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ADAPTATION OF GAS TAGGING FOR FAILED FUEL IDENTIFICATION IN LIGHT WATER REACTORS*

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Lambert et al. (1978) and updated to the end of EBR-II operations in 1994 by Lambert et al. (1995). Briefly, the core of EBR-II contained 20-30 experimental assemblies every reactor cycle, each having its own unique tag; some assemblies contained batches of replacement elements with different tags. Thus, a total of up to 40 Xe tags were present in-core in a typical cycle. Despite this number of tags the method was used with considerable certainty to identify failures in experimental assemblies (~150 over 1974-94).

The technology was employed in the Fast Flux Test Facility (FFTF) at Hanford to identify failures in both experimental fuel assemblies and in-reactor pressurized creep capsules. Gas tagging was also exported to Japan for use in fuel assemblies for the 350 MW(e) LMR MONJU. For FFTF and MONJU application, stable isotopes of Kr (Kr^{78} , Kr^{80} , Kr^{82}) were used in addition to Xe isotopes in order to increase the number of possible separate tag compositions.

Tags for LWRs

Noble gas tags were developed for LMRs with no particular regard to economy. There were two reasons for this: the small number of unique tags required (≤ 50 for EBR-II, ~100 for FFTF/MONJU), and the few reactors involved (≤ 5). Thus, the artificially enriched isotopes of Xe and Kr (which were expensive in 1975-80) could be used at acceptable cost. In contrast, the cost of the enriched noble gases will be of primary concern in tagging commercial reactor fuel because of the larger number of tag compositions (250 if assembly tags are used in PWRs) and the potentially much larger volumes of total gas that would be needed. To illustrate the latter point, there are 72 PWRs in the U.S. alone; their annual gas tag requirements would be approximately:

$$\begin{aligned} &72 \text{ [# reactors]} \\ &\quad \times 70 \text{ [# new assemblies/reactor/year]} \\ &\quad \quad \times 250 \text{ [#rods/assembly]} \\ &\quad \quad \quad \times 1 \text{ mL [volume/rod (no wastage)]} \\ &= 1260 \text{ L of enriched tag.} \end{aligned}$$

Although the cost of isotope enrichment has dropped by an order of magnitude since 1975-80, the competitive nature of nuclear power will mandate the additional use of the cheaper Ar and Ne isotopes.

The feasibility of using Ar tags in PWRs was established by Brigger et al. (1978) in preliminary tests in the Palisades reactor. For commercial application, Ne isotopes will also likely be needed; the isotopes would be Ar^{36} , Ar^{38} and Ar^{40} , and Ne^{20} , Ne^{21} and Ne^{22} , respectively. Of these isotopes, Ar^{40} is unlikely to be useful in that it comprises 99.6% of naturally occurring Ar, which is present in air at the 1% level, air being an RCS contaminant (see later discussion).

The small number of Xe and Xe/Kr tags needed for LMR applications did not require rigorous optimization of the tag compositions: they could be simply disposed in tag-ratio space in a linear fashion without fear of confusion during analysis because of multiple failures. The ~250 tags needed for a PWR will require more careful disposition of tag-gas compositions. Gross (1979) described the method of spacing the tags which will likely have to be used: the tag compositions lie on the surface of concentric spheres in such a way that a line joining two compositions intersects no other composition. Such a configuration ensures minimal ambiguity in identifying multiple fuel failures.

In 1992-93, when utility interest in LWR tagging was rekindled, it was clear from EBR-II experience that the method held considerable promise for the on-line identification of failures. But

three questions needed positive answers before much further development could take place: (1) will gas tagging be an attractive method for failure identification in commercial reactors?; (2) will tags experience similar (or smaller) shifts in composition in a thermal neutron spectrum than the manageable shifts observed in LMRs?; and (3) will the collection and analysis of tag gas samples be as simple and precise for water reactors as they are for sodium reactors? The remainder of this paper describes the work carried out at ANL over the last three years to answer these basic questions.

