alkali metals in the reactor coolant.

With core temperatures below about 1200°C, experiments have shown that the fission products released are only those which have already accumulated in the gap spaces, with additional releases from grain boundaries or the UO<sub>2</sub> fuel matrix being insignificant.<sup>64</sup> Above 1200°C, releases from the fuel by several other mechanisms become important. Therefore, as fuel temperatures initially begin to rise, the cumulative gap inventory dictates which fission products will be released into the reactor coolant. These are predominately the noble

gases, halogens and alkali metals.

Increases in ratios of longer-lived to shorter-lived fission product activities can alert operators to initially rising core temperatures, since long-lived fission products are released from the  $\text{UO}_2$  fuel predominantly by the temperature dependent mechanism of diffusion, whereas shorter-lived fission products are released mainly by knockout and recoil.<sup>40</sup> This effect can also serve as a means of estimating the average temperature of fuel with defects. An example of a useful ratio is  $^{133}\text{Xe}$  to  $^{135}\text{Xe}$ , since it increases steadily from 800 to 1800°C, whereas other ratios such as  $^{138}\text{Xe}$  to  $^{135}\text{Xe}$  do not vary.<sup>40</sup>

Cesium radionuclides can be useful indicators of rising core temperatures since significant releases are not expected below 1300°C, even though the precise temperature is not known and cesium releases can be quite variable. At about 1300°C, a change in cesium's predominant chemical form results, that is, below 1300°C cesium is highly reactive and readily retained in gap spaces, but at higher temperatures cesium behaves more like a gas, becoming volatile and easily released.<sup>65</sup> Probably the most useful cesium radionuclides to select as indicators are those without xenon precursors, since the contribution from xenon decay can complicate the analysis and xenon is also released in great quantity by rising core temperatures.

Fission products and actinides can roughly be divided into anticipated release groups, based on the ease with which they are

volatized as a function of temperature. Although not precise, since other factors also exert a definite influence (such as oxidation state, chemical interactions, etc.), such groupings can approximate the order in which fission products will be released into the reactor coolant as fuel temperatures rise.<sup>44</sup> One grouping arranges them in order of decreasing volatility, but such a listing provides only a rough estimate of release for the purpose of estimating fuel integrity.

1. Noble Gases (Kr, Xe)
2. Halogens (I, Br)
3. Alkali metals (Cs, Rb)
4. Tellurium
5. Alkaline earths (Sr, Ba)
6. Noble metals
7. Rare earths and actinides
8. Refractory oxides of zirconium and niobium

Another grouping arranges fission products into three broad categories (elements of high, medium, and low volatility) based on temperatures at which considerable releases can be expected.

The highly volatile category includes the noble gases, halogens, ruthenium, cesium, tellerium and cadmium. They are characterized by boiling points less than 1300°C for both the elemental and all known oxide forms. It is anticipated that core inventory releases will be complete when the fuel is heated from about 1700 to 2000°C, except for the cesiums, telluriums, and iodines which require temperatures

greater than 2000° C.<sup>66</sup>

Medium volatility elements are characterized by boiling points of the metal form, or at least one of the oxide forms, of less than 2700° C. However, releases can only be predicted with large uncertainty because of their chemical and physical properties and other phenomena.<sup>66</sup> Included in this grouping are radioisotopes of Mo, Tc, Ru, Rh, Ba, Cr, Mn, Co, Ni and Sn.

Although they cannot alert operators of the onset of rising fuel temperatures, low volatility fission products can be useful in assessing the degree of core damage generated by an accident. These fission products have boiling points greater than 3000°C. Thus, they will only be released under the most degraded core conditions, and then only in low concentrations.<sup>66</sup> They are primarily released from melted fuel by the slow process of reactor coolant leaching. Included in this category are Y, Zr, Ni, Np, Pu, Am and the rare earths.

The feasibility of reactor coolant isotopic analysis for the indication of elevated core temperatures is questionable. The accuracy of an analysis is dependent on the representativeness of a sample and in order for a sample to be representative it must be transported rapidly from the core to the sampling point. Elevated core, temperatures are caused by a loss of reactor coolant in the core making the requirement for sample representativeness hard to meet.

### Indicators of Core Melting

The temperature at which the core will melt is about 2800°C.<sup>42</sup> Indicators of core melting, although important in assessing the severity of core degradation, can only provide after-the-fact information and therefore are not as important as those "key" radionuclides which can alert operators to the initial conditions that will lead to a core melt. In addition, if a core melts, multiple plant measuring systems will reflect abnormal conditions. However, isotopic analysis could confirm the occurrence of melting where the other parameters would not supply as definitive information.

