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JUSTIFICATION FOR INERT-ATMOSPHERE
SHIELDED SAMPLING AND ANALYTICAL
FACILITIES FOR THE FFTF

November 1969

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AND ANALYTICAL FACILITIES FOR THE FFTF

By

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November 1969

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ABSTRACT

Extensive sodium sampling and analysis facilities are necessary to meet the FFTF goals of safe sustained operation, high use factor, and controlled test conditions. Shielded sampling facilities must include remotely operated equipment for obtaining sodium metal samples and distillation residues. The analytical facility must be capable of determining a wide variety of trace-level impurities in radioactive and nonradioactive sodium. The cost of remote, shielded sampling and analytical facilities is amply justified by the ultimate capability, flexibility, and safety they provide.

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INTRODUCTION

Present plans call for the use of shielded, remotely operated, sampling and analytical facilities for the FFTF. The purpose of this document is to review the need and justification for remote facilities which are required for handling radioactive sodium. The document refers only to coolant analysis and omits consideration of requirements for analysis of cover gas, aqueous process streams, and reactor materials. Fuel examination facilities will not be considered here as part of the analytical facilities.

DISCUSSION

NEED FOR ANALYSIS OF THE FFTF COOLANT

Design criteria⁽¹⁾ for the FFTF repeatedly emphasize the need for coolant purity control. This control is essential to meet three of the FFTF goals: (1) safe, sustained operations, (2) a high use factor, and (3) the performance of test irradiations under controlled conditions.

In order to achieve both safe, sustained operation and a high use factor, impurity concentrations in the coolant must be maintained below levels which will cause damage to reactor components through corrosion or mass transport. Purification of sodium for use in primary and secondary loops is required. Sodium processing systems are required to furnish sodium of specified purity and to store purified sodium. Sodium quality assurance during operation is essential. These requirements for quality control carry the implicit assumption that the levels of impurities can be verified by chemical analysis.

Design criteria⁽¹⁾ also explicitly state the requirement for analysis of sodium in the reactor, in the heat removal systems, and in the storage facilities. The necessity for providing good analytical capabilities is graphically portrayed in the experience of other liquid-metal cooled reactors. Lack of reliable, timely analytical data, even in reactors not designed to be research tools, has proved to be very costly in terms of serious impurity excursions (often detected weeks after the event), lost time, expensive correction studies, compromise of safety factors, and poor operational decisions.

Argonne National Laboratory, in the operation of EBR-II, encountered serious difficulties when flow rates in the primary plugging loop were drastically reduced by copper which had deposited from the primary coolant.⁽²⁾ Investigation revealed that badly eroded copper electrodes had jeopardized the reliability of the auxiliary primary pump and created a serious safety hazard in reactor operation. The seriousness of the situation was compounded by the unawareness that the erosion was taking place. The hazard might have been avoided and a very expensive investigation program averted had a good analytical program been functioning to reveal the presence of copper in the coolant and to give early warning of the situation.

----- During the operation of the Sodium Reactor Experiment (SRE), severe damage to fuel elements (requiring reactor shutdown, repair, and modification) resulted from blockage of coolant passages by decomposition products of tetralin (a naphthalene derivative) which had leaked into the sodium primary coolant.^(3,4) The leakage was not discovered until anomalous temperature readings and erratic reactor behavior forced shutdown and subsequent investigation. The investigation revealed large amounts of impurities in the coolant. Routine coolant analysis might have detected the increasing amount of impurities, given

warning of the impending emergency, and resulted in significant savings of time and money. Edison Company, during operation of the Enrico Fermi Atomic Plant,⁽⁵⁾ experienced an unusually high carbon dioxide content in the cover gas. Chemical analysis of both cover gas and sodium coolant immediately became necessary. Carbonaceous contaminants were found on vessel and coolant surfaces. The impurities associated with the sodium were found to be carbon, sodium carbonate, and sodium hydroxide. Analysis of the cover gas showed significant quantities of hydrogen, oxygen, methane, carbon monoxide, and nitrogen. Appraisal of the situation would have been impossible without extensive chemical analysis.

These incidents, together with experiences showing severe hydriding of Zircaloy,⁽⁶⁾ nitriding of stainless steel,⁽⁸⁻¹²⁾ and corrosion and carburization of steels⁽⁷⁾ in sodium systems containing oxygen, carbon, hydrogen, and nitrogen emphasize the importance of accurate and constant monitoring of the sodium, especially in a reactor such as the FFTF in which high operating temperatures make the system chemistry even more critical.

