214
2/19/69

667

IN-1257
January 1969

MASTER

**EQUIPMENT AND TECHNIQUES FOR SEALING
AMPOULES CONTAINING IRRADIATED U-235 SOLUTIONS**

IDAHO NUCLEAR CORPORATION
NATIONAL REACTOR TESTING STATION
IDAHO FALLS, IDAHO

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

U. S. ATOMIC ENERGY COMMISSION

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

Printed in the United States of America
Available from
Clearinghouse for Federal Scientific and Technical Information
National Bureau of Standards, U. S. Department of Commerce
Springfield, Virginia 22151
Price: Printed Copy \$3.00; Microfiche \$0.65

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, express or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

IN-1257
Issued: January 1969
Chemistry
TID-4500

EQUIPMENT AND TECHNIQUES FOR SEALING
AMPOULES CONTAINING IRRADIATED U-235 SOLUTIONS

D. R. Trammell, M. A. Wade, E. M. Fortsch,
H. G. Brinkley, G. A. Huff

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

IDAHO NUCLEAR CORPORATION

A JOINTLY OWNED SUBSIDIARY OF
AEROJET ALLIED
GENERAL CHEMICAL
CORPORATION CORPORATION

U. S. Atomic Energy Commission Research and Development Report
Issued Under Contract AT(10-1)-1230
Idaho Operations Office

ABSTRACT

Evaporation through poorly made seals is suggested as the cause of irradiated U-235 solutions concentrating in ampoules during storage. Recently developed improved ampoule sealing techniques and equipment are described which overcome this problem.

SUMMARY

Referee input measurement samples from dissolved reactor fuel must be stored for long periods. These highly radioactive samples are sealed in ampoules shielded by a lead jacket. A disproportionately high number of samples treated in this manner have concentrated in storage. The most likely cause of concentration is evaporation through poorly made seals. A program was inaugurated to develop improved ampoule sealing techniques and equipment. The causes of sample evaporation from ampoules, the analysis of stored dissolved irradiated fuel samples, and remote equipment developed for sealing ampoules containing highly radioactive samples are discussed.

Three possible causes of sample evaporation from the ampoules are (1) evaporation while awaiting sealing, (2) evaporation during sealing, and (3) evaporation through poorly made seals. Studies were initiated to examine each of the above causes and to supply corrective measures necessary to insure good samples for storage. The first study shows that prior to ampoule sealing, samples can be left in temporarily stoppered ampoules up to 72 hr with no concentration change. The second study shows that there is no evaporation from samples during ampoule sealing provided ampoules are not overfilled. The third study shows that of two sealing techniques investigated the drawn-shut technique gives a more reliable seal than the pinched-shut technique.

The reliability of storing samples in ampoules was demonstrated by analyzing a group of dissolved irradiated fuel samples for total uranium, total acid, and specific gravity. The values obtained were compared statistically with prestorage values. Each comparison showed no significant change.

A prototype remote ampoule sealing system has been built to overcome the radiation exposure problem encountered in sealing dissolved irradiated fuel samples in ampoules. Furthermore, the sealing system has consistently produced reliable seals and does not require the operating skill needed for the manual sealing technique. The prototype sealing system consists of (1) a sample transfer pipet, (2) an ampoule sealer, and (3) an ampoule leak tester.

The transfer pipet permits the remote transfer of solutions to ampoules from sample bottles. The transfer pipet consists of a stainless steel hypodermic needle attached to the bottom of a 10-cc glass reservoir. A long length of polyethylene tubing connects the top of the reservoir to a syringe which is used to fill and empty the reservoir. The pipet is mounted on a motor driven vertical lift to permit insertion and withdrawal from sample bottles and ampoules.

The ampoule sealer permits the remote sealing of ampoules. The sealer consists of a hydrogen-oxygen flame torch, a fuel generator, a flame igniter, a safety control, and a vertically mounted glass lathe. The ampoule is sealed by rotating its neck in the flame until soft,

and then drawing the neck shut by elevating the top chuck of the glass lathe. The reliability of this system was demonstrated when ten water samples sealed in ampoules and stored for 96 hr at 110°C showed no weight loss.

