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IMPACT OF IRRADIATION ON IITB52 ANTIFOAM EFFECTIVENESS IN LABORATORY SCALE FOAMING COLUMN STUDIES (U)

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April 12, 2001

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LIST OF ACRONYMS

KTPB	Potassium Tetraphenylborate
NaTPB	Sodium Tetraphenylborate
STTP	Small Tank Tetraphenylborate Process
IIT B52	Antifoam agent developed by Illinois Institute of Technology under contract to WSRC.
MST	Monosodium titanate
SRS	Savannah River Site

1.0 EXECUTIVE SUMMARY

IITB52 antifoam is added to the Continuous Stir Tank Reactors (CSTRs), the concentration tank and the wash tank in the Small Tank Tetraphenylborate Precipitation (STTP) to control foam. The antifoam, composed of a surfactant bis(2-ethylhexyl) sulfosuccinate, sodium salt ($C_{20}H_{37}O_7SNa$) dissolved in a diluant, will be subject to the radiation dose from the waste, primarily ^{137}Cs. During normal processing, the CSTR slurry has approximately a half-day residence time in each CSTR, a two-day residence time in the concentration tank and a two-day residence time in the wash tank. It will then have a four-day residence time in the product tank prior to hydrolysis of the precipitate.

A test program was designed to measure damage to the antifoam agent (i.e. its loss of effectiveness, if any, as an antifoam agent) caused by the irradiation during processing and storage. To simulate the radioactive dose, simulated KTPB precipitate (at an insoluble solids concentration of 9-11 wt %) was combined with IITB52 antifoam (at concentrations of 300 ppm_v and 1000 ppm_v) and irradiated in the SRTC's cobalt well to match the expected dose in the STTP. In addition, test slurries without antifoam were irradiated and the antifoam added post irradiation at comparable levels. Foam column testing was used to measure the effectiveness of the antifoam in the precipitate.

The addition of 1000 ppm_v of antifoam was more effective than the 300 ppm_v in testing. In addition, 1000 ppm_v of fresh antifoam added to irradiated precipitate without antifoam was slightly more effective than 1000 ppm_v of antifoam that had been irradiated with the precipitate. This was likely due to the degradation of the antifoam in the five days of storage post irradiation.

The antifoam was not effective in the washed precipitate as measured by the foam column testing. This suggests that the wash tank is the most likely source of foaming during normal processing. Because of the aggressiveness of this testing, it does not necessarily mean that there will be foam problems in a properly designed agitated tank. However, further testing should be completed to understand why the washed precipitate is so much foamier.

One additional finding is that the irradiation of the precipitate decreases its foaming tendency. The foamiest mixture tested was an unirradiated precipitate with 1000 ppm_v of antifoam added five days before testing. This suggests that chemical degradation of the antifoam is much more significant to the antifoam's effectiveness than the irradiation expected during normal processing.

2.0 INTRODUCTION

One of the alternatives to processing the highly radioactive salt solutions in the SRS Waste Tanks is to precipitate the radioactive cesium with sodium tetraphenylborate and then concentrate and wash the precipitate slurry. Hydrolysis will be done in a new Salt Disposition Facility prior to subsequent processing in the Defense Waste Processing Facility (DWPF). This alternative salt disposition process is called the Small Tank Tetraphenylborate Precipitation process (STTP). In the STTP precipitation process, soluble ions of cesium, potassium and ammonium are precipitated as insoluble TPB (tetraphenylborate) salts. Strontium, uranium, and plutonium are sorbed on solid monosodium titanate (MST). The resulting slurry, which now contains most of the radionuclides as insoluble solids, is filtered to concentrate the solids. After washing the solids to reduce the concentration of soluble sodium salts in the slurry, the precipitate is processed in the Salt Disposition Facility and incorporated into glass in the DWPF. The decontaminated salt solution or filtrate would be transferred to Z Area for processing and disposal as Saltstone.[1] A report detailing the precipitate preparation can be found in reference.[2]

In recent tests of the precipitation process using actual radioactive waste material excessive foaming was observed.[3] Foaming was also observed in testing at ORNL using slurry spiked with radioactive cesium.[4] Foaming during the precipitation, concentration and washing steps using simulants was also observed at SRTC.[5] As a result of these experiences with foam generation during the proposed STTP process steps, an investigation into finding suitable antifoam agents that could eliminate or mitigate the consequences of foam generation during normal operation process was undertaken.