The data base of fission products that are indicative of core melting is small, coming basically from experiments, but the incident at TMI provides an excellent case study. A variety of experiments reported in the literature provide information on the magnitude of fission product release fractions from fuel under various conditions. However, because of large fuel surface to volume ratios in such experiments compared to that in a molten core, the experimentally derived release fractions would be greater than those anticipated with actual reactor meltdown conditions.<sup>67</sup>

It is estimated that from the most severely damaged fuel at TMI, nearly complete releases of fuel inventories of the noble gases, halogens, and most other volatile fission products occurred, whereas a major fraction of the alkali metals (Cs, Rb), a smaller fraction of tellurium, and only minimal concentrations of Sr, Ba, Ru, Rh, Pd, Mo,

Tc, Zr, Nb, and the actinides escaped from fuel which reached the highest temperatures.<sup>26</sup> The principal mechanism for release of these refractory elements was probably reactor coolant leaching, with rates being slow. The low fractions of tellurium and ruthenium specifically indicate that a small volume of the core actually melted. The extent of core damage incurred was more precisely estimated by analyzing the fraction of the total core inventory of  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ , and  $^{131}\text{I}$  found in the reactor coolant. The estimated damage was in remarkably good agreement with estimates based on the amount of hydrogen produced. However, attempts to correlate the degree of core damage by other fission product concentrations proved unfruitful because the range of uncertainty in their release fractions was too high.<sup>33</sup>

Reactor coolant conditions indicative of core melting include:

1. Extreme activities of the highly volatile fission products such as the noble gases, halogens, and cesiums;
2. Elevated concentrations of refractory elements; and
3. Indicators of massive cladding rupturing.

When core melting occurs, at least some fraction of all fission products can be released.<sup>26</sup> It is estimated that highly volatile fission products, such as the noble gases, halogens, and cesiums, will represent about 95% of the activity released from melted fuel.<sup>66</sup> The fractional release is estimated at up to 15%, although with a large degree of uncertainty. The most refractory elements are anticipated to contribute only about 1% of the

overall activity, and releases will occur mainly by the slow process of leaching.<sup>26</sup>

A complication in assessing the degree of fuel melting is that the effective surface area whereby refractory elements can be released is almost impossible to estimate.<sup>26</sup> The greater the available surface area, the higher the fraction possibly released.



## CHAPTER V

## SPECTRAL INTERFERENCES

Introduction

The interpretation of irradiated reactor fuel gamma ray spectra is quite complicated since there are many fission products represented and many decay emitting a profusion of gamma rays. In such complicated spectra, spectral interferences can be anticipated which arise because of both the Compton continuum and interfering photopeaks. Although difficult to quantify, interferences due to Compton scattering of photons in the detector are progressively more severe at lower photon energies. The Compton continuum for a photon of energy  $E$  ranges in energy from zero to  $2E^2/(E_0 + 2E)$ , where  $E_0$  is the rest energy of an electron (511 keV). The Compton continua for a multi-photon spectrum thus tend to build up to higher levels at lower energies, thereby obscuring photopeaks in that energy region to a greater extent than for higher energy photopeaks.

Photopeak spectral interferences also result when separate radionuclides emit gamma rays at about the same energy such that their corresponding photopeaks cannot be resolved without special data reduction techniques. The effect of such spectral interferences on analytical results can be threefold: (1) calculation of specific reactor coolant fission product concentrations may be inaccurate because of attributing the photopeak area to only a single radionu-

clide, (2) the identification of key photopeaks may be obscured, and (3) radionuclide identification may be erroneous due to an apparently invalid ratio of photopeak intensities. The significance of any particular interference depends on the importance of the information supplied by the radionuclide in evaluating core conditions and on the magnitude of the interference; as such it may vary with each specific reactor situation.

Spectral interferences exist even in reactor coolant spectra collected under normal operating conditions, although under normal operating temperatures most of the fission products remain within the confines of the fuel pellets. Furthermore, with the occurrence of cladding breaches, there is relatively little fission product escape into the reactor coolant, as there are only a few elements that are highly volatile and mobile. The extent of spectral interferences increases as the diversity of fission products present in the reactor coolant rises.

In an accident scenario, the core temperature rises. The initial radionuclides released at the onset of the temperature increase are those that are the most volatile and concentrated in the gap spaces of the fuel pins. It is important to accurately analyze these nuclides, since they are indicative of the onset of elevated temperatures which may lead to core damage. Further elevated core temperatures result in two major effects in the reactor coolant isotopic profile, namely, an increase in the number of different fission products present and an elevation in the gross radioactivity level.