The leak tester permits the remote checking of seal reliability. The tester is an ampoule holding chamber coupled to a vacuum pump and a thermocouple vacuum gauge. The integrity of an ampoule seal is assured if the vacuum pump drops the pressure in the chamber to 17μ in 5 min and the pressure remains constant when the vacuum is turned off.

CONTENTS

	<u>Page</u>
ABSTRACT.	ii
SUMMARY	iii
I. INTRODUCTION.	1
II. CAUSES OF SAMPLE EVAPORATION FROM AMPOULES.	2
1. STUDY ON THE EVAPORATION FROM TEMPORARILY STORED SAMPLES	2
2. STUDY ON EVAPORATION DURING SEALING	2
3. STUDY ON SEAL RELIABILITY	3
III. STORED INPUT SAMPLES STABILITY STUDY.	5
IV. REMOTE AMPOULE SEALING SYSTEM	6
1. SAMPLE TRANSFER PIPET	6
2. AMPOULE SEALER.	8
3. SEAL TESTER	9
V. CONCLUSIONS	10

FIGURES

1. Prototype Remote Ampoule Sealing System	7
1-A. Transfer Pipet.	7
1-B. Ampoule Sealer.	7
1-C. Ampoule Leak Tester	7
2. Vacuum Waste System.	8

TABLES

I. Comparison of Weight Loss from Water-Filled and Empty Ampoules During Sealing	3
II. Comparison of Analyses Values of Irradiated Fuel Samples Before and After Storage.	4
III. t-Value Comparison.	5

I. INTRODUCTION

The Analytical Chemistry Branch is required to store referee input measurement samples from dissolved reactor fuel for long periods. These highly radioactive samples are manually sealed in ampoules[1] using an oxygen-propane flame torch. To afford the glass worker some radiation protection, the sealing operation is carried out with the ampoule shielded by a lead jacket with only the neck exposed. This working arrangement makes it awkward for even an experienced glass worker to produce good seals. In a recent study[2] to measure the stability of samples sealed and stored in this manner, it was found that uranium concentration of the samples had increased. The study postulates that the most likely cause of this change is evaporation during storage through poorly made seals. A program was inaugurated to develop improved ampoule sealing techniques and equipment to overcome this problem and to analyze stored samples and compare results to original data. The results of this program are reported. Section II discusses the causes of sample evaporation from ampoules, Section III discusses the application of the findings to a group of dissolved irradiated fuel samples. Section IV describes remote equipment developed for sealing ampoules containing highly radioactive samples.

[1] Wheaton Gold Band "Vacules" 12649, 5-ml capacity.

[2] D. R. Trammell, Idaho Nuclear Corporation, personal communication (December 1967).

II. CAUSES OF SAMPLE EVAPORATION FROM AMPOULES

There are three possible causes of samples evaporating from ampoules during the sealing procedure. The first is evaporation from the filled ampoule while awaiting sealing. Ampoules are filled in groups and temporarily closed with $\text{\textcircled{F}}$ plastic stoppers until sealed. The second is evaporation during sealing caused by heat from the sealing flame. The ampoule neck is first heated to softness and either pinched shut with a tong or drawn shut with a glass rod. The third is evaporation through poorly made seals.

Three studies were initiated to examine each of the above possible causes of evaporation, and to supply the corrective measures necessary to insure good samples for storage.

1. STUDY ON EVAPORATION FROM TEMPORARILY STORED SAMPLES.

Ampoules are rarely sealed immediately after filling. Instead, groups of ampoules are filled, temporarily stoppered and sealed at a later time (usually within 16 to 64 hr). This study was conducted to determine if there is any evaporation problem created by not immediately sealing the ampoules. Ten ampoules were filled with water, stoppered, weighed and stored at room temperature. After 72 hr, the ampoules were reweighed to show an average weight loss of 0.9 mg. This weight loss is negligible compared to an initial sample weight of 3000 mg. The conclusion from this study is that samples can be stored up to 72 hr at room temperature in stoppered ampoules without weight loss.

