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ORNL-4529

LEAK TESTING ENCAPSULATED RADIOACTIVE SOURCES

R. G. Niemeyer

OAK RIDGE NATIONAL LABORATORY
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JULY 1972

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LEAK TESTING ENCAPSULATED RADIOACTIVE SOURCES

R. G. Hiemeyer

ABSTRACT

The common methods used for leak testing radioactive sources are presented along with such factors as test reliability, sensitivity, and source design which affect the choice of the leak test to be used. Related data obtained from leak tests of experimental sources having known leak hole sizes are given.

1. INTRODUCTION

Encapsulated radioactive sources are routinely leak tested at the time of manufacture, and in some cases additional leak testing is performed during the lifetime of the sources. The various leak test procedures are designed to detect the presence of leak paths in the containment walls of sources through which radioactive material might escape to the surroundings.

There are three basic types of leak tests in common use: (1) detecting escaping source material (wipe test; leach tests); (2) detecting escaping air (bubble tests); and (3) detecting escaping tracer gas (helium mass spectrometer test, ^{85}Kr test). Each of these tests has several variations which affect the sensitivity of the test.

The sensitivities of the various leak tests vary from $\sim 10^{-4}$ to 10^{-10} atm cm³/sec.* The higher sensitivity methods, such as the helium mass spectrometer and ^{85}Kr leak tests, are more costly and time-consuming than the lower sensitivity methods, and it is felt that their use is not justified except in special circumstances. The leak hole sizes which correspond to leak rates in this range are more than an order of magnitude smaller than the effective pore sizes in high efficiency filters that are routinely used throughout the nuclear industry to filter gaseous effluents from radioactive operations prior to discharge to the atmosphere.

Each of the leak tests has inherent advantages and disadvantages which are considered when selecting a leak test for a given source design. For example, the vacuum leak test which has a high sensitivity ($\sim 10^{-6}$ atm cm³/sec) is not normally used on sources having < 0.25 cm³ of internal void space, since sufficient void space must be available to support a stream of bubbles indicating a leak.

The report is divided into two sections. The first section (Leak Test Methods) describes in detail each of the common leak test methods,

*Atm cm³ as used in this report means cm³ of gas stated at a pressure of 14.7 psia and a temperature of 25°C.

emphasizing the limitations in test reliability due to source design and identifying inherent characteristics of the test procedures which can cause uncertainties in the test results.

The second section (Laboratory Evaluations of Leak Tests) contains data that were obtained by using manufactured leak holes of known size and shape. These data support the leak test evaluations given in the first section. Approximate sensitivities of the various leak tests are given in Appendix A, and typical examples of leak test procedures are given in Appendix B.

2. LEAK TEST METHODS

The following discussions of the common methods for leak testing sources are designed to provide the necessary background information for selecting reliable leak tests with suitable sensitivities. Test reliability is usually limited somewhat by the amount of void volume within the source. All of the leak tests discussed require at least some void volume in order to be effective in detecting leaks. For example, the weight-gain test and immersion tests require enough void volume for water to enter the source capsule, the bubble tests require sufficient void volume to support a stream of bubbles during the test, the ^{85}Kr and helium tests require sufficient void volume for the tracer gas within the source capsule, and the smear test requires sufficient void volume for activity to migrate to the outer surface of the capsule. Other limitations which are imposed by source design, leak hole size, and inherent characteristics of the various leak tests are also discussed.

2.1 Immersion Test

The immersion test involves immersing the source in a water bath for a specified time and at a specified temperature (25-50°C). The test is sometimes performed in an ultrasonic cleaner. For water-insoluble source forms, a soluble tracer is sometimes mixed with the source material as an aid to detecting leaks. When nonradioactive tracers, such as soluble lithium or cesium salts are used, the bath water is analyzed by flame photometry¹ or some equally sensitive method. Standard counting procedures are used for soluble radioactive tracers or source material. For maximum sensitivity, the liquid volume is reduced by evaporation before analysis.

Several liquid baths, each containing a source, can be operated simultaneously by a single operator, since observation of the sources for leak indications during testing is normally not necessary. This is one of the few methods that can be used to test for large leaks in sources having very small free void volumes.