REVIEW OF LWR FUEL FAILURES

The last 15 years witnessed a steady improvement in LWR fuel reliability through the virtual elimination of several of the earlier causes of failure, such as pellet-cladding interaction (PCI) and primary hydriding of cladding. Despite such improvements, however, Ozer (1994) concluded in his review of U.S. reactor performance that fuel failure persists at an averaged annual rate of 0.5 to 1.5 defective assemblies/GW(e), with a sizeable number of failures in the "cause unknown" category. Although the failure rate in boiling water reactors (BWRs) is currently about half that in PWRs, failures that do occur degrade badly from H_2 embrittlement.

Boumay (1994) described similar experience in French PWRs: of 30,000 assemblies loaded into EdF plants over 1982-94, 400 were identified as leaking. The averaged PWR assembly failure rate was thus 400/30,000 or 1.3%. For a 1 GW(e) plant with a third of the assemblies (~83) discharged per year, this rate translates to a value of $0.013 \times 83 = 1.1$ assemblies/GW(e), comparable with the ~1.5 assemblies/GW(e) for U.S. PWRs over 1989-93.

Failure Detection/Identification

Failures can be detected in both PWRs and BWRs during operation despite high background activities due to tramp fuel from previous failures. Gas activities are measured in the laboratory using grab samples from various off-gas points in the RCS, as reviewed in general terms by Kreider and Schneider (1990) and in detail for BWRs by Turnage et al. (1994). Failures often seem to become dormant; more accurately, their activities merge with the high recoil background from fuel released by previous failures. The certain way to detect a benign failure is the I^{131} activity spikes caused by steam/water/ dissolved iodine being released during power maneuvers; again, these spikes are detected by analysis of water samples. Only when failures seriously degrade, i.e., develop secondary hydride cracks that rip the cladding, are they made obvious by the presence of Np^{239} in the coolant.

The on-line identification of failures is possible only in BWRs, where local control rods can be used to modulate the fission-product release from a failure. This procedure of flux tilting has to be performed throughout the core at $\leq 70\%$ full power and involves counting many grab samples; it is time consuming and frequently only isolates the failure to a group of 4-8 assemblies. However, power in the assemblies may then be locally depressed by the adjacent control rods to limit subsequent degradation of the failure.

No direct on-line identification is possible in PWRs, and non-specific methods like I, Cs and Xe isotope "age" ratios are used to narrow down suspects to a class of assemblies, as discussed by White and Turnbull (1994). Iodine activity measurements supply input to codes like IODYNE (Clink and Freeburn, 1987) and CHIRON (Rudling et al., 1990) which estimate the number of failures; these codes consistently tend to underestimate the number

of failures when there are more than one or two. Actual identification is then performed off-line after shutdown using one or more of four NDE techniques: (1) vacuum sipping in BWRs, where gas released from an assembly when it is isolated in the spent fuel pool and sparged with gas is monitored for activity (Xe^{133} or Kr^{85}); (2) in-mast sipping in PWRs, where gas released from an assembly being raised from the grid is monitored for activity; (3) underwater visual inspection with video cameras; and (4) ultrasonic testing (U/T), where an array of probes is inserted between rows of rods to identify those containing water, and therefore failed; the probe array is inserted at several axial positions and the process repeated at 90° for identification.

Vacuum sipping is the only NDE technique applicable to BWR assemblies because they are contained in ducts. The success rate for this method is claimed to be high, see Sunderland et al. (1994). Combined with the ability to prelocate failures at power to a group of assemblies by flux tilting, the identification of BWR failures is relatively efficient. However, the power changes involved in flux tilting can seriously affect the condition of those failures which are caused by secondary hydriding.