Antifoam experts at IIT (Drs. A. D. Nikolov and D. T. Wasan) were contracted (Contract # AE-14869S) to develop and recommend an antifoam agent for the STTP process. The recommended agent is IITB52 (Batch 2 Lot # ANAEPG) which was tested in laboratory scale demonstrations of the precipitation, concentration and washing steps using simulated waste. This report describes the testing completed to determine if irradiation of the antifoam will lead to degraded performance. Since the antifoam agent is an organic compound, it may degrade in a radiation field.

3.0 EXPERIMENTAL EQUIPMENT

All experimental testing was conducted in a glass foam column fabricated by Savannah River Technology Center (SRTC) glass shop. A photograph of the foam column is shown in Figure 1. The glass foam column is 2.5 inches (6.45 cm) in internal diameter and 24 inches (60 cm) in height. A calibration curve can be found in Appendix B to correlate the column height with volume.

Attached to the bottom portion of the column is a sintered glass frit with an average pore size of 10-15 microns to retain the slurry in the column while allowing small bubbles of nitrogen gas to enter into the slurry. Foam is produced by purposely-introducing gas sub-surface at a sparge rate of 0.5 Liters/min into the slurry. The foam column is a very aggressive test of the foam generation rate in slurries. The failure of a foam column test does not necessarily translate into a failure in a well-designed, agitated tank. However, it provides a quick and reproducible way to test the relative foaminess of various mixtures and the relative effectiveness of antifoam agents.