2. STUDY ON EVAPORATION DURING SEALING

This study was conducted to determine if there is significant evaporation of samples during ampoule sealing. Ten empty control ampoules were weighed, sealed and weighed again. This was done to measure weight loss caused by vaporization of glass and surface moisture during sealing. Ten water-filled[3] ampoules were weighed, sealed and weighed again. A comparison of the weight changes of the two groups of ampoules (Table I) showed no significant difference, indicating that any weight loss during sealing is attributed to loss of either glass or surface moisture on the glass and not from the sample. The conclusion from this study is that no sample is lost through evaporation during ampoule sealing provided the ampoule is not overfilled into the neck.

[3] The ampoules were filled with water to just below the neck.

TABLE I

COMPARISON OF WEIGHT LOSS FROM WATER-FILLED
AND EMPTY AMPOULES DURING SEALING

<u>Weight Loss (mg)</u>	
<u>Water Filled</u>	<u>Empty</u>
1.0	0.7
1.0	0.6
0.9	0.9
1.0	0.7
0.9	0.6
1.1	0.8
1.1	1.1
1.3	1.0
1.0	1.1
<u>1.1</u>	<u>1.3</u>
Average = 1.04	Average = 0.9

3. STUDY ON SEAL RELIABILITY

This study was conducted to determine if reliable seals can be made by pinching or drawing the ampoules shut. Seventeen ampoules were filled with water and divided into two groups. Seven ampoules were sealed by drawing the neck closed with a glass rod and 10 ampoules were sealed by pinching the neck shut with metal tongs. All of the ampoules were weighed and stored at 90 to 93°C for 6 days and reweighed. The seven ampoules drawn shut showed no weight loss, but two of the pinched-shut ampoules lost 5 and 15% respectively. Pinching the ampoules shut with metal tongs creates stress in the glass, and these seals are more susceptible to leakage. The conclusions from this study are that ampoules can be successfully sealed using the drawn-shut technique and the pinched-shut technique is not very successful.

TABLE II

COMPARISON OF ANALYSES VALUES OF IRRADIATED FUEL
SAMPLES BEFORE AND AFTER STORAGE

<u>Sample Code</u>	<u>Total Uranium (g/l)^[a]</u>		<u>Specific Gravity (@ 20°C)^[b]</u>		<u>Total Acid (N)^[c]</u>	
	<u>Initial</u>	<u>Final</u>	<u>Initial</u>	<u>Final</u>	<u>Initial</u>	<u>Final</u>
1938	8.20	8.19	1.250	1.244		
1864	8.09	8.11	1.240	1.241		
1843	7.90	7.00				
1917	7.89	7.91				
1950	0.31	0.31			5.50	5.22
4979	1.61	1.61	1.239	1.239		
4996	1.69	1.68				
5063	0.71	0.71			2.30	2.20
5050	1.56	1.58				
4973	1.49	1.51				
4988	1.58	1.57				
4991	1.56	1.54	1.234	1.233		
5017	1.71	1.71			0.98	0.92
5034	1.56	1.56				
5039	1.57	1.58	1.231	1.233		
5043	1.61	1.63			0.79	0.77
5046	1.61	1.62	1.234	1.237		
5057	1.51	1.52			0.83	0.74

[a] Relative standard deviation for total uranium is 1.46% for initial analyses and 0.66% for final analyses. Initial analyses were performed on remote-pipetted aliquots. Final analyses were performed on hand-pipetted aliquots.

[b] Relative standard deviation for specific gravity is 0.23%

[c] Standard deviation for total acid is 0.1N.

III. STORED INPUT SAMPLES STABILITY STUDY

The reliability of storing samples in ampoules was demonstrated by analyzing 18 stored samples of dissolved reactor fuel, and comparing the data with prestorage analysis values. The sealing of these samples was complicated because they were shielded in a lead jacket during the filling and sealing operation. This was done to protect the workers from excessive radiation exposure. This precaution created an awkward work arrangement, but having an experienced glass worker make the seals kept to a minimum the chances for seal failure. The integrity of the seals was checked by using a seal tester described in Section IV.