For this test to be reliable on doubly encapsulated sources, the liquid must flow from the bath through the leak hole in the outer source capsule and continue flowing into the void space between the inner and outer capsule until it reaches the leak hole in the inner capsule. It then must pass through the inner leak hole and continue flowing until it reaches and dissolves detectable amounts of the source material which must then migrate

back through the liquid and enter the liquid bath. It is uncertain whether these events can be relied upon to occur. For some sources the leak path might be excessively long, requiring a correspondingly long immersion time. Evidence found in the ORNL leach tests of CsCl sources indicated that solid pieces of the source material sometimes plugged the leak holes. In other cases, the leak holes were plugged with what appeared to be corrosion products, which probably resulted from the water solution of cesium chloride attacking the stainless steel leak hole surfaces (see Sect. 3.4.1).

Since the immersion time required for this test is unpredictable, the time normally allotted is usually quite long (1 to 24 hr). In the case of the IAEA leak test for "special form material"² and the British test for sealed sources,³ the source capsule is immersed in the solution for 8 hr at $50 \pm 5^\circ\text{C}$, stored in air for 7 days, and immersed in a fresh solution for 8 hr at $50 \pm 5^\circ\text{C}$. Each solution is analyzed for activity.

This test will not detect leaks in the outer capsule only, since no activity will be found in the leach water unless both inner and outer capsules leak. A very clean work area is required to prevent background contamination from interfering. The exposed surfaces of the source capsule must be very clean so that the test will not indicate a leak (due to capsule contamination) when none is present. Control samples are necessary to determine the effects of either background or cross contamination, as was done in the IAEA leak test of CsCl test sources (see Sect. 3.6).

Testing at elevated temperatures is advantageous, since source materials are usually more soluble and liquid viscosities are lower. A suitable liquid might be difficult to obtain, since the liquid must dissolve the source material or the tracer but not attack the source capsule. This test does not locate leak holes as the bubble tests do (see Sect. 2.2).

The maximum sensitivities for this test when very soluble lithium, cesium, or radioactive tracers are used are 0.0002 $\mu\text{g/ml}$, 0.003 $\mu\text{g/ml}$, and ~ 100 dis/min.ml, respectively.

2.2 Bubble Leak Tests

The vacuum leak test, the hot-water bubble test, and a part of the vacuum leach test fall into a general class of tests commonly referred to as bubble leak tests. The source capsule is immersed in a liquid to a depth of about 2 in., and a pressure differential is established between the inside of the capsule and the surrounding such that gas can pass from the capsule interior through a leak hole in the capsule and be detected by observing the resulting stream of bubbles emanating from the point of leakage. The pressure differential is obtained by prepressurization of the capsule, by reducing the pressure above the liquid, or by placing the capsule into a liquid which is at a higher temperature. There must be sufficient free void volume within the source capsule to support a stream of bubbles for this type of test to be valid. Bubble leak tests have been used successfully on source capsules having free volumes as small as 0.1 cm^3 ;⁴ however, this is considered to be the minimum volume at which it should be used. At ORNL, an arbitrary minimum void volume of 0.25 cm^3 is used for bubble leak testing.

When prepressurization is used, the pressure and time must be selected so that sufficient gas will leak into the source to give the desired sensitivity. Additional time must be allowed under pressure to account for loss of internal capsule pressure during the time interval between pressurization and testing. If helium is used for prepressurization, the full advantages of helium will not be obtained unless the air is first removed from the capsule by evacuation. The use of flammable gases or liquids in hot cells is not normally advisable due to fire and explosion hazards. When the pressure differential is obtained by a vacuum over the liquid, some of the liquid can be drawn inside the capsule through a leak hole when the vacuum is released at the end of the test. Unless this liquid can be removed, serious difficulties could be encountered if attempts were made to repair the leak. If the capsule is sealed with residual liquid inside, excessive pressures may be generated later inside the capsule by radiolysis of the liquid, or by abnormally high environmental temperatures which will cause liquid expansion or vaporization.