The French place high confidence in in-mast sipping of PWR assemblies; in fact, they use the activity measurements to estimate defect size and to re-insert a failed assembly when the defect is below a critical dimension (Bourmay, 1994). The technique is new to U.S. PWRs but its use is growing. Clearly all assemblies in the core that are to continue irradiation must be examined at shutdown in order for in-mast sipping, visual or U/T inspection to identify a failure: lengthy tasks which could easily become critical-path activities. Experience with U/T inspection is mixed: in general, the technique gives over-calls of the number of failures; it does, however, indicate which rods are failed in an assembly so that they may be replaced and the assembly re-inserted.

The Case for LWR Gas Tagging

The state-of-the-art with regard to identifying LWR fuel failures can be summarized as follows. The success of flux tilting in BWRs to localize failures, coupled with the reliable (General Electric) vacuum sipping system for off-line confirmation of the failures, is offset by: (a) the need to reduce reactor power to ~65%; (b) the lengthy search process itself; and (c) the significant risk of serious degradation of secondary hydride failures (the now prevalent form of the small number of current BWR failures).

The situation is less certain for PWRs, at least in the U.S. French experience says in-mast sipping is a reliable identification method but domestic experience is limited. And yet all assemblies must still be fuel handled: those being discharged, and those remaining, the latter in order to detect release of fission gas from a failure. Visual inspection is generally limited to the first and second rows of rods. U/T is time-consuming and often gives overcalls.

It seems clear that a non-intrusive technique like gas tagging, which offers *on-line* identification of fuel failures to the assembly level, would substantially benefit operation at both types of commercial LWR. Gas tagging will have no tangible effect on reactor operations, it will account for all failures, and it will allow core management of failed fuel in a given cycle to be planned before shutdown of that cycle. Moreover, it could be easily coupled with (indeed would supplement) present off-line NDE techniques like U/T for rapid identification of failed rods. As such, it would ensure that leakers are efficiently removed during critical downtime and

that failed fuel will not be returned to core. Thus, gas tagging could substantially reduce radiation exposure to plant operators by minimizing failed fuel operation and the need for extensive poolside NDE inspection of discharged assemblies.

NEUTRONIC STABILITY OF LWR TAGS

Early experience at EBR-II showed that changes can occur in tag isotopes in an LMR environment (Lambert et al., 1976). The application of tags to LWRs thus requires an assessment of the influence of thermal neutron spectra on tags over the fuel life. Tag ratio stability requires that both tag production by fission or fission product decay and tag destruction by transmutation be negligible in-reactor. Alternatively, if neutron effects are large for some isotopes then these changes could be used as age indicators.

The stability of Ar, Ne, Kr and Xe isotopes was assessed using the ORIGEN code and additional cross sections generated by the ENDFB/V5 library (Kinsey, 1979) and the MCC-2 code (Toppel et al., 1978); fission yields were from Meek and Rider (1974). The fuel model was based on one developed by Ludwig and Renier (1989) for a Westinghouse 17×17 assembly with 4.2% enrichment. Full-power operation for three 500-day cycles was simulated, for a residence of 1500 days and an average burnout of 50 GWd/MTU.

The Ar and Ne tag isotopes have low absorption cross sections in a typical LWR spectrum and thus exhibit good stability in-reactor. Maximum burnout after 1500 days is $\leq 2.5\%$ for Ne^{21} and $\leq 1.1\%$ for Ar^{36} , and less for the other isotopes. Several of the Kr and Xe isotopes have absorption cross sections in the 1-10 barns range and are significantly affected by in-reactor exposure, Kr^{80} and Xe^{124} exhibiting the largest decreases due to burnout, 85% and 30% after 1500 days, respectively. Figure 1 shows the calculated changes in the Kr and Xe isotopes with reactor residence time.

It was concluded that all the Ne and Ar isotopes could be used as tag isotopes whose ratios with one another would not change significantly in-reactor. The changes in Kr^{78} , Xe^{126} and Xe^{129} were sufficiently small that these isotopes could also be used for LWR tags. Kr^{80} , Kr^{82} , and Xe^{124} exhibited substantial burnout and these isotopes might best be used as age indicators; for example, to differentiate between failures in different reloads of fuel.