Initially, the elevated activity results primarily from the volatile fission products experiencing a more rapid diffusion out of the fuel pellets and from some previously non-volatile elements becoming volatilized, with both groups escaping out of existing cladding breaches. However, with a sustained temperature increase, massive fuel failure can eventually result, exposing large quantities of irradiated fuel pellets directly to the reactor coolant and then releasing transuranics and refractory elements into the reactor coolant.

The magnitude of the spectral interference problem is somewhat dependent on the type of monitoring system and analysis techniques used. Since on-line monitoring allows the analysis of shorter-lived radionuclides, a greater diversity of fission products and higher gross activity will be observed resulting in increased spectral interference. Indeed, the photopeaks of highly radioactive short-lived fission products may totally obscure detection of longer-lived radionuclides, either in the Compton continuum or actual photopeak areas of the short-lived nuclides. However, it can be assumed that a successful reactor scram will occur upon the advent of adverse core conditions leading to severe fuel degradation. Therefore, before significant releases of fission products will occur, a large fraction of short-lived radionuclides will have decayed to negligible levels, making this distinction between on-line monitoring and grab sample analysis under accident conditions less important. Under routine operating conditions, the previously described distinction

between monitoring systems also applies, although the extent of spectral interferences is lower.

Since krypton and xenon fission products (both noble gases) are important contributors to spectral interferences, it has been estimated that spectral interferences can be reduced up to 75% by analyzing dissolved gases and reactor coolant liquid separately.<sup>20</sup> Specifically, in the analysis of  $^{135}\text{Xe}$  and  $^{131}\text{I}$ , an improvement in accuracy by a factor of up to 50 may occur when dissolved gas stripping techniques are used.<sup>20</sup> However, even though techniques to separate fission gases reduce interferences and subsequently improve the accuracy of analysis, it is not always feasible to use such measures since in the time required for degassing, valuable short-lived radioisotopes important for assessing core conditions may be lost.<sup>21</sup>

To aid in the assessment of reactor core conditions, only the analysis of a few key radionuclides whose presence signifies a range of conditions is required. A list of these nuclides is given in Table V-1. A review of the relationship of these key radionuclides to reactor core conditions is found in Chapter IV.

Table V-1

## KEY RADIONUCLIDES RELATED TO REACTOR CORE CONDITION

(Refer to Chapter IV)

Radionuclide $^{85m}\text{Kr}$  $^{87}\text{Kr}$  $^{88}\text{Kr}$  $^{88}\text{Rb}$  $^{131}\text{I}$  $^{132}\text{I}$  $^{133}\text{I}$  $^{133}\text{Xe}$  $^{134}\text{I}$  $^{134}\text{Cs}$  $^{135}\text{I}$  $^{135}\text{Xe}$  $^{137}\text{Cs}$  $^{138}\text{Xe}$  $^{138}\text{Cs}$ Computer Study

It is difficult to predict where significant spectral interferences may occur under extreme accident conditions, since each accident scenario will be unique. Moreover, because of decay, the

gamma ray energies where the interferences occur, and the significance of such interferences, change with time. Therefore, a computer study was undertaken to determine the energies where possible spectral interferences might be significant during the initial stages of a design basis accident, and to determine how the relative interference patterns change within the hour following shutdown. In this assessment consideration was given to fission products that would be initially released in significant fractions at the onset of rising core temperatures for a core in which most fission products are at equilibrium at various times after a reactor scram. With this set of conditions it then appeared appropriate to consider the noble gases, halogens, cesium, and rubidium radionuclides. Refractory radionuclides, which are released significantly into the reactor coolant only after the core has degraded severely, were ignored.

Because of a time lapse which occurs between the initial stages of an accident and the culmination of an accident with a core meltdown, the significant interfering radionuclides vary with the time progression of the accident. Consequently, spectral interferences were analyzed for times of 10 and 60 minutes after a reactor scram. These time intervals were selected since it is important to accurately and swiftly analyze the initial indicators of elevated temperatures so that operators will have time to take corrective actions.

