MIXING AND SAMPLING TESTS FOR RADIOCHEMICAL PLANT

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Introduction

Among the more difficult sources of measurement uncertainty to assess are mixing and sampling. Because there are no standards, sampling studies and/or tests are required to assess sampling systems statistical contributions to overall measurement uncertainty. Over the years in the U.S., there have been a number of approaches to such tests. This paper describes results and test procedures used to evaluate uncertainty and bias effects introduced by the sampler systems of a radiochemical plant, and similar parameters associated with mixing. In addition, these tests are also the basis to establish operational parameters to assure minimum effects of sampling and mixing on overall measurement uncertainties.

Design of a radiochemical plant and the processing equipment is always a compromise between competing requirements and disciplines. Safeguards must compete with operational and criticality concerns. Safeguards must compromise in these arenas yet achieve acceptable measurement statistics in light of other concerns. The tests and evaluations described can guarantee acceptable measurement performance within constraints of other concerns.

This report will concentrate on experiences at the Barnwell Nuclear Fuels Plant (BNFP). In the history of reprocessing in the U.S., initial experiences were in the weapons plants at Hanford and Savannah River built in the 40's and 50's. The first commercial reprocessing plant built in the U.S., which deviated from the canyon concepts at Hanford and Savannah River, was at West Valley. This plant operated from 1966-72 and set the groundwork for advanced reprocessing concepts in the U.S. Some experiences were gained at the Idaho Chemical Processing Plant (ICPP) as they planned for upgrades through the 1970's. However, the BNFP plant incorporated the latest technologies and experiences from the U.S. reprocessing histories. It was on a par with the THORP plant currently operating in the U.K with respect to equipment design and the efforts to qualify measurement equipment and technologies. This paper will concentrate on experiences at the BNFP and discuss related experiences in the earlier generation plants as well.

Tank/Equipment Design

As mentioned, a number of concerns must be mediated in the final design of plant equipment, and thus measurement systems. When it comes to the overall vessel configuration, several variations are available to the designers. The simplest is the simple cylindrical tank, oriented with the straight- walled sections horizontal or vertical. The issue here is criticality. Concentra-

tions of fissile material must be limited in these tanks or soluble poisons must be added. Typically the tanks are used in the head end area of a light water reactor processing facility, in chemical or waste handling areas, or in uranium finishing areas where the isotopic and overall elemental concentrations ate limited and controlled. Internal structures for heating and cooling as well as mixing and sampling can complicate issues related to measuring, but the sampling and mixing issues are peculiar to the cylindrical design.

Sometimes vessel configuration becomes more complex such as in the case of evaporators and concentrators. These are special cases for the sampling and mixing questions. Generally, they pose the concerns when addressing the requirements during inventory taking to close material balances during conventional shutdown inventories or on-line inventories to support near-real-time accounting or other timely lost detection techniques.

In areas of a plant where high fissile material concentrations are expected, such as plutonium purification and conversion or highly enriched uranium processing, other tanks designs are required for criticality control. Annular or slab tanks are typically used in these applications. These designs pose a separate set of questions related to sampling and mixing. Another option in tank design under criticality constraints is a tall, small diameter tank, often referred to as "pencil tank". It again presents a unique challenge to operations related to mixing and sampling. Designers have resorted to other configurations for criticality control. One design uses runs of critically safe pipe slightly off the horizontal and connected at the ends by vertical runs of critically safe pipe. These look like a harp and are often referred to as "harp tanks". In a few cases, critically safe geometry pipe has been connected in a "D' shape to control criticality and facilitate mixing/re-circulation. All of these options present challenges to the safeguard measurement technologist.

The question of capacity also relates to design and considerations for mixing and sampling. Throughput or temporary storage requirements dictate capacity. Physical space requirements also influence design. In specific areas of processing plants. As examples, slab tanks or pencil tanks may be interconnected to form a tank system. The safeguard measurement specialist must consider these options.

Most of this discussion related to liquid processing considerations. An entirely new dimension is added in sections of processing plants where solids or slurries are handled. This paper will concentrate on liquid processing measurement problems.