Prior to sealing, the samples were analyzed for total uranium, total acid, and specific gravity. After storing for 13 to 16 months, the samples were opened and analyzed for total uranium, and random samples were analyzed for total acid and specific gravity. The comparison of data before and after storage (Table II) shows excellent agreement.

The uranium, acid, and specific gravity values were separately compared statistically using the t-test to determine if there is a significant difference between initial and final results^[4]. To show significant difference, the calculated t-value must be greater than the table t-value.

$$t = \frac{\bar{x} - 0}{\sqrt{\frac{s_x^2}{n}}}$$

where

x = the difference between the two analyses

n = the number of samples analyzed.

The comparison of the table and calculated t-values is in Table III. In all three determinations the t-value comparison shows no significant concentration change.

TABLE III

t-Value Comparison

<u>Determination</u>	<u>n</u>	<u>Table-t</u>	<u>Calculated-t</u>
Total uranium	18	2.11	1.03
Total acid	5	2.78	2.46
Specific gravity	6	2.57	0.1

[4] Jerome Li, Introduction to Statistical Inference, Ann Arbor, Michigan: Edwards Brothers, 1957, p 97.

IV. REMOTE AMPOULE SEALING SYSTEM

Even though it has been demonstrated that dissolved reactor fuel samples can be satisfactorily sealed manually in ampoules for long-term storage, a persistent problem in preparing the samples in this manner is the radiation exposure. This problem has been overcome by building a prototype remote ampoule sealing system. This semi-automatic system can be installed behind a protective barricade equipped with manipulators, and thus enable the worker to fill, seal, and leak-check the ampoules with no exposure to his person. Furthermore, the sealing device has consistently produced reliable seals and does not require the operating skill needed for the manual sealing technique.

The prototype remote ampoule sealing system consists of (1) a sample transfer pipet (Figure 1-A), (2) an ampoule sealer (Figure 1-B), and an ampoule leak tester, (Figure 1-C). Each of these components and their operation is discussed.

1. SAMPLE TRANSFER PIPET

The sample transfer pipet permits the remote transfer of solutions to ampoules from sample bottles covered by screw caps with penetrable rubber diaphragm centers. The sample transfer pipet (Figure 1-A) consists of a 2-ft long 14-gauge stainless steel hypodermic needle attached to a 10-cc clear glass reservoir. The needle is shaped to form an inverted U, with the crest above the reservoir. This configuration provides an air lock to prevent the sample from inadvertently leaking out of the pipet. The constricted butt end of the reservoir is attached to one end of a short length of plastic tube. The other end of the tube is attached by a quick disconnect to one side of a safety overflow trap. The other side of the trap is attached to a small diameter polyethylene tube that passes out of the shielded area and is attached to a conventional hypodermic syringe in the working area. The pipet assembly, including the overflow safety trap, is mounted on a motor-driven vertical lift. The sample bottle and ampoule are held in a sliding rack directly below the pipet. The pipet is filled by positioning a sample bottle directly below the pipet tip, lowering the pipet through the rubber diaphragm into the bottle, and drawing the pipet reservoir full by the syringe plunger. The pipet is then elevated, the sample-ampoule rack repositioned so the ampoule is below the pipet tip, and the pipet introduced into the ampoule. The reservoir is emptied into the ampoule by pushing down on the syringe plunger. Care is taken during ampoule filling to allow no liquid to cling to the ampoule neck; otherwise during sealing, the liquid will volatilize and the sample concentration will change.