To some extent, both reliability and sensitivity of these tests depend on the operator performing the test. Careful observation is required throughout a test to be sure that leaks will not be missed. Small bubbles or a low bubbling rate can make very small leaks difficult to see, and very large leaks in which the gas escapes in one or two bubbles might be missed. Corrosion or air pockets on the external surfaces of the capsule can give false indications of a leak. Good lighting is important and moderate magnification (2 to 4X) is helpful. The area where leaks are most likely to occur (seal areas) should be clearly visible to the operator. The liquid used should be outgassed so that extraneous bubbling will not interfere with the test. When the pressure differential is obtained by means of vacuum, the vacuum should be applied slowly, and the operator should be observing for leaks from the moment the vacuum is applied to avoid missing large leaks.

When one of the bubble leak tests is used, a stream of bubbles emanating from a source capsule indicates that the outer source capsule is leaking. The inner source capsule may or may not be leaking. In order to be reasonably certain that the inner capsule does not leak, it must be leak tested before it is loaded into the outer source capsule, or the assembled source must be leak tested using a method which will detect leaks in both inner and outer source capsules. The bubble leak tests will indicate both the presence and the location of all leaks in the outer source capsule provided the free void volume is large enough and the pressure drop across each leak is sufficient to cause bubbling from each leak.

For bubble leak tests where the leak flow is into a liquid under vacuum, such as the vacuum leak test and vacuum leach test, the pressure differential (ΔP) necessary to initiate bubbling is approximately $4T/D$, where T is the surface tension of the liquid and D is the diameter of the leak hole. The development of this equation and experimental data demonstrating its validity are given in the section on evaluation of the vacuum leak tests (Sect. 3.10). For leak tests, such as the hot-water bubble test, where the flow is into a liquid at atmospheric pressure, the relationship $\Delta P = 4T/D$ is invalid.

2.3 Vacuum Leak Test

The source is immersed in isopropyl alcohol or ethylene glycol to a depth of about 2 in. below the surface. The pressure above the liquid is then slowly reduced to 2.5 psia. A leak is indicated by a stream of bubbles rising through the liquid from a point on the source capsule (Fig. 2.1).

Fig. 2.1. A Stream of Bubbles Rising Through the Liquid from a Leaking Source Capsule During the Vacuum Leak Test.

The sensitivity of this test⁴ has been determined by preparing a number of calibrated leaks, measuring the hole sizes microscopically, and measuring the leak rates. Under the test conditions described, leaks as small as $\sim 4 \times 10^{-6}$ atm cm³/sec (for 1- μ m diam) can be readily detected, and smaller leaks have been detected by pressurizing the capsules with air immediately before the leak test. The sensitivity of the vacuum leak test can be brought almost to the level of the helium and ⁸⁵Kr leak-detection methods (10^{-8} to 10^{-10} atm cm³/sec) by the use of prepressurization techniques (see Sect. 3.10).

The discussion of bubble testing should be taken into consideration when the vacuum leak test is used.

2.4 Vacuum Leach Test

The vacuum leach test^{5,6} can be regarded as a combination of the immersion test and the vacuum leak test. The discussions of these two tests should be considered when the vacuum leach test is used.

The source is immersed in liquid (usually water), and the pressure above the liquid is decreased to 2.5 psia for 3 min and then vented to atmosphere. The vacuum-venting procedure is repeated three times. The liquid is then analyzed in the same manner as described for the immersion test. This test, like the immersion test, can detect a leak path which extends through both the inner and outer capsules. During the initial vacuum period, air is drawn through a leak hole from the free void volumes within the outer and inner capsules as in the vacuum leak test. During the initial venting period, the test liquid is drawn into the partially evacuated free void volumes. During the remaining vacuum-venting cycles, additional air can be removed from the source and the test liquid can move in, dissolve source material, and move out to the liquid bath again. This test will detect a leak in the outer capsule of a source even though the inner capsule is not leaking

since a leak in the outer capsule is indicated by a stream of air bubbles emanating from the capsule during the vacuum periods, provided there is sufficient free void volume to support a stream of bubbles.