TAG INJECTION TESTS

The viability of LWR tagging depends on whether the enriched noble gas isotopes can be collected and measured in off-gas samples from the RCS with sufficient precision to allow identification of a particular batch or assembly tag. The pioneering tag test in the Palisades PWR by Brigger et al. (1978) had shown that tag-gas isotopes were resolvable with a ca. 1975 mass spectrometer. But it was not clear what might be the true sensitivity of the method. Accordingly, tag tests were performed in 1993 in the Byron-2 and Oconee-2 PWRs; and again at Byron-2 in 1994. Byron-2 is a 4-loop Westinghouse plant operated at 3411 MW(t) by Commonwealth Edison Company in Byron, Illinois; Oconee-2 is a 2-loop B & W plant operated at 2568 MW(t) by Duke Power Company in Oconee, South Carolina.

The 1993 tests were aimed at developing methods to sample the RCS and perform mass spectrometry. Tag release was simulated by injecting a 1-gallon sample of water containing 40 mL of dissolved tag gases into the RCS through a chemical addition tank in the chemical volume control system. Gas samples were drawn from the volume control tank (VCT) and from the VCT gas space with the

FIG. 1 XE/KR TAG ISOTOPE STABILITY IN AN LWR

pressurizer steam space sample line connected to it. Samples were collected before and for 24 hours after injection. A distinctive set of γ -ray photopeaks observed when counting off-gas samples taken after injection were identified with Xe^{125} ($\tau_{1/2} = 17.1$ hr), which is not a fission product and thus derived from activation of Xe^{124} . This finding confirmed that tag had indeed been injected into the RCS (Xe^{125} activity was detected for several weeks). Mass spectrometry confirmed the presence of tag isotopes in the RCS of both reactors, but unfortunately in the Byron test, the valves on the sample cylinders leaked, while in the Oconee tests, very little tag gas actually entered the RCS because of loss during a lengthy injection process at too high an ambient temperature. No further quantitative data were therefore obtained from the tests.

Using this 1993 experience, a more carefully controlled injection was carried out at Byron-2 in November 1994. A set of new sample cylinders were supplied by Westinghouse and sampling included: (a) more extensive monitoring of the VCT for 24 hours after tag injection; and (b) monitoring the waste gas decay tank (WGDT), the eventual repository for all off-gas, at intervals of 1, 3 and 5 days after injection. The Xe^{125} activity again confirmed the presence of tag gases in the RCS. The sample times, places and Xe^{125} activities are given in Table 1. These data are being used to model gas movement in the RCS, which may be described in a later paper.

Mass Spectrometry

All the 500-mL sample cylinders were shipped to Los Alamos National Laboratory (LANL) for analysis. Prior work at ANL-West

TABLE 1 BYRON-2 TEST 2:
INJECTED AND DRAWN SAMPLES

No.	ID	Point	Volume (mL)	Relative Time (hh:mm:ss)	Xe^{125} Activity (Ci/m ³)
1	Injection	RCS	4000	00:00:00	0.000E-0
2	VB-03	VCT	500	-1:42:10	0.000E-0
3	VR-01	VCT	500	-1:22:00	0.000E-0
4	VR+01	VCT	500	1:00:00	6.825E-5
5	VR+02	VCT	500	2:00:00	3.854E-4
6	VB+02	VCT	500	2:20:11	5.323E-4
7	VR+04	VCT	500	4:00:00	1.294E-3
8	VR+12	VCT	500	12:03:00	3.166E-3
9	VR+24	VCT	500	24:08:00	4.935E-3
10	VB+24	VCT	500	24:23:00	5.104E-3
11	WR-02	WGDT	500	-2:41:30	0.000E-0
12	WR+24	WGDT	500	23:03:00	2.962E-6
13	WB+72	WGDT	500	72:23:00	1.766E-4
14	WR+72	WGDT	500	72:38:00	1.700E-4
15	WR+48	WGDT	500	113:13:00	2.912E-5

had shown that sample purification and concentration were needed before the gas could be admitted to a mass spectrometer. Figure 2 shows the apparatus that was used at LANL, consisting of a quick-disconnect sample inlet port, a 50-mL sampling cylinder, a cryostat collection container, a getter bed, a baratron, a 2-L heated inlet system, a mass spectrometer, and connections to vacuum pumps. Dry ice was available to condense water vapor in the sample cylinders but generally was not needed.