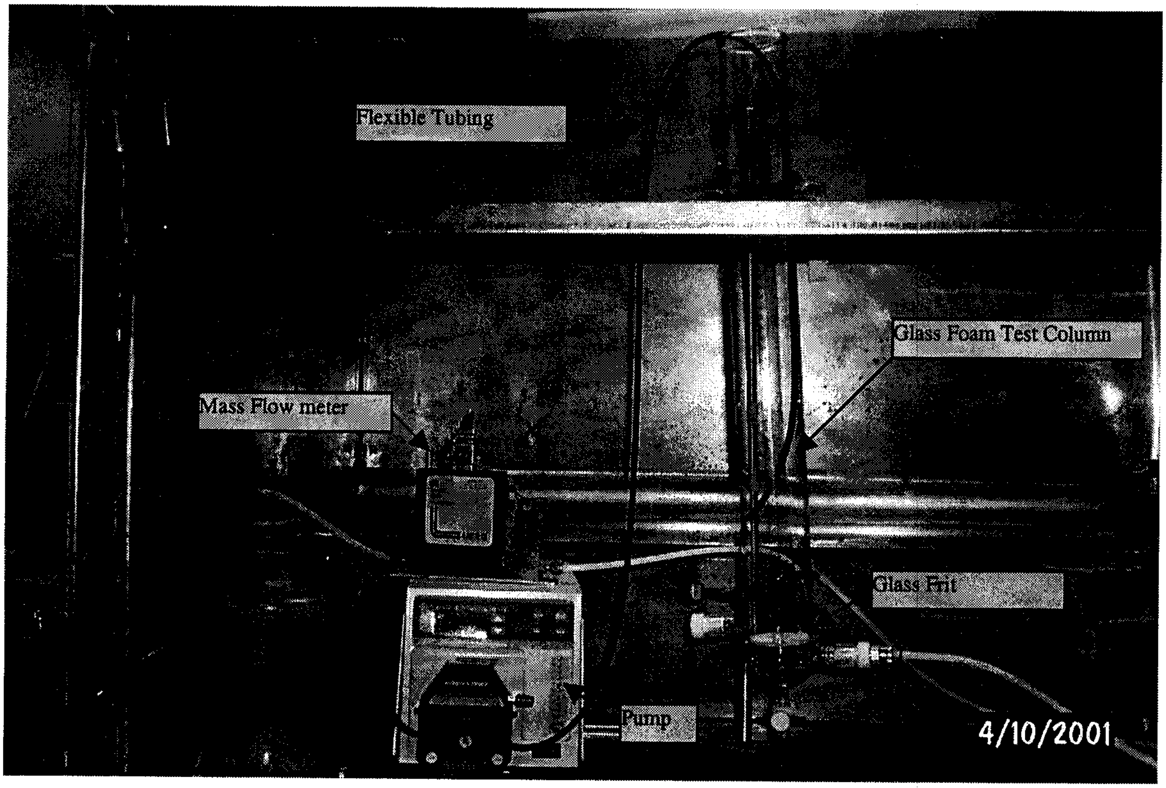

Figure 1 Antifoam Test Column Configuration

Unwashed precipitate used for these tests was produced in experiments where no antifoam was added during the preparation of the precipitate.[2] The washed precipitate was produced in experiments where IITB52 antifoam was added throughout the precipitate preparation.

The precipitate samples that underwent the irradiation process were placed into a sample chamber and lowered into a cylindrical array of Co-60 sources. The precipitate was irradiated to a total dose of $3.56X10^6$ Rad/hr. The calculation to determine total irradiation time is to take the total desired dose and divide that number by the total dose of the source to determine the total irradiation time. At the end of this time, the sample is removed from the radiation field and returned to the customer. The results of the irradiation calculation can be found in Appendix C.

The antifoam used throughout the testing was IITB52 antifoam-labeled IIT B52 9-14-00 (Batch 2 Lot # ANAEPG). The antifoam is 75 % active agent in propylene glycol and was further diluted 100:1 with deionized water.

The procedure for operating the foam column is included in Appendix A-1. The procedure for decontaminating and cleaning the foam column between runs can be found in Appendix A-2. The foam column experiments were completed by measuring the amount of precipitate to be used, adding additional antifoam agent, if needed, mixing the precipitate and slowly pumping the precipitate into the foam column using a peristaltic pump. After each experiment, the foam column was cleaned to prevent the accumulation of antifoam in the fritted disc from impacting test results.

4.0 EXPERIMENTAL RESULTS

Four different tests were conducted using both the unwashed and washed precipitate (total of eight experiments). The results and discussion in the following sections refer to these tests as follows:

Test #	Irradiated/Unirradiated	IITB52 Addition	IIT B52 Amount (ppm)
1	Irradiated	After irradiation	1000 ppm
2	Irradiated	Before irradiation	300 ppm
3	Irradiated	Before irradiation	1000 ppm
4	Unirradiated	Pre-mixed	1000 ppm

4.1 Irradiated Unwashed Samples from No-Antifoam Run

Results from the test with 10-wt % unwashed irradiated precipitate are reported in figure 2:

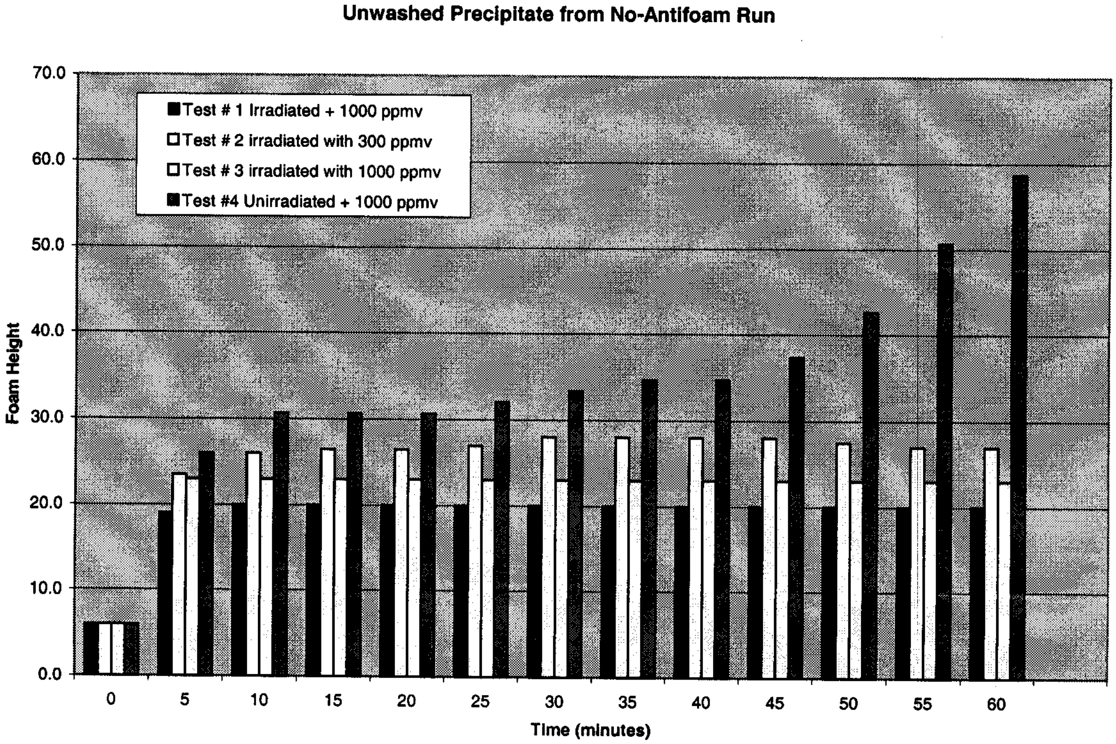

Figure 2. Results from Unwashed No-Antifoam Run

All of the experiments utilizing irradiated feeds (Tests # 1, 2 and 3) had very similar performance in the foam test column. All increased in height from approximately 6 cm to 20 to 25 cm and then held relatively constant during the 60 minute test period. The foam column tests were completed approximately five days after the solutions were prepared (prior to irradiation). Figure 2 clearly show that the addition of 1000 ppm$_v$ of fresh IITB52 antifoam before or after irradiation is the most effective amount of antifoam (minimum foam). Test #2 with 300 ppmv of antifoam consistently had the most foam of the three tests. There was little difference between irradiating the samples with antifoam (Test #3) or adding fresh antifoam to the precipitate (Test #1). Tests (1,2, & 3) clearly show that the irradiation process tends to reduce the amount of foam.

To understand whether the five-day wait prior to analysis caused excessive degradation of the antifoam, an additional test (Test #4) was run. 1000 ppm$_v$ of IITB52 antifoam was added to an unirradiated precipitate sample. After preparation, the sample was held for 5 days to mimic for the time period between preparing the samples, irradiating the samples, and testing the samples in the first three tests. Test 4 demonstrates that antifoam performance was significantly poorer in this test possibly caused by degradation of the antifoam. Results show more than twice the foam height of the other three test runs. The results of the four tests imply that irradiation altered the nature of the precipitate solution itself leading to a significantly lower tendency for foam production.

4.2 Irradiated Washed Samples from Antifoam Run

The IITB52 Antifoam Agent (at 300 & 1000 ppm) was not effective in reducing the foam volume level of the washed precipitate in any of the testing. In each of the three tests, the experiment was halted after approximately three minutes to prevent the foam level from exceeding the height of the column. The results are reported in the following figure:

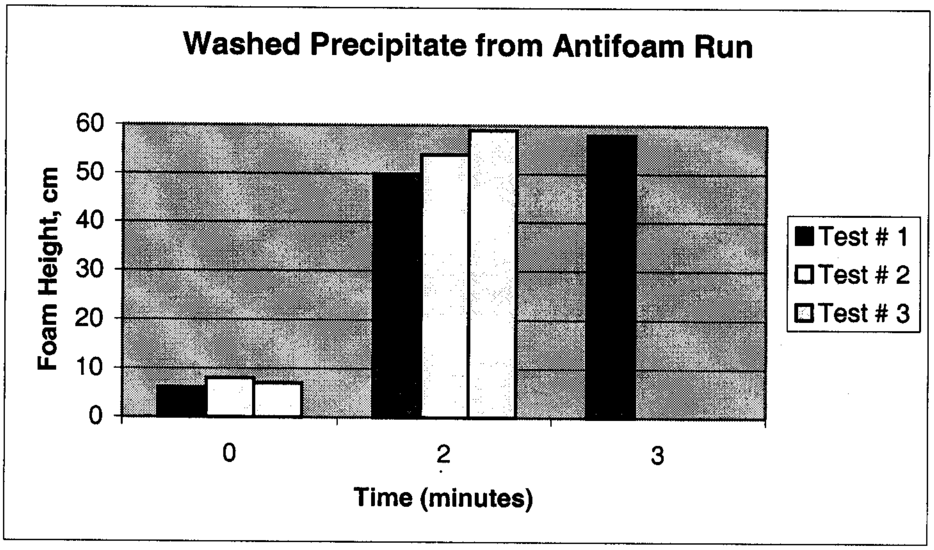

Figure 3. Washed Precipitate from Antifoam Run

5.0 CONCLUSIONS

Foam column testing of irradiated precipitate and antifoam was performed to determine whether the irradiation of the antifoam would significantly degrade its effectiveness. Testing was completed using unwashed, irradiated precipitate and washed, irradiated precipitate. This testing demonstrated that there is minimal degradation resulting from a 3.6 Mrad irradiation of the precipitate with 300 and 1000 ppm_v of antifoam present. The 3.6 Mrad dose were calculated assuming 217 hours (approximately nine days) of irradiation during normal processing, including storage prior to hydrolysis.

The addition of 1000 ppm_v of antifoam was more effective than the 300 ppm_v in testing. In addition, 1000 ppm_v of fresh antifoam added to irradiated precipitate without antifoam was slightly more effective than 1000 ppm_v of antifoam that had been irradiated with the precipitate. This was likely due to the degradation of the antifoam in the five days of storage post irradiation.

One additional interesting finding is that the irradiation of the precipitate decreases its foaming tendency. The foamiest mixture tested was an unirradiated precipitate with 1000 ppm_v of antifoam added five days before testing. This suggests that chemical degradation of the antifoam is much more significant to the antifoam's effectiveness than the irradiation expected during normal processing.

The washed, irradiated precipitate foamed severely in testing. An understanding of why this precipitate is so much foamier than unwashed precipitate is needed. Future tests on the irradiation processes should be conducted to gain a clear understanding of how the irradiation process affects the precipitate (ionic strength, soluble salt) and the IITB52 antifoam.

6.0 REFERENCES

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[2] T.B. Calloway, M.A. Baich and D.P. Lambert, "Fate of IITB52 Antifoam Agent Across the Small Tank TPB Process", WSRC-TR-2000-00261 Revision 0, 2001.

[3] R. A. Peterson and J. O. Burgess, "The Demonstration of Continuous Stirred Tank Reactor Operations with High Level waste", WSRC-TR-99-00345, Revision 1, February 15, 2000.

[4] D. D. Lee and J. L. Collins, "Continuous-Flow Stirred-Tank Reactor 20-L Demonstration Test: Final Report, ORNL/TM-1999/234.

[5] Personal Communication with Dr. Mike Poirier of WRSC, June 5, 2000.

APPENDIX A-1

TEST PROCEDURE

Equipment Configuration

Setup equipment per attached Figure A-1.
Critical specifications:

Glass Foam Column

Internal Diameter	2.5 inches
Height	24 inches

Sintered Glass Frit

Average Pore Size	10 – 16 microns
Vendor Spec	

Air Purge Rate	0.5 Liters/min

1.1.1 Test Procedure

1. Transfer 250 mL of well-agitated unwashed 10-wt % TPB slurry to a beaker.
2. Calculate the quantity of antifoam for an antifoam concentration in the 250-mL of test slurry of 1000 ppm (by weight).
3. Dilute the quantity determined in Step 2 by a dilution factor of 100:1 using water.
4. Add the diluted antifoam to the 250-mL of test slurry.
5. Mix for a minimum period of 2 minutes
6. Transfer the test slurry into the column until the slurry level is at the 200-mL mark on the column.
7. Initiate air flow at 0.5 Liters/min
8. Maintain the air sparge for a minimum of 1.