Core inventories (in curies) were generated by the RIBD computer code.<sup>68</sup> Only those fission products with core inventories greater

than 10 curies at 10 minutes after reactor scram were used in the analysis (refer to Table V-2). The reference reactor used in the RIBD calculations was assumed to have a thermal power of 3565 MW, an operating period of 650 days, and a coolant volume of 87,000 gallons (about  $3.29 \times 10^7$  ml). Although no actual reactor would likely have an identical history, these inventories can approximate the relative ratios among the fission products since many fission products reach radiological equilibrium fairly rapidly. Core release fractions in the presence of severe core degradation conditions were estimated as outlined in NRC Regulatory Guide 1.4<sup>8</sup> for a design basis, non-line-break accident in a PWR, with the exception of cesium whose release fraction was assumed to be 50% rather than 1%. Thus the fission product release fractions were divided into three groups: the noble gases, the halogens and cesium, and all others with assumed release fractions of 1.00, 0.50, and 0.01, respectively.

Table V-2

## EQUILIBRIUM CORE INVENTORY

<u>Fission Product</u>	<u>Curies 10 Minutes After Scram</u>	<u>Curies 60 Minutes After Scram</u>	<u>Release Fraction</u>
Br-82	1.464 E+04	1.441 E+04	0.50
Br-83	1.151 E+07	9.400 E+06	0.50
Br-84	1.814 E+07	6.147 E+06	0.50
Br-84m	1.414 E+05	4.385 E+02	0.50
Br-85	3.597 E+06	3.452 E+01	0.50
Kr-85	8.466 E+05	8.466 E+05	1.00
Kr-85m	2.986 E+07	2.622 E+07	1.00
Br-86	2.385 E+04	4.506 E-13	0.50
Br-87	4.178 E+04	1.592 E-12	0.50
Kr-87	5.280 E+07	3.346 E+07	1.00
Kr-88	7.707 E+07	6.271 E+07	1.00
Rb-88	7.970 E+07	6.926 E+07	0.01
Kr-89	1.135 E+07	2.244 E+02	1.00
Kr-90	3.865 E+02	1.652 E-25	1.00
Rb-90	1.417 E+07	9.150 E+01	0.01
Rb-91	2.898 E+05	8.441 E-08	0.01
I-128	6.935 E+04	1.734 E+04	0.50
I-130	1.414 E+06	1.350 E+06	0.50
I-131	1.023 E+08	1.022 E+08	0.50
Xe-131m	8.485 E+05	8.483 E+05	1.00
I-132	1.443 E+08	1.441 E+08	0.50
I-133	1.775 E+08	1.747 E+08	0.50
Xe-133	2.014 E+06	2.013 E+08	1.00
Xe-133m	4.764 E+06	4.751 E+06	1.00
I-134	2.235 E+08	1.730 E+08	0.50
Cs-134	2.225 E+06	2.225 E+06	0.50
Cs-134m	6.590 E+05	5.188 E+05	0.50
I-135	1.916 E+08	1.758 E+08	0.50
Xe-135	4.226 E+07	5.097 E+07	1.00
Xe-135m	5.365 E+07	4.940 E+07	1.00
I-136	5.305 E+05	6.997 E-06	0.50
Cs-136	2.142 E+06	2.138 E+06	0.50
Cs-137	7.660 E+06	7.660 E+06	0.50
Xe-138	1.047 E+08	8.808 E+06	1.00
Cs-138	1.723 E+08	7.940 E+07	0.50
Cs-139	8.907 E+07	2.335 E+06	0.50
Xe-139	5.554 E+03	5.206 E-19	1.00
Cs-140	3.781 E+05	7.848 E-09	0.50



Gamma ray energies for the fission products listed in Table V-2 were arranged in increasing order. Gamma ray emission rates (gammas per second per milliliter of reactor coolant) were calculated at 10 and 60 minutes after reactor scram. A data file was first established listing the gamma ray energies emitted with an absolute intensity of  $\geq 0.2\%$ . The specific gamma ray emission rate in the reactor coolant is the product of the core inventory, the absolute gamma ray photofraction, the release fraction, and appropriate conversion factors, divided by the coolant volume:

$$\begin{aligned} \text{Gammas per second} = & \text{core inventory (Ci)} \times \text{photofraction \%}/100(\gamma/\text{dis}) \\ & \times \text{release fraction} \times g^* \end{aligned}$$

$$^*g = 3.7 \times 10^{10} \text{ disintegrations per second per curie}$$

Calculating the gamma ray emission rate permitted an evaluation of the expected degree of interferences due to photopeak overlap at any given energy.

An energy tolerance, the maximum energy difference between gamma ray energies that would result in spectral interference, was set at 2 keV. Fission products whose gamma ray energies differed by less than 2 keV are listed together in Table V-3 for the case of 10 minutes and 60 minutes after a reactor scram.