The vacuum leach test is more reliable than the immersion test, since the repeated vacuum cycles provide a ΔP of 12.3 psi to force liquid in and out of a leaking source. The test also provides the high sensitivity of the vacuum leak test to detect leaks in the outer capsule when sufficient free void volume is present.

The vacuum leach test can be used on sources having free void volumes which are too small to support a stream of bubbles and on sources having very large diameter leaks. The time required to perform the test is short, ~24 min. Several sources can be tested at one time by a single operator unless observations for bubbles are required. The liquid used must be one that will dissolve the source material without attacking the source capsule. When desirable, soluble radioactive or nonradioactive tracers are used as in the immersion test. The maximum sensitivity of this test is the same as the vacuum leak test when a stream of bubbles is evolved. When bubbles are not evolved, the sensitivity is limited by the detectable quantity of tracer as in the immersion test (see Sect. 2.1).

2.5 Hot-Water Bubble Test

The source, which is at ambient temperature, is immersed in water that is just below the boiling point (~90°C). A leak is indicated by a stream of bubbles emanating from a point on the source capsule due to the increase of internal gas pressure caused by the increase in temperature (Fig. 2.2). The source must have sufficient free void volume to support a stream of bubbles. Several variations of this test exist, including prepressurization of the source with air or helium, using a liquid with a higher boiling point, or precooling the source just before the test. Each of these variations is designed to provide increased gas pressure inside the source capsules in order to increase the sensitivity of the test.

It is possible that a large leak might be missed if all the air escapes in one or two bubbles. Prepressurization before the test can be effective for small leaks where the leak rate is low enough so that the pressure will not bleed off before the leak test starts. This can be further improved by conducting both the prepressurization and

Fig. 2.2. Hot Water Bubble Test of Experimental Source Capsule.

the leak test in the same chamber to reduce the time in which bleed-off can occur. Precooling the capsule can be effective, since it can cause more gas to flow into the capsule, thus giving a higher internal pressure when heated in the test. For doubly encapsulated sources, this test will indicate leaks in the outer source capsule only. The use of liquids having high viscosities is not recommended, since these liquids sometimes plug the leak holes and prevent leaks from being detected. The sensitivity of this test is $\sim 2.8 \times 10^{-4}$ atm cm³/sec (8.5- μ m-diam leak). The discussion of bubble testing should be taken into consideration in using the hot-water bubble test.

2.6 Weight-Gain Test

The source is weighed and placed in a water-filled pressure vessel (fig. 2.3) where the water pressure is increased to the desired value and held for the desired length of time. The source is then air dried and weighed; a gain in weight indicates that water has entered the capsule through a leak. There are many variations of this test, particularly in the pressure used and the length of the test. A typical set of conditions is a water pressure of 300 psig for a 1-hr period.

Fig. 2.3. Apparatus Used for Testing Source Capsules by the Weight-Gain Method.

The weight gain test is unreliable when there is extraneous material on the source capsule that can be dislodged by the water. The internal void volume should be large enough to allow water to enter the capsule through a leak under the test conditions. The weight of the water entering a leak should be at least five times the sensitivity of the weighing equipment. The time allotted for the test must be sufficient to reach this weight of water at the test pressure. Clean distilled water should be used so that solid particles will not plug the leak hole. In the case of very large diameter leak holes in sources having small internal void volumes, the water might be lost in the drying step, and the leak will not be found. The water pressure should not be high enough to deform the source capsule. For small leaks an evacuation step immediately preceding the pressurization can remove air from the void space and provide additional space for water to enter the source through a leak. The sensitivity of this test is estimated to be in the range of 10^{-5} to 10^{-7} atm cm³/sec (1.7- to 0.17- μ m diam).

2.7 Helium Mass Spectrometer and ⁸⁵Kr Leak Tests

The tracer gas (helium or ⁸⁵Kr) is incorporated into the source capsule either before sealing by using a closed chamber welder or after sealing by pressurization in an atmosphere containing the tracer gas. The closed chamber welder has provisions for evacuation, gas introduction, and temperature and pressure measurement. Any gas (with a known tracer content) consistent with good welding practice can be used. With this equipment, the air can be evacuated and the welder chamber filled with the weld gas. The void space of the outer capsule will then contain gas with a known tracer content. If desired, the inner capsule seal can be performed in the same manner.