If acceptable, continue with Step 9.

9. De-inventory test column
10. Decontaminate column per procedure in Appendix A.
11. Repeat Steps 1 through 8 above using washed, 10-wt % test slurry.

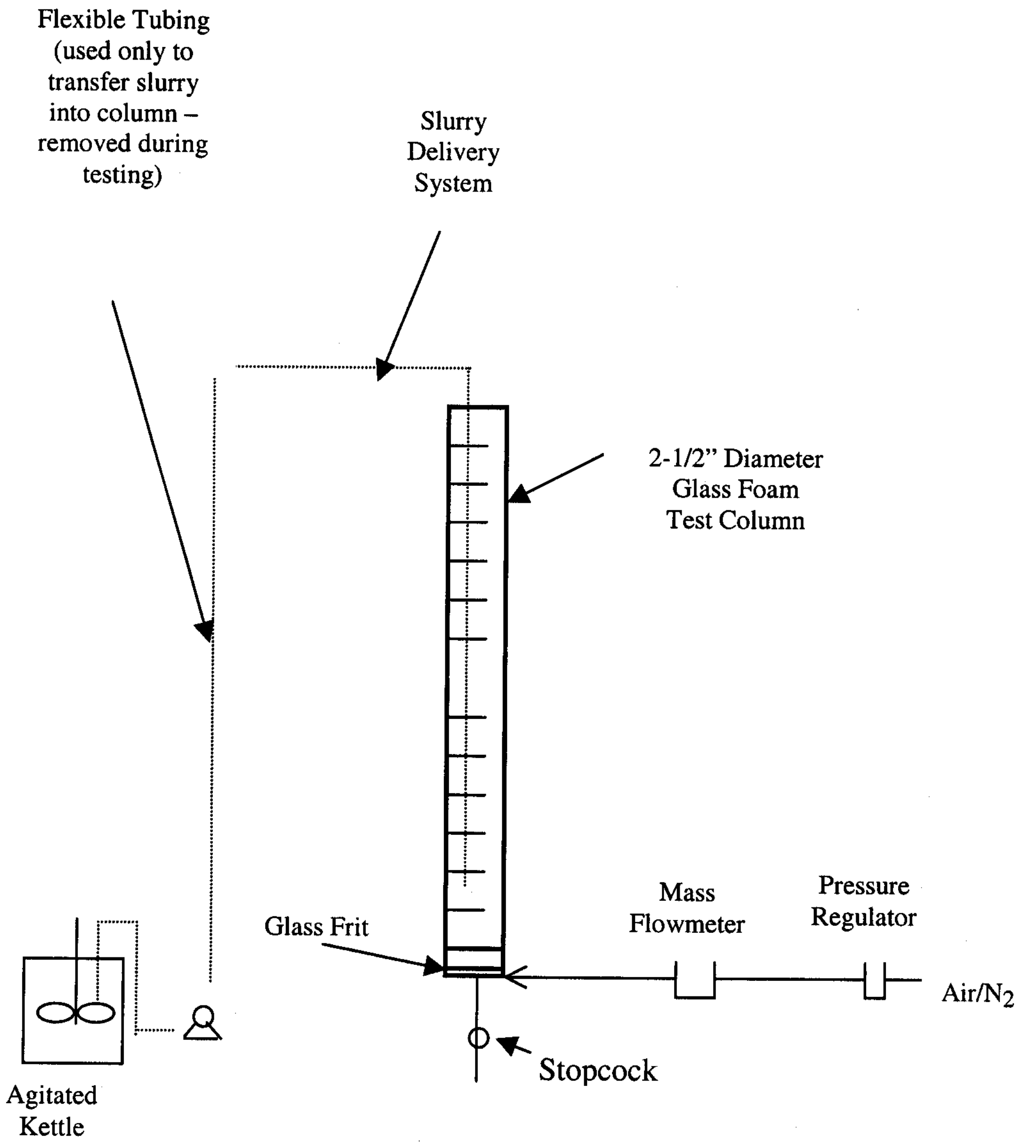


FIGURE A-1 ANTIFOAM TEST COLUMN CONFIGURATION

APPENDIX A-2

PROCEDURE FOR DECONTAMINATING THE FOAM TEST COLUMN

Safety

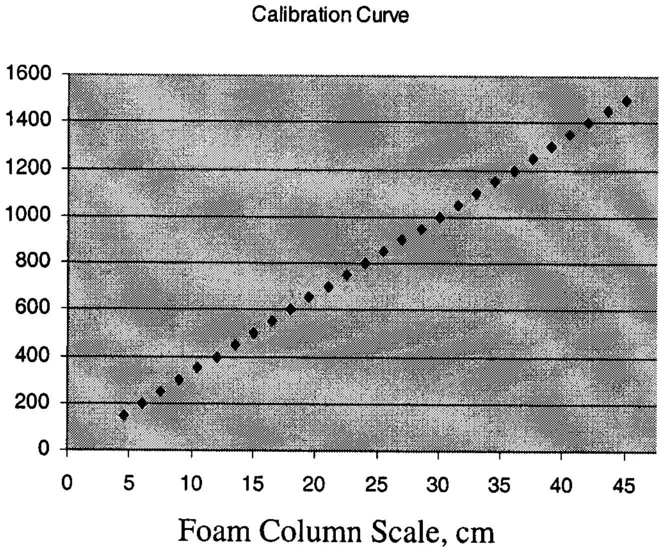
1. Wear PPE when handling cleaning solutions. This includes at the minimum safety glasses, rubber gloves, and lab coat.
2. Make certain that no oxidizing acids (e.g. HNO_3) are used in this procedure.

Cleaning Procedure

1. De-inventory the column by placing new tubing into the column and pump out the solution into a small carboy.
2. Prepare to clean the column.
3. First, rinse the column with 200-mL DI water. Then dispose the DI water into an appropriate organic rinse container.
4. Next, pour 200 mL of methanol into the portion of column above the sintered glass frit as well as 20 mL below the sintered glass frit.
5. Sparge the methanol above and below the sintered glass frit for about 10 minutes each.
6. Turn off sparge. Take the column off the racks and place a rubber stopper on top of the column.
7. Shake the column manually for several minutes. This ensures that no antifoam is left on the sides of the column.
8. Take column to the organic rinse carboy and drain the column.
9. Next, rinse the column with DI water and dispose it into the organic rinse carboy.
10. Pour 200 mL of acetone into the portion of column above the sintered glass frit as well as 20 mL below the sintered glass frit.