In all designs, the issue is to guarantee a homogeneous solution in the process vessel to allow a representative sample to be drawn. This requires a method for mixing/agitation. In some applications mechanical means may be applied such as motor driven paddles. Various forms of pumps can be used to re-circulate solutions. Both of these methods are limited to applications where maintenance on the pumps or motors can be carried out. There are some air drive pump techniques that can be used for highly radioactive solution measurement points, but remote/no-maintenance methods are usually required. These are usually air-driven techniques to stimulate agitation/mixing, such as internal air-lift mixers, or air spargers. Spargers may take the form of submerged rings with a series of air holes drilled where the air bubbles exiting under pressure create to turbulence to simple tubes where the bubbles create turbulence and stimulate re-circulation by convection.

Sampling is another matter. Where radiation exposure and contamination concerns can be addressed by gloveboxes or ventilated hoods, samples may be drawn from simple gravity fed devices such as valves/spigots. There are sampling accuracy concerns here, but these are rather simply addressed by pulling enough samples to "flush" the valve. In radio-chemical plants the majority of samples must be remotely drawn. In the U.S., the most widely used sampling tech-

nology is the vacuum assisted airlift, re-circulating sampler. This is a rather elegant device that uses an airlift in a sampling location to deliver solutions part way to a sampling location, usually in a hot cell remote from the processing operations. The actual sampling station consists of a sampling block with two needles. The airlift pipe to the tank/solution terminates at one needle. The other needle connects to a vacuum supply and drains back to the tank/process. The airlift design is not sufficient to deliver solution all the way to the needle. A sample bottle with a rubber diaphragm cap is pushed onto the two needles. Installation of the bottle closes the system between the airlift and the vacuum and the vacuum assist pulls the solution the rest of the way up the supply line to create circulation of the solution through the bottle back to the tank/process. The safeguards questions here relate to the minimum re-circulation times to guarantee a "representative sample from the solution, and to quantify potential concentration effects from the vacuum/airlift.

In the current safeguards regime, all system must be qualified to establish statistical parameters associated with the contribution to measurement uncertainty and establish operational parameters guarantee performance and to minimize the statistical effects. The test method to establish these parameters is the subject of the remainder of this paper.

The Role of Density Measurements in Tests and Operations

If there is one message to be conveyed in this paper it is the importance of process solution density measurements. It can be argued that this is the single most important analytical measurement that can be made in radio-chemical plants. The state-of-the-art technique for solution density measurement is the vibrating tube method. This method relates the harmonics of a vibrating glass capillary tube with process solution to the tube with air and de-mineralized water. Such equipment is manufactured and distributed by Paar-Mettler as one source. The Paar-Mettler equipment is capable of density measurement precision in the fourth or fifth decimal place, depending on the model. Accuracy is dependent on calibration and accuracy related to process measurement must consider temperature effects.

The implication of temperature effects is that a totally homogeneous solution will change in density with temperature, but according to predictable relationships. The accuracy of commercially available equipment can be guaranteed by control of temperature. A controlled water bath for samples, coupled to modern vibrating tube density measurement equipment can ensure accuracy to the fourth or fifth decimal place. The implication here is that solutions measured in the controlled temperature measurement application can be compared at the fourth or fifth decimal place. Again, results at the fourth or fifth decimal place level depend on the model equipment purchased, but decisions on comparisons of solution can be made at the hundredth of a percent or better.

Mixing Tests

The objective of mixing tests to establish the effectiveness of the installed mixing systems and the operating parameters to guarantee performance of the systems during routine operations. As an example, a particular process tank is equipped with a sparge ring mixing system. How long must that mixer be operated to guarantee a "homogeneous" solution and to what degree is that solution "homogeneous".

Some qualitative tests were run over the years a several test facilities. For example, the Idaho plant built a plexi-glass slab tank to study mixing. They added acids and acid sensitive dyes to the tank. They initiated mixing and looked for color based indications of homogeneity and local-

ized dead spots for mixing. These were generally tests with slab tanks and sparge tubes and they were able to optimize placement of the tubes for mixing and elimination of dead spots. However the dye methods did not necessarily quantify mixing parameters.