### Conclusions

Reference to Table V-3 indicates that most of the nuclides listed in Table V-1 are relatively clear of spectral interferences

Table V-3.

## EXTEND OF SPECTRAL INTERFERENCES FOR KEY RADIONUCLIDES

Energy (keV)	Gamma/s-ml in coolant after		Nuclide Code		Half life	Abs. Int. (%)
	10 Minutes	60 Minutes				
80.18 #	1.506E+08	1.504E+08	I-131	HFP	8.04 D	2.620
81.00	8.260E+07	8.256E+07	XE-133	FG	5.25 D	36.500
151.18 *	2.526E+07	2.218E+07	KR-85M	FG	4.48 H	75.300
196.32	2.251E+07	1.832E+07	KR-88	FG	2.84 H	26.000
197.50 +	2.321E+05	4.589E+00	KR-89	FG	3.16 M	1.820
249.79 *	4.269E+07	5.149E+07	XE-135	FG	9.11 H	89.900
258.31 *	3.706E+07	3.117E+06	XE-138	FG	14.13 M	31.500
304.87 *	4.697E+06	4.124E+06	KR-85M	FG	4.48 H	14.000
340.57	5.836E+07	5.825E+07	CS-136	HFP	13.16 D	48.500
364.48 *	4.667E+09	4.662E+09	I-131	HFP	8.04 D	81.200
401.36 +	2.553E+06	2.148E+05	XE-138	FG	14.13 M	2.170
402.58	2.937E+07	1.861E+07	KR-87	FG	76.30 M	49.500
403.03 \$	2.497E+07	2.291E+07	I-135	HFP	6.61 H	.232
433.35 #	5.261E+08	4.072E+08	I-134	HFP	52.60 M	4.190
433.74 \$	5.942E+07	5.452E+07	I-135	HFP	6.61 H	.552
434.18 +	2.474E+05	3.253E-06	I-136	HFP	.83 S	.830
434.49	2.388E+07	2.009E+06	XE-138	FG	14.13 M	20.300

Table V- 3 (continued)

Energy (keV)	Gamma/s-ml in coolant after		Nuclide Code	Half life	Abs. Int. (%)
	10 Minutes	60 Minutes			
462.79 *	2.972E+09	1.369E+09	CS-138 HFP	32.20 M	30.700
510.53 &	1.805E+08	1.776E+08	I-133 HFP	20.80 H	1.810
529.87 *	8.606E+09	8.470E+09	I-133 HFP	20.80 H	86.300
563.23 *	1.048E+07	1.048E+07	CS-134 HFP	2.06 Y	8.380
565.52	1.105E+08	8.553E+07	I-134 HFP	52.60 M	.880
568.53 +	3.600E+05	3.028E+04	XE-138 FG	14.13 M	.306
569.32	1.929E+07	1.929E+07	CS-134 HFP	2.06 Y	15.430
570.75 \$	2.637E+07	2.041E+07	I-134 HFP	52.60 M	.210
604.70	1.220E+08	1.220E+08	CS-134 HFP	2.06 Y	97.600
604.80 \$	1.834E+07	6.216E+06	BR-84 HFP	31.80 M	1.800
608.19 *	1.372E+06	1.655E+06	XE-135 FG	9.11 H	2.890
636.97 *	4.172E+08	4.168E+08	I-131 HFP	8.04 D	7.260
661.65 *	3.872E+08	3.872E+08	CS-137 HFP	30.17 Y	89.980
667.69 *	8.001E+09	7.990E+09	I-132 HFP	2.30 H	98.700
706.58	1.486E+08	1.462E+08	I-133 HFP	20.80 H	1.490
706.65 \$	1.042E+08	8.067E+07	I-134 HFP	52.60 M	.830
707.92 \$	7.104E+07	6.518E+07	I-135 HFP	6.61 H	.660
772.61 *	6.177E+09	6.169E+09	I-132 HFP	2.30 H	76.200

Table V-3 (continued)