Carefully controlled closed-loop environmental conditions, including coolant purity, are demanded by the design criteria. As in any controlled experiment, the research conducted with FFTF closed loops will require the measurement and control of each variable. It is only when the variables can be independently controlled and voluntarily altered that meaningful scientific studies can be conducted. In the case of the FFTF test programs, this requires the capability of analytically characterizing and controlling the purity of the intensely radioactive sodium coolant in the closed loops.

CAPABILITIES REQUIRED OF THE ANALYTICAL FACILITY

A very important requirement imposed upon the analytical system will be the speed with which analytical data can be

obtained. Other requirements will be flexibility to perform a wide variety of determinations and the ability to meet exacting requirements of sensitivity and reliability.

Rapid analyses are especially important to the success of the FFTF as a research facility. For research purposes, test conditions, including coolant quality, must not only be controlled but also accurately known and recorded. Control must be exercised constantly during a test loop experiment to prevent loss of valuable data by fluctuating conditions. Undetected excursions from the programmed conditions may make the data from an experiment meaningless or misleading. For the required control, "after-the-fact" analyses are not adequate; prompt and rapid analyses are essential.

A great deal of flexibility will be required of the analytical system. Many of the impurities in the sodium coolant will affect the operation of the reactor and the conduct of the test programs. Measurement of some of these impurities will be done on a routine basis. Other analyses will be made infrequently. Impurities of interest will certainly include oxygen, carbon, hydrogen, and the transition elements. Test loop programs will generate a need for measurement of nitrogen, fission products, halides, fuel materials, boron, particulates, and chemical compounds of the various impurity elements. Information concerning ionic and molecular species of impurities is becoming increasingly significant as continuing investigations reveal correlations between corrosion phenomena and species concentration. The flexibility required to provide this information when it is demanded must be provided by the analytical laboratory.

The concentration levels of many of the impurities of interest will be exceedingly low. Analyses will be valuable, in many cases, only if their sensitivities permit accurate

numbers in the parts-per-million or even parts-per-billion range. This requirement makes it most important that the analytical system be designed for the very best in contamination control, analytical versatility, and sample preservation. Because sodium is extremely reactive, samples must be protected from contamination resulting from exposure to water, oxygen, and carbon dioxide. This protection can be accomplished only in an analytical facility furnished with an ultra-pure inert atmosphere.

CAPABILITIES REQUIRED OF THE SAMPLING FACILITY

The sampling system will be required to obtain reliable, representative samples of two types--sodium metal and distillation residue.

Sodium metal samples will be required to meet the FFTF analytical requirements. Correlation of data from the FFTF programs with those from other LMFBR programs will be meaningful only if these data are obtained from the same type of samples, namely sodium metal. This is the only type of sample that can be preserved in sample archives for analysis at a later time. Some impurities can be determined only from metal samples, particularly volatile components which would be lost during distillation sampling. Notable examples of such impurities are hydrogen, hydrogen-containing compounds, mercury, cadmium, and tin.

The mechanical operations required to obtain metallic sodium samples by the removable-section technique are relatively simple and adaptable to remote operation. Experience in manually operated facilities has shown that reliable, sustained operation of this type of sampler is possible, and it is expected that this experience can be extended to a remotely operated facility.

Distillation sampling is also a required function of the sampling system. Because this type of sampling removes the large bulk of highly radioactive sodium-24, it produces a sample which, in many cases, can be analyzed immediately without using the heavily shielded, remotely operated laboratory. Because of the high cost of operating the remote laboratory, distillation sampling can effect a considerable reduction in the cost of analytical support. For some determinations, distillation sampling will also provide sensitivity not attainable with sodium metal samples. Analysis of distillation residues is presently the best available technique for the determination of oxygen concentrations at levels below 10 ppm.⁽¹³⁾ Measurements in this range will be required during some of the more demanding test runs. Distillation sampling is also particularly well suited to the determination of nonvolatile metal impurities in very low concentrations.

The sampling system must be capable of removing representative samples of the coolant for determination of any one or many impurity components at sufficiently short time intervals to immediately detect any changes in coolant chemistry. The integrity of these samples must be carefully guarded so that a minimum change in the amount or in the chemical form of substances of interest takes place during sampling or sample handling. Since simultaneous measurement of several components may be required, the samples taken must be of appropriate form and size to permit multiple analyses, or the system must have the capability of obtaining replicate samples. Because coolant quality control will depend upon analytical measurements, these measurements must have a short delay time between sampling and reporting. It may be necessary to hold the delay time to 4 hr or less for some routine measurements.