When the tracer gas is introduced after sealing, the tracer, under pressure, can flow through a leak hole into the free void volume of the outer capsule. In order for this test to be valid, there must be an internal void volume sufficient to contain enough gas to last through any evacuations required by the test procedure (helium test only). It is possible in the case of large leaks that all of the tracer gas will be lost during the evacuation period of the helium test. During pressurization, the gas pressure must be high enough and the soak time long enough so that detectable amounts of tracer will flow through any leaks into the internal void volume.

When the tracer gas is helium, the leak test is made with the helium mass spectrometer.⁷ The usual method employed is the dynamic method in which the source is placed in a chamber that can be valved directly to the sample port of the leak detector. The chamber is then evacuated and monitored for helium. A second method involves "sniffing" the outer surfaces of the source with the leak detector sniffing attachment.

When the tracer gas is ⁸⁵Kr,⁸ the gas escaping through a leak is collected in a chamber at atmospheric pressure and analyzed for ⁸⁵Kr by conventional counting techniques. The leak rate is then calculated from the count rate.

After completion of the helium leak test, it is necessary to perform an additional leak test which is sensitive to leaks of 10^{-5} atm cm³/sec and larger. Leaks in this range can be missed due to the evacuation procedures.

In using the helium and ⁸⁵Kr leak tests, it should be remembered that both of these gases are readily absorbed by many organics, oxide films, and dust particles. Any contamination of the capsule or the test equipment will cause background problems with the smaller leaks. The diffusion rates of helium and ⁸⁵Kr through rubber, glass, plastics, and other porous materials used in these tests should be checked to be sure they do not limit the sensitivity of the test. The test areas should be adequately ventilated so that extraneous tracer gas does not interfere with the test. This is particularly important in using the helium sniffer test. The test should be conducted as soon after loading the tracer gas as possible to prevent loss of tracer gas.

The sensitivity of the helium leak test using the dynamic method is about 10^{-8} to 10^{-10} atm cm³/sec (0.05- to 0.005- μ m diam). The helium sniffer method has a sensitivity of about 10^{-4} to 10^{-6} atm cm³/sec (5 to 0.5- μ m diam). When the helium in the source capsule is diluted with air, nitrogen, argon, etc., the sensitivity of the test is degraded by the volume ratio of the helium to the total gas. The sensitivity of the ⁸⁵Kr method is about 10^{-8} to 10^{-10} atm cm³/sec.

2.8 Smear Test

The use of the smear test for detecting leaks in radioactive sources is based on the assumption that some of the radioactive material in a source can pass through a leak hole and be deposited on the outer surfaces of the source where it can be detected as transferable activity. The test is made by thoroughly wiping (no attempt is made to scrub) all accessible surfaces of the source with a piece of filter paper, a cotton medical applicator, or other suitable material of high wet strength and absorptive capacity. Both wet and dry smears are used. Wet smears are moistened with a liquid (usually water) that will not attack the source capsule material but which under the test conditions will effectively remove the radioisotope involved. The total activity of the smear is counted using a procedure which has been demonstrated to be effective in counting the radioisotope involved.

The smear test does not always give an indication of a leaking source capsule. Wet smears normally pick up a higher count than dry smears. However, in comparison tests where the wet smear showed a leak, the corresponding dry smear also showed a leak.⁵ When wet smears are used, consideration must be given in cases where attenuation of the radiation by the water is significant. This is particularly true of alpha particles having low penetrating power. When in doubt, it is advisable to count the smears both while wet and then after drying. Smear results may be confusing since it is difficult to tell a leaking capsule from a contaminated nonleaking capsule. For this reason, the capsule should be leak tested by at least one other method. The smear test is commonly used to observe for leaks of sources installed inside devices.⁹ When such sources are inaccessible,

the surrounding areas are smeared for evidence of a leak. When used in this manner, the periodic smear tests of sources in use would probably detect leakage of activity that would otherwise be unnoticed.