Energy (keV)	Gamma/s-ml in coolant after		Nuclide Code	Half life	Abs. Int. (%)
	10 Minutes	60 Minutes			
795.85 *	1.068E+08	1.068E+08	CS-134 HFP	2.06 Y	85.400
816.38	6.529E+07	5.054E+07	I-134 HFP	52.60 M	.520
818.00 *	1.200E+08	1.198E+08	CS-136 HFP	13.16 D	99.700
845.43	4.331E+06	2.744E+06	KR-87 FG	76.30 M	7.300
847.03 *#	1.198E+10	9.273E+09	I-134 HFP	52.60 M	95.410
856.28	1.227E+08	1.207E+08	I-133 HFP	20.80 H	1.230
857.29 #	8.739E+08	6.764E+08	I-134 HFP	52.60 M	6.960
875.33	4.457E+08	4.387E+08	I-133 HFP	20.80 H	4.470
876.80 \$	8.755E+07	8.743E+07	I-132 HFP	2.30 H	1.080
884.09 *	8.199E+09	6.347E+09	I-134 HFP	52.60 M	65.300
896.87 +	3.764E+05	3.167E+04	XE-138 FG	14.13 M	.320
898.02	1.254E+07	1.089E+07	RB-88 FP	17.80 M	14.000
1009.78 *	2.885E+09	1.329E+09	CS-138 HFP	32.20 M	29.800
1131.51 *	2.422E+09	2.222E+09	I-135 HFP	6.61 H	22.500
1235.34 \$	2.371E+07	2.366E+07	CS-136 HFP	13.16 D	19.700
1236.41	1.486E+08	1.462E+08	I-133 HFP	20.80 H	1.490
1260.41 *	3.079E+09	2.825E+09	I-135 HFP	6.61 H	28.600

Table V-3 (continued)

Energy (keV)	Gamma/s-ml in coolant after		Nuclide Code		Half life	Abs. Int. (%)
	10 Minutes	60 Minutes				
1297.60 \$	7.215E+07	7.205E+07	I-132	HFP	2.30 H	.890
1298.22	2.323E+08	2.287E+08	I-133	HFP	20.80 H	2.330
1435.86 *	7.386E+09	3.403E+09	CS-138	HFP	32.20 M	76.300
1768.26 *	1.965E+07	1.653E+06	XE-138	FG	14.13 M	16.700
1836.01 *	1.916E+07	1.665E+07	RB-88	FP	17.80 M	21.400
2015.82 *	1.447E+07	1.217E+06	XE-138	FG	14.13 M	12.300
2195.84 *	1.143E+07	9.301E+06	KR-88	FG	2.84 H	13.200
2392.11 *	2.996E+07	2.438E+07	KR-88	FG	2.84 H	34.600
2554.80 *	5.517E+06	3.496E+06	KR-87	FG	76.30 M	9.300
2558.10 *	2.314E+06	1.466E+06	KR-87	FG	76.30 M	3.900
2677.86 *	1.755E+06	1.525E+06	RB-88	FP	17.80 M	1.960

Code:

- \* - Photopeak for key radionuclide
- + - Weak interference with key radionuclide
- \$ - Strong interference with key radionuclide
- # - Severe interference with key radionuclide
- % - Photopeak used for analysis of key radionuclide
- & - Interference from annihilation photons

from nearby photopeaks. In order to positively identify the concentration of a fission product, it is helpful to have two interference-free photopeaks to estimate the possibility that another nuclide is contributing to the photopeak of the nuclide of interest. Table V-4 indicates the extent of interference to the most prominent photopeaks of the nuclides listed in Table V-1. For all but two nuclides ( $^{133}\text{I}$  and  $^{133}\text{Xe}$ ), at least two prominent photopeaks can be found with no interfering photopeaks within 2 keV (which have gamma ray emission rate greater than 1% of that of the photopeak of interest). Thus it appears feasible to monitor nearly all of these key radionuclides under conditions of elevated core temperatures, assuming of course, that count rate reduction techniques are applied to reduce the count rate from all nuclides to an acceptable level and that spectral interference due to Compton continuum background is not severe.

Table V-4

## EXTEND OF SPECTRAL INTERFERENCES FOR KEY RADIONUCLIDES

Nuclide		E <sub>γ</sub> , keV	Interfering Nuclides						Key Photopeaks for analysis
			10 minutes after shutdown			60 minutes after shutdown			
			severe	strong	weak	severe	strong	weak	
85m	Kr	151.180 <sup>*</sup> 304.870 <sup>+</sup>							X <sup>*</sup> X <sup>+</sup>
87	Kr	402.578 <sup>+</sup> 2554.80 845.43 2558.10		135 I	138 Xe		135 I	138 Xe	X  X
88	Kr	2392.11 <sup>*</sup> 196.320 2195.842			89 Kr				X  X
88	Rb	1836.01 898.021							X X
131	I	364.980 636.973							X <sup>+</sup> X
132	I	667.69 772.61							X X