Before sample collection, the system was evacuated to high vacuum to ensure removal of residuals from prior analyses. The getter bed was regenerated by heating to 673 K for 3 hours and then held at 573 K to absorb all but the noble gas species. After 1 hour of gettering, the sample was admitted to the cryostill containing 0.5 gm of silica gel at 40 K to absorb all the nobles except He. After 5 minutes the gas phase over the cryostill was pumped away, the cryostill valve opened, and its temperature raised to 70 K to release Ne. After measuring the Ne peaks, the cryo valve was closed and temperature raised to 138 K to release about 85% of the Ar and $\leq 10\%$ of the Kr; the Ar peaks were measured. The temperature was then raised to 230 K to release the remaining Ar, $\sim 90\%$ of the Kr, and $\gg 90\%$ of the Xe. After completing analysis, the cryostill was raised to 320 K and pumped for 15 minutes to remove residuals (some water vapor, hydrocarbons, and a small amount of Xe).

The mass spectrometer was a Finnegan MAT 271 equipped with an electron multiplier and capable of analyzing a 10^{-7} mL sample within 1%. This configuration enabled analysis of smaller samples

FIG. 2 LANL APPARATUS FOR TAG GAS SEPARATION, CONCENTRATION AND ANALYSIS

with greater accuracy than had been available at ANL-West. In addition, high mass spectrometer resolution permitted a partial elimination of spectral interference at mass numbers 78 and 80, due to trace hydrocarbons or other impurities which may have derived from the sealing/cleaning compounds used on the sample cylinders.

Test Results

Table 2 illustrates the type of information obtained from the second Byron-2 test with the analyses of two samples from the VCT: a pre-test (background) sample (VB-03) taken about 2 hours before tag injection, and a post-test sample (VR+02) taken 2 hours after injection. Also included are the compositions of the injected tag gases and the natural abundances of the tag isotopes. The background (pre-test) values obtained before the injection matched closely the natural abundances of the isotopes, suggesting that the background values derived from air that was entrained in the RCS. Although this air content could not be determined directly from the analyses, estimates were made from the Ar^{41} activity measured in off-gas samples (typically $\sim 1.5 \times 10^{-2} \mu\text{Ci/mL}$ at Byron 2). With a number of assumptions, this value was found to correspond with about 73 L of Ar, 150 mL of Ne, 10 mL of Kr, and ~ 0.7 mL of Xe in the RCS.

TABLE 2 BYRON-2 TEST 2: MEASURED NOBLE GAS COMPOSITIONS

Gas	Isotope	Gas Compositions				
		Injected Tag	Natural Gas	Pre-Test ^(a) VB-03	Post-Test VR+02	% Shift ^(b)
Ar	36	49.91	0.337	0.3401	0.3473	0.014
	38	5.417	0.063	0.06466	0.06525	0.010
	40	44.67	99.600	99.5952	99.5875	0.013
Ne	20	44.86	90.48	90.749	88.808	4.4
	21	0.9408	0.27	0.26	0.296	5.5
	22	54.2	9.25	8.991	10.9	4.4
Kr	78	0.1313	0.35	0.701	0.35	61.
	80	2.378	2.25	2.578	2.539	13.
	82	61.47	11.6	12.153	33.385	43.
	83	26.56	11.5	11.357	17.97	43.
	84	9.424	57.0	56.322	36.139	43.
	86	0.0325	17.3	16.887	9.62	43.
Xe	124	10.04	0.10	ND ^(c)	8.739	87.
	126	5.013	0.09	ND ^(c)	4.364	87.
	128	18.29	1.91	1.89	15.89	85.
	129	62.35	26.4	25.13	56.97	85.
	130	1.948	4.1	3.87	2.215	86.
	131	1.877	21.2	20.29	4.575	85.
	132	0.4614	26.9	28.24	4.34	86.
	134	0.0149	10.4	11.8	1.63	86.
	136	0.0074	8.9	8.785	1.277	85.