The questions were answered at the Barnwell Nuclear Fuels plant using a mixing test involving density measurements. The tank in question was partially filled with an acid of one concentration. Acid of a different concentration, and consequently a significantly different density was carefully added to created a "stratified" condition in the vessel. At this point the mixing system was started and a sample drawn immediately. Subsequent samples were drawn every five minutes. All samples were analyzed for density using the vibrating tube density measurement equipment with the constant temperature water bath. Sequential density results were considered. When sequential samples agreed within the Precision/accuracy of the density measurement, it was concluded homogeneity was attained. When subsequent samples (two or three) agreed within the statistical parameters, it was assumed the homogeneity was within those limits of the density method. For the Barnwell tests, agreement to within 0.0008 was the criteria with solutions of density in the neighborhood of 1.2-1.3mg/ml. This corresponds to an "accuracy" in mixing on the order of 0.07%. This was with density measurement equipment capable of accuracy in the fourth decimal place. Modern equipment is capable of measurements to the fifth decimal place, with corresponding improvements in assessment of mixing capabilities.

These tests were run on a number of process vessels at Barnwell. Mixing time to achieve accuracy of less than 0.1% were in the neighborhood of 15-30 minutes for most single tank systems. A number of tests were run on systems consisting of several connected slab storage tanks. These tanks are mixed by re-circulating pump systems. In this test, a single tank was filled with acid of considerably different density from the others. Mixing was initiated and sampling commenced in the same way as for single tanks. Initial testes showed minimum mixing times of several hours to achieve homogeneity to the accuracy of the density method. Re-circulation pump capacities were increased and re-tests were conducted. Satisfactory mixing (within the accuracy of the density method) could then be achieved within 30-60 minutes. But the testing method using density as the parameter allowed for this judgement and corrective action.

Sampling Tests

Sampling parameters for simple sampling systems can be established based on calculations, once the homogeneity of the solution in the vessel is established. For instance, a spigot sampler draws liquid from a tank. Mixing will not generally affect the solution trapped in the spigot itself, but will guarantee the homogeneity in the vessel. The sampling procedure must simply guarantee a flush of the full capacity of the spigot to guarantee the "homogeneous" solution from the tank is delivered.

The problem becomes more complex with more complex samplers. For example, consider the re-circulating airlift sampler system. In this case the solution must enter the sampler system, flush the lines, flush the sample bottle and consider any systematic effects such as concentration of the sample by the flow of air involved.

Again, going back to the Barnwell experience in the U.S., a test was developed to quantify sampler system effects. Test equipment was fabricated to back flush the samplers on the air-lift side with water. At the conclusion of the mixing experiments, when the density of the homogeneous acid solutions in the tanks had been established, the sampler systems were back-flushed with water. Re-circulation in the sampler was by installing a bottle on the needle block. Sample bottles were exchanged on the block every 3-5 minutes. This provided a "new" sample of the

delivered solution with each bottle. All bottles were analyzed for density. Again, when subsequent bottles agreed within the accuracy of the density method, it was concluded that the sampler was delivering the homogeneous solution from the tank.

The minimum re-circulation times were established by the test. Based on the test results, it was concluded the with minimum re-circulation times, the sampler system could deliver samples within a "precision" equal to the "accuracy" of the density measurement. The question of "systematic" uncertainties remained. To answer this question, during the pre-operational tests at Barnwell, grab samples from the tanks were taken and analyzed for density at the conclusion of the sample tests. Comparison of the density of the sampler delivered samples to the grab samples was made. It was concluded in these tests that sampler system systematic errors were less than the precision of the density method (i.e., less than 0.07% with the equipment used in those tests). Tests for systematic effects in operating facilities where "grab" samples are not attainable would be problematic and rely on in-tank density measurement comparisons.

Conclusions

Mixing and sampling tests can be conducted to establish the statistical parameters for those activities related to overall measurement uncertainties. Density measurements by state-of-the-art, commercially available equipment is the key to conducting those tests. Experience in the U.S. suggests the statistical contribution of mixing and sampling can be controlled to less than 0.01% and with new equipment and new tests in operating facilities might be controllable to better accuracy.