Table V-4 continued

Nuclide	E $\gamma$ , keV	Interfering Nuclides						Key Photopeaks for analysis
		10 minutes after shutdown			60 minutes after shutdown			
		severe	strong	weak	severe	strong	weak	
$^{133}\text{I}$	529.872 875.329 1298.223 <sup>++</sup> 510.530 1236.411 706.578 856.278		$^{132}\text{I}$ $^{132}\text{I}$ $^{136}\text{I}$	$^{135}\text{I}$	$^{134}\text{I}$ $^{134}\text{I}$	$^{136}\text{I}$	$^{135}\text{I}$	X (only strong peak avail- able)
$^{133}\text{Xe}$	80.997 <sup>*</sup>	$^{133}\text{I}$			$^{133}\text{I}$			none
$^{134}\text{I}$	847.025 884.090							X X
$^{134}\text{Cs}$	604.699 795.845 569.315 563.227	$^{84}\text{Br}$	$^{134}\text{I}$	$^{138}\text{Xe}$		$^{134}\text{I}$	$^{84}\text{Br}$	X X
$^{135}\text{I}$	1260.409 1131.185							X X
$^{135}\text{Xe}$	249.794 <sup>*</sup> 608.185							X <sup>*</sup> X
$^{137}\text{Cs}$	661.649							X (only $\gamma$ )
$^{138}\text{Xe}$	258.31 <sup>*</sup> 434.49 <sup>+</sup> 1768.26 2015.82	$^{134}\text{I}$	$^{135}\text{I}$	$^{136}\text{I}$	$^{134}\text{I}$	$^{135}\text{I}$		X X



Table V-4 continued

Nuclide	E $\gamma$ , keV	Interfering Nuclides						Key Photopeaks for analysis
		10 minutes after shutdown			60 minutes after shutdown			
		severe	strong	weak	severe	strong	weak	
$^{138}\text{Cs}$	1435.86							X
	462.785 <sup>+</sup>							X
	1009.78							

\* Indicates possible severe Compton continuum interference.

<sup>+</sup> Indicates possible moderate Compton continuum interference.

<sup>++</sup> Indicates possible interference from annihilation photons (511 keV).

<sup>\*\*</sup> Degree of interference for nuclides within 2 keV:

severe -  $(\gamma/s-m) \text{ for key nuclide} / (\gamma/s-m) \text{ for interfering nuclide} = 1$   
strong -  $(\gamma/s-m) \text{ for key nuclide} / (\gamma/s-m) \text{ for interfering nuclide} = 1 \text{ to } 10$   
weak -  $(\gamma/s-m) \text{ for key nuclide} / (\gamma/s-m) \text{ for interfering nuclide} = 10 \text{ to } 100$   
none -  $(\gamma/s-m) \text{ for key nuclide} / (\gamma/s-m) \text{ for interfering nuclide}$

## CHAPTER VI.

### CONCLUSIONS

#### On-Line Monitoring versus Grab Sample Analysis

There is no universal solution as to which technique (i.e., on-line monitoring or grab sample analysis) is more desirable for post-accident reactor coolant analysis, since each method has unique advantages and disadvantages. Although there are several factors to consider in comparing the merits of the two types of systems, the most important objective to consider in post-accident monitoring is the capability to promptly obtain useful information by the analysis of highly radioactive reactor coolant samples without excessive personnel exposure.

The important areas where on-line monitoring is superior to grab sample analysis include:

1. Reduction of personnel exposure: Because of radiation health considerations and the extreme radiation dose rates encountered in an accident environment, analysis of grab samples could become unfeasible, whereas on-line monitoring could still be conducted.
2. Ability to provide a prompt analysis: Prompt analytical results are critical so that operators can swiftly obtain information needed to determine the

appropriate corrective steps.

3. Ability to obtain and analyze highly representative samples: The analysis of representative samples enables greater accuracy in core diagnosis because higher quality information is provided.

The important areas where grab sample analysis is superior to on-line monitoring include:

1. Ease of adapting sample count rates to detector system capabilities: The probability that a detector will become inoperational because of high count rates is reduced with the analysis of a grab sample, since the sample activity and background are both lower.
2. Greater analytical flexibility: Since it is uncertain what specific information will be the most useful during an accident, flexibility in the kind of analysis that can be performed is important.

Each organization obviously must assess which type of system is better for its specific plant conditions.