^(a)Background in RCS

^(b)Change from background due to injection

^(c)Not detectable

It is against this high background of air in the RCS that the injected enriched noble tag isotopes had to be measured. It was gratifying that a shift was measured in the composition of all tag isotopes due to injection. Moreover, the shift was consistent between the isotopes of each gas (with the exception of Kr^{78} and Kr^{80} that were affected by hydrocarbons) and mirrored the relative abundance of the particular gas in air. That is, the measured shift was largest for Xe (85%), smaller for Kr (43%), smaller again for Ne (4.4%), and smallest for Ar (0.013%). If the calculated background Xe content is correct, however, these values imply that not all the tag gases that were originally dissolved in the 1-gallon water sample (10 mL each of Ar, Ne, Kr and Xe) found their way into the RCS. Although every precaution was taken, it is quite possible that some of the dissolved gas was lost from the water either in storage, during transport, or in the final injection.

What can be stated from the test is that for a maximum of 10 mL of each tag added to the RCS (and in practice probably less than that volume), the enriched tag isotopes were clearly detected. Indeed, with the possible exception of Ar isotopes, it seems likely that at least an order of magnitude less volume (≤ 1 mL) of each tag gas could have been distinguished from air contamination in the RCS, even with the Finnegan MAT 271 mass spectrometer. This LANL instrument is designed for rapid analysis of comparatively large samples. Although available at the time (and therefore used), hindsight suggests it was not the ideal machine for the current application. A noble gas mass spectrometer with a small inlet manifold operated in the static mode could give a 3-4 orders of magnitude improvement in resolution for a likely smaller capital

cost (Abernathy, 1995). Such improvement would come at the expense of sample throughput but this is not a large concern.

SUMMARY

A review of LWR fuel failures and methods to identify them revealed both a need and an incentive for noble gas tagging of commercial reactor fuel. The need is to eliminate as far as possible the downtime associated with identifying failures off-line by the current means of in-mast sipping, vacuum sipping or U/T inspection of assemblies. The incentive is the reduction in personnel exposure that would come from minimizing the poolside examination of fuel.

Fast reactor experience has demonstrated that noble gas tagging works well in pinpointing fuel failures on-line; the question is whether such a technique will translate to commercial water reactors. Work at Argonne over 1992-95 has indicated it very well might. Physics calculations showed that Ar and Ne isotopes do not undergo significant transmutation in a PWR neutron spectrum; three of the six candidate Kr and Xe isotopes that do change significantly in-reactor could be used as "age" indicators to tag fuel reloads.

Injection tests of enriched tag gas isotopes in the Byron-2 and Oconee-2 PWRs gave encouraging results. For example, activation of one of the tag isotopes in-reactor ($Xe^{124}n, \gamma Xe^{125}$) produced a non-fission product that acted as a "tag of a tag." In the final test in Byron-2 all tag isotopes (Ar, Ne, Kr and Xe) could be readily detected against a high background of entrained air in the RCS (which of course contains tag isotopes in their natural abundances). Moreover, the gas tags were detected using a mass spectrometer that was not optimized for measuring small quantities of noble gases. This result suggests that a high-resolution static mass spectrometer could likely provide the sensitivity required to identify assembly-specific tags in the range of 0.1-1 mL per rod. Such capability will satisfy one requirement for the commercial practicality of tagging; the other two requirements—inexpensive tags and simple ways to load them into fuel rods—will be the concern of fuel vendors.

Any future test to pin down the minimum amount of tag that can be detected in the RCS (i.e., how much must be added to fuel rods to begin with) should use a surer method of simulating tag release in-reactor, and a static mass spectrometer for analysis.