NEED FOR SHIELDING

Sodium from the FFTF primary loop will be extremely radioactive because of the high energy gamma radiation from the short-lived, high energy sodium-24 isotope produced by neutron irradiation of the coolant. It has been calculated that a typical 10-g analytical sample will have a surface radiation level greater than 500 R/hr from sodium-24 activity alone.* Such a sample can be handled only with remote equipment and with adequate shielding.

Because of the relatively short (15-hr) half-life, a decay period of two weeks will reduce sodium-24 activity to a level at which analyses can be performed without shielding. However, in view of the urgency for continuous definition and control of operating conditions, such a delay is clearly unacceptable. Analyses must be made rapidly and without delay.

Vacuum distillation can be used to reduce radiation levels by removing from the sample all sodium which is in metallic form. Only the nonvolatile residue is then available for analysis. Despite its obvious advantage in reducing radioactivity prior to analysis, the distillation sampler must be shielded and remotely operated and it is limited in its use. It is of no value in the determination of components completely or partially volatilized during the vacuum distillation of the sodium. It is of little value in determining compounds whose concentrations change due to shifting chemical equilibria during the distillation process. Even for the determination of oxygen, distillation may fail to reduce the activity to acceptable levels because of the sodium-24 chemically combined in nonvolatile compounds such as sodium oxide. For example, distillation residue from 10 g of sodium containing 100 ppm oxygen would have a radiation level of 5000 mR/hr.

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* *This assumes an equilibrium condition of 0.04 Ci/cm³ of primary coolant.*

Neither short periods of decay time nor distillation sampling is effective in reducing the activity levels caused by some activation products and fission products. Long-lived activation products, present in the coolant because of corrosion of reactor materials, may contribute significantly to the radiation hazard of handling coolant samples.^(14,15) High fission product concentrations in the coolant will be caused by vented or ruptured fuel elements.

Prompt analyses will be especially important after a fuel element has ruptured. A rupture, whether a deliberate or an unintentional failure of fuel cladding, may release to a test loop as much as one megacurie of fission products, resulting in radiation levels (from fission products alone) as high as 5 R/hr at one foot from a 10-g sample. If this event is the climax of a test, accurate analyses are of utmost importance for the successful evaluation of the test. If, on the other hand, the failure is unplanned, emergency conditions exist. Immediate analysis of the radioactive coolant then becomes essential to safe reactor operation. In both instances of fuel element failure, coolant analysis is most urgent and only with a remotely operated, shielded facility can the analysis be obtained.

FLEXIBILITY AND RELIABILITY OF A REMOTE FACILITY

The techniques and equipment for performing chemical analysis in a remotely operated, shielded facility are well developed. Remote equipment can be designed to perform almost any operation that can be done manually. This permits determination of any substance for which a method has been developed. Specially engineered, remote manipulators of superior quality are commercially available, both for air-atmosphere and inert-atmosphere applications.

Accuracy, precision, and sensitivity have been established for the determination by manual methods of many impurities in sodium. Reliability of these methods in a remote facility and the operational reliability of the remote equipment will not be certain until they have been adapted to remote operation and repeatedly demonstrated in an inert-atmosphere facility. However, it is anticipated that reliability will prove to be very similar to that already observed with the corresponding manual methods.

LOCATION OF ANALYTICAL FACILITY

Because of the highly specialized type of laboratory required for analysis of radioactive sodium, no existing facility is suitable. The very reactive nature of sodium requires an ultra-pure inert atmosphere and the high radiation levels of primary coolant require heavy shielding. No existing facility can be satisfactorily modified to fulfill both of these requirements. Because transporting samples in shielded casks from the reactor to the analytical facility is time-consuming and costly, it is important that the analytical facility be located close enough to permit transfer of samples by pneumatic tubes. Pneumatic-tube transfer of samples will make possible the immediate analysis that will be so important, especially in emergency situations. A new facility near the reactor is essential to successful operation of the analytical laboratory.

CONCLUSIONS

- Extensive sodium sampling and analysis facilities are necessary to meet the FFTF goals of safe sustained operation, a high use factor, and controlled test conditions.
- Sampling facilities must include shielded, remotely operated equipment for obtaining sodium metal samples and vacuum distillation residues from the primary coolant and from each of the test loops.

- The analytical facility must be capable of determining a wide variety of trace impurities in radioactive and non-radioactive sodium.
- The analytical facility must have the versatility to perform a wide variety of determinations.
- The cost of remote, shielded sampling and analytical facilities is amply justified by the ultimate capability, flexibility, and safety provided.

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