11. Sparge the acetone above and below the sintered glass frit for about 10 minutes each.
12. Turn off the sparge. Take the column off the racks and place a rubber stopper on top of the column.
13. Shake the column manually for several minutes. This ensures that no antifoam or methanol is left on the sides of the column.
14. Take the column to the organic rinse carboy and drain the column.
15. Pour 200 mL of DI water into the portion of column above the sintered glass frit as well as 20 mL below the sintered glass frit.
16. Sparge the DI water above and below the sintered glass frit for about 10 minutes each.
17. Turn off the sparge. Take the column off the racks and place a rubber stopper on top of the column.
18. Shake the column manually for several minutes. This ensures that no methanol or acetone is left on the sides of the column.
19. Drain DI water into organic rinse container and let the column air dry.
20. Steps 1-19 are to be conducted between every antifoam test.

Appendix B

Calibration curve of volume, ml in the test column versus the actual scale of the column.



Appendix C

Calculation of dose rate for the irradiation process.

	Ci/gallon	density (g/cc)	time (hours)
for CSTR #1	3.2	1.138	10.5
for CSTR #2	3.2	1.138	10.5
for Concentration Tank	42	1.162	48
for Wash Tank	42	1.03	48
for Feed Tank	42	1.031	100

	Dose Rate
for CSTR #1	13,423.37
for CSTR #2	13,423.37
for Conc	788,767.33
for Wash	889,852.07
for Feed	1,852,060.37
Total RADS	3,557,526.51
Total mRADS	3.56