In the ORNL smear tests of leaking $^{137}\text{CsCl}$ test sources it was found that singly encapsulated test sources having leak holes of 30.0- to 46.7- μm diameter did not always deposit enough activity on the source surfaces over a 7- to 21-day period to be judged as leaking sources. Smear tests of doubly encapsulated test sources having inner and outer leak hole diameters of 19.1 and 24.6 μm , respectively, did not reveal any transferable activity over a period of 11 weeks when smeared at 7-day intervals (see Sect. 3.7.1).

2.9 Visual Examination

The value of a thorough visual examination of the source surfaces should not be discounted. In many instances, examination of the seal areas under moderate (5-30X) magnification clearly reveals porosity.

3. LABORATORY EVALUATIONS OF LEAK TESTS

3.1 Leak Tests of Leaking Cesium Chloride Test Sources

Various leak tests were performed on both singly and doubly encapsulated leaking CsCl test sources to obtain information relating to the reliability and sensitivity of the various tests. The tests were made to provide quantitative data on leakage of activity for various leak hole diameters under the test conditions. Most of the tests were performed using non-radioactive CsCl pellets as the source material to simplify the procedures. In some cases, radioactive $^{137}\text{CsCl}$ pellets were used in the test sources.

3.2 Test Source Design

Seven sets of stainless steel capsules were used in the tests. Each set had both an inner and an outer capsule of 316 stainless steel (Fig. 3.1). The end caps were sealed with neoprene O-rings. The 0.010-in.-thick capsule windows were made of type 304 stainless steel, and each window had a leak hole drilled at a point near its center. The leak hole sizes ranged from 12.7 to 57.9 μm . The capsules were interchangeable so that any inner capsule could be used with any outer capsule to obtain doubly encapsulated sources having various leak hole combinations. Tests were made to show that under the test conditions the O-ring seals did not leak. The source material used was either nonradioactive CsCl or radioactive $^{137}\text{CsCl}$, pressed into pellets at a pressure of $\sim 20,000$ psi. The pellets, weighing ~ 0.34 g (8.5 Ci), were 0.30 in. in diameter and 0.08 in. thick. They were positioned inside the capsules so that they would be in contact with the leak holes during the tests. Cesium chloride was chosen as the source material because of its high solubility in water, its wide use in commercial sources, and its high sensitivity to detection by means of flame photometry. The leak holes were made using the virtual electrode process.¹⁰ In

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Fig. 3.1. Inner and Outer Test Capsule Parts.

this process, the hole drilling action results when a virtual cathode (a very small diameter hollow glass tube containing an electrolyte) is advanced into the work (anode). The resulting electrolytic action forms a nearly round hole through the capsule window (Fig. 3.2). The leak hole diameters were measured by two methods — microscope and microphotography. A beam of light was passed through the leak holes to aid in these measurements. The internal void volume of the test sources was 0.84 cm^3 for the singly encapsulated sources and 5.11 cm^3 for the doubly encapsulated sources.

Fig. 3.2. Leak Hole Through Window of Test Capsule. The average diameter of the hole is $\sim 50 \text{ }\mu\text{m}$.

3.3 Leak Test Procedures Used on Test Sources

The procedures used for leak testing the CsCl sources are described below. In some cases, where appropriate, the leak test procedure is described in the section where the test results are given.

3.3.1 Water Leach Test

The source capsule was immersed with the leak hole facing down in 40 ml of distilled water (except where noted) at ambient temperature and pressure for various lengths of time. The water was not agitated except immediately prior to sampling. The leach water vessels were covered (but not sealed) to prevent contamination from outside sources and loss of water by evaporation. The leach water was maintained at constant volume by replacing the water removed in sampling. The data given in the tables are the total amounts of cesium leached from the source up to the time of sampling and have been adjusted to include the amounts previously removed for analysis. Data for the nonradioactive sources are expressed in micrograms of cesium (1 μg of fission-product cesium contains $\sim 32 \mu\text{Ci}$ of ^{137}Cs), and data for the radioactive ^{137}Cs are expressed in microcuries. The nonradioactive samples were analyzed by flame photometry, which has a sensitivity of 0.003 μg of cesium per milliliter. The radioactive samples were analyzed by a standard counting technique having a sensitivity of 100 dis/min.ml.