#### The Technique of Collimation in Reducing High Count Rates

Collimation is quite effective in reducing count rates; however increases in reduction factors may be accompanied by decreases in spectral quality. Photopeak-to-total count rate ratios indicate spectral quality, and higher values provide more useful information by which to determine fission product concentrations and ultimately

evaluate core conditions. Both aperture size and gamma ray energy affect the photopeak-to-total count rate ratios, with reductions in aperture sizes resulting in lower ratio values, especially at lower gamma ray energies.

Although no additional photopeak information is really generated by collimation, at higher gamma ray energies an "apparent" enhancement in the photopeak-to-total ratio is observed. This general collimation effect results from a greater reduction in the total spectrum counts compared to photopeak counts because of greater attenuation of lower energy gamma rays coupled with a buildup of the Compton continuum.

Count rate reduction factors are also highly dependent on aperture size and gamma ray energy, with reduction factors increasing as either the aperture size or gamma ray energy decrease. Therefore, for greater accuracy, independent calibration procedures are necessary for each aperture size and gamma ray energy.

For post-accident monitoring the range in count rates obtainable by collimation alone does not satisfy the current NRC requirements of six orders of magnitude. Hence, for post-accident monitoring use, other additional count rate reduction techniques should also be implemented.

### Key Radionuclides

Although not completely definitive in the type of information that can be provided, isotopic analysis of the reactor coolant can be

useful in indicating certain core conditions. Isotopic analysis is the only means of evaluating the extent and characteristics of fuel pin failures under normal operating conditions and can be an important technique for quickly (especially if using on-line monitoring techniques) alerting operators of elevated core temperatures. Post-accident reactor coolant isotopic analysis also proved helpful in evaluating the extent of core melting that resulted from the TMI accident.

Because elevated core temperatures will be a rare occurrence (if at all), isotopic analysis will probably only be used to evaluate core conditions in a normal operating environment; however the ability to alert operators to elevated temperatures is valuable. The collection of isotopic data during normal operations is not only useful in the evaluation of fuel failures, but also supplies baseline data of steady-state and spiking conditions against which accident and fuel failure releases may be gauged.

Although not definitive because of the multiplicity of factors affecting the reactor coolant isotopic profile, estimates of the size of defect and fuel burnup of existing fuel failures under normal operating conditions can be provided. Such information is supplied by evaluating certain reactor coolant fission product ratios as well as individual concentrations. Useful ratios include those between fission products of different elements, different half lives or different chemical forms. The greatest amount of information is supplied by ratios where only one variable differs. Defect size is

indicated by analyzing short- to long-lived fission product ratios. Fuel burnup is estimated by evaluating ratios of either cesiums or transuranics.

The usefulness of reactor coolant isotopic analysis in indicating elevated core temperatures or melting is questionable since for such conditions to exist the reactor coolant system may be compromised in some manner (i.e., either by not circulating, by not reaching the entire core or by a reduced overall flow). In such cases, reactor coolant samples may not be obtainable and/or representative.

Fission products can be grouped into categories based on their volatility to estimate the order by which they would be released with rising core temperatures. However, since other factors also exert a large influence on releases, this categorization would supply only a rough estimate of release order. In addition, only the group of highest volatility would be released at the onset of elevated temperatures, which is most critical to discover.

Indications of the onset of elevated core temperatures include alterations in the ratios of longer-lived fission products to other fission products, increases in gross radioactivity levels, and increases in the range of noble gases, halogens, and alkali metals.

Use of reactor coolant isotopic analysis to indicate the extent of core melting is only useful in after-the-fact evaluations, since indicators are only released significantly after the core has already sustained considerable damage. It is estimated that with core

melting, fission products of high, medium, and low volatility will comprise 95%, up to 15%, and 1% of the total activity released, respectively. However, great uncertainty exists as to the actual fraction of the medium volatility group that would be released.

Because of the many uncertainties involved in using coolant isotopic analysis to evaluate severe core damage, a group of key radionuclides should be carefully selected. The considerations in making the selections include capability to serve as indicators of specific core conditions, feasible decay characteristics, high core inventories, and suitable chemical behavior. One frequently used group which fulfills most of the important criteria is listed in Table V-1.

### Spectral Interferences

A computer study was conducted which revealed that most of the key volatile fission products (listed in Table V-1) are relatively clear of significant spectral interferences, other than those due to Compton continuum background. Indeed, for all of them except  $^{133}\text{I}$  and  $^{133}\text{Xe}$ , at least two prominent interference free photopeaks exist. It is important that two photopeaks be available for analysis so that verification of the proper radionuclide can be made. Therefore, it appears feasible to accurately analyze reactor coolant for the indicators of initially rising core temperatures without concern of considerable overlapping photopeaks.

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