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ABSTRACT

Gas tagging for failed fuel identification involves adding a small amount of stable noble gas isotopes to the helium fill gas of fuel rods during fabrication. When a rod fails, it releases a portion of this "tag" into the reactor coolant system (RCS) along with the Xe and Kr fission gases. By interrogating a small sample of the off-gas from the RCS with a mass spectrometer, one can pinpoint the assembly (or assemblies) containing the leaker(s). Gas tagging was initially developed for fast reactors at Argonne National Laboratory (ANL) and successfully used in the Laboratory's Experimental Breeder Reactor-II (EBR-II) from the early 1970's. It eliminated the need to use sipping and flux tilting to identify failures, and saved much critical downtime over the final 20 years of reactor operation.

This paper discusses experience with noble gas tagging and its adaptation to commercial reactors. It reviews the recent incidence of fuel failures in light water reactors, and methods used to identify failures, and concludes that the on-line technique of gas tagging could significantly augment present flux tilting, sipping and ultrasonic testing of assemblies. The paper describes calculations on tag gas stability in-reactor, and tag injection tests that were carried out collaboratively with Commonwealth Edison Company in the Byron-2 pressurized water reactor (PWR) and with Duke Power Company and Babcock and Wilcox Fuel Company in the Oconee-2 PWR. The tests gave information on: (a) noble gas concentration dynamics as the tag gases were dissolved in and eventually removed from subsystems of the RCS; and (b) the suitability of candidate Ar, Ne, Kr and Xe isotopes for tagging PWR fuel. It was found that the activity of Xe^{125} (the activation product of the tag isotope Xe^{124}) acted as a "tag of a tag" and tracked gas through the reactor; measured activities are being used to model gas movement in the RCS. Several interference molecules (trace contaminants normally present at sub-ppm concentrations in RCS samples) and entrained air in the RCS were found to affect mass spectrometer sensitivity for tag isotopes. In all instances the contaminants could be differentiated from the tag isotopes by operating the mass spectrometer at high resolution (2500). Similarly, it was possible to distinguish all

the candidate tag gases against a high background of air. The test results suggested, however, that for routine analysis a high-resolution static mass spectrometer will be preferable to the dynamic instrument used for the present analyses.

BACKGROUND ON GAS TAGGING

The principle of gas tagging is deceptively simple. It consists of adding small quantities (typically 1 mL) of enriched noble gas isotopes to each fuel rod in an assembly (or group of assemblies if done by fuel batch or reactor zone) during fuel rod manufacture. By varying the tag gas compositions from one assembly, fuel batch or zone to the next one, each will contain its own unique tag. If a fuel rod fails during operation, part of its tag gas will be released to the RCS along with the fission gases that build up inside the rod. Mass spectrometric analysis of an off-gas sample from the RCS will then identify the assembly, fuel batch or zone containing the failure.

The enabling technologies for gas tagging are isotopic enrichment, which allows the compositions of the noble gases to be made significantly different from those occurring in nature, and mass spectrometry, which enables minuscule quantities of these gases to be identified. An extreme example of the sensitivity of gas mass spectrometry was the ability reported by Hurst et al. (1985) to analyze Ar^{41} in samples of the polar ice cap at concentrations of ~5000 atoms per litre.

Tags for LMRs

The gas tagging method was developed at ANL using stable isotopes of Xe that have zero or small fission yield: specifically, Xe^{124} , Xe^{126} , Xe^{128} , and Xe^{129} . The method was used very successfully at EBR-II over 1974-94 to identify failures in experimental liquid metal reactor (LMR) assemblies. Up to 1977 samples of the argon cover gas were obtained manually and the Xe cryogenically separated in the laboratory to produce mass spectrometer samples; after 1977 the process was made part of an automatic cover gas cleanup system with an ancillary on-line mass spectrometer. Pre-1977 experience with tagging was summarized by