The pipet is cleaned by disconnecting the pipet line from the safety trap via the quick disconnect and attaching it to a vacuum wash system. The vacuum wash system (Figure 2) is a 500-cc cylindrical glass container equipped with a side arm inlet from the pipet, a top

Fig. 1 Prototype remote ampoule sealing system.

outlet to the vacuum source, a bottom outlet to the waste system, and a ball valve-float assembly. The bottom outlet contains a top-sealing ball valve to close off the drain when vacuum is applied to the wash system. When the container becomes about half full, the ball valve-float assembly closes off the vacuum causing the bottom outlet drain to open. The contents of the cylinder are thus discharged to the waste system.

Fig. 2 Vacuum waste system

With the waste system coupled to the pipet and the vacuum turned on, dilute nitric acid, demineralized water, and acetone are successively drawn through the pipet to remove any residual sample. Following the acetone rinse, the vacuum is left on to air dry the pipet. When the pipet is dried, the vacuum is turned off and the pipet line is reconnected to the sample trap in preparation for the next sample.

2. AMPOULE SEALER

The ampoule sealer (Figure 1-B) consists of a horizontally swivel-mounted hydrogen-oxygen flame torch, a Henes' water welder gas generator, a Clevite spark pump, a safety control and a vertically mounted glass lathe with remote control drive. The gas generator produces oxygen and hydrogen by the electrolysis of distilled water. The two gases are thoroughly mixed, filtered, and supplied to the torch for combustion. The Clevite spark pump ignites the gas with a spark produced by the piezoelectric effect. A thermocouple, located in the flame area is connected to an automatic safety control.

If the flame goes out, the control automatically shuts off the gas generator, thus preventing an accidental build up of an explosive mixture of oxygen and hydrogen in the sealing area. The vertically mounted glass lathe holds, rotates, and draws the ampoule shut during sealing. The top and bottom lathe chucks are coupled to a common drive system, and the top chuck can be raised and lowered.

Samples are remotely sealed in the following manner. Using a manipulator, the filled ampoule is placed into the lower chuck of the glass lathe. The top chuck is lowered onto the neck of the ampoule and the lathe is turned on. With the torch held away from the ampoule, the gas generator is turned on and the gas is ignited. The flame is brought into the sealing position, and the ampoule neck is rotated until the glass is soft enough to draw out. The top chuck of the lathe is then slowly raised until the ampoule seals and the upper part of the neck separates from the ampoule. The ampoule is rotated several additional times in the flame to fire polish the seal after which the torch is moved away from the ampoule. The sealed ampoule is then removed from the lathe, and replaced with the next ampoule to be sealed. There is no need to extinguish and reignite the flame between ampoules.

The reliability of the ampoule sealer was demonstrated by sealing 10 water samples in ampoules using the above procedure, weighing the ampoules, and storing them for 96 hr at 110°C. The ampoules were reweighed twice during the storage period (at 24 hr and again at 96 hr). These ampoules showed no weight loss.

3. SEAL TESTER

The seal tester is an ampoule holding chamber coupled to a vacuum pump and a pressure measuring device (Figure 1-C). The holding chamber consists of a 55-cc glass tube with a $\frac{1}{8}$ inch mouth, and a $\frac{1}{8}$ inch stopper with a T vent. One arm of the vent is attached to a vacuum pump, and the other arm is attached to a thermocouple vacuum gauge. The ampoule to be tested is placed into the tube and the stopper is set in place. The vacuum pump is turned on and the vacuum gauge is observed. If the ampoule is properly sealed, the pressure should drop to 17 μ in 5 min and remain constant when the vacuum is turned off. If the ampoule is improperly sealed, the pressure will not go below 140 μ in 5 min and will slowly rise after the vacuum pump is turned off.

V. CONCLUSIONS

As the findings of this report demonstrate, dissolved reactor fuel samples can successfully be stored for long time periods in ampoules provided proper care is taken to make good seals. Obtaining seals manually is complicated if the samples are radioactive enough to require shielding, but as was shown by the analyses of the stored reactor fuel samples, good seals are possible under these circumstances provided an experienced glass worker does the sealing. Many of the samples requiring storage in ampoules are too radioactive to permit even limited manual handling. The remote ampoule sealing system not only overcomes the manual handling difficulties but consistently produces easily made reliable seals.