3.3.2 Vacuum Leach Test

The source was immersed in 100 ml of distilled water with the leak holes facing down, and the pressure above the water was slowly reduced to 2.5 psia for 3 min and then vented to atmosphere for 3 min. This vacuum-venting cycle was repeated four times. The leach water was then analyzed for cesium using the same analytical methods as used in the water leach tests.

3.3.3 Smear Tests

The smear test was used to detect transferable activity on selected surfaces of the sources and the test equipment. These surfaces were thoroughly wiped with cotton medical applicators using moderate pressure. Both wet and dry smears were used. The smears were counted in a shielded GM counter (end-window type) except as otherwise noted.

3.4 Water Leach and Vacuum Leach Tests of Leaking Nonradioactive Cesium Chloride Test Sources

3.4.1 Singly Encapsulated Test Sources

Tests were performed on singly encapsulated sources to obtain data on leach rate versus leak diameter, and test reliability. In the first two tests, four sources were immersed with the leak holes horizontal in the leach water. Both tests were performed using the same conditions to check for reproducibility. The degree of leak hole plugging at the end of each of

these two tests was estimated by microscopic observation of a beam of light passed through the leak hole and is therefore not quantitative.

Although the amounts of cesium leached from the sources for the first and second tests differ (Table 3.1), they appear to relate to the leak diameters and to leach time. The problem of leak plugging is evident, especially in the 12.7- μ m leak used in the second test where no cesium was detected in the leach water over the 7-day period. Some of the plugs appeared to be small pieces of CsCl that could usually be removed with water and methyl alcohol at the end of a test. However, plugs like those found in the 12.7- μ m leak (second test) required dilute nitric acid for removal. Examination of the pellets showed that CsCl had dissolved in the area of the leak hole in all cases except the source with the 12.7- μ m leak hole.

Table 3.1. Water Leach Tests of Singly Encapsulated Nonradioactive Cesium Chloride Test Sources

Leak Hole Diameter (μ m)	Amount of Cesium Leached from Capsule (μ g)					
	5 min ^a	1/2 hr ^a	2 hr ^a	1 day ^a	3 days ^a	7 days ^a
<u>Capsules Horizontal – First Test</u>						
12.7	b	56	82	599	—	1,185 ^c
22.9	b	b	90	—	1,806	84 ^c
33.0	176	244	406	1772	—	12 ^c
40.6	1624	1646	1644	1676	—	30
<u>Capsules Horizontal – Second Test</u>						
12.7	b	b	b	b	b	^c
22.9	b	b	b	b	140	109 ^d
33.0	18	54	124	3292	10,296	16,056 ^d
40.6	608	780	1801	2038	2,025	13,192

^aElapsed time before sample was taken.

^bSought, but not found.

^cLeak hole plugged at end of test.

^dLeak hole partly plugged at end of test.

All of the sources showed positive leak indications within 3 days except the source with the 12.7- μ m-diam leak (second test), and all sources having leak holes of 33.0- μ m diameter or larger had positive leak indications during the first 5 min of the tests.

The possibility that the CsCl pellets were not in contact with the leak holes throughout the tests could account for variation in the leach rates. Therefore, a test was performed in which the capsule having the 22.9- μ m leak hole was leach tested in a vertical position with the leak hole facing up (position of least pressure of the source pellet against the leak hole),

and the capsule having the 33.0- μ m leak hole was tested in a vertical position with the leak hole facing down (position of maximum pressure of the source pellet against the leak hole). This test was inconclusive since no cesium was found in the leach water of the capsule having the 33.0- μ m leak until the 2-hr sample was taken; however, the leach rate of the capsule with the 22.9- μ m leak was much higher than in the previous tests, and, for this reason, all four capsules were given a 3-day leach test with the leak holes facing up. During this 3-day test, none of the four capsules showed detectable cesium leaching. Examination of the source pellets showed that water had leaked into the pellets during the test. For comparison with these results, the test was repeated with the leak holes facing down (position of maximum pressure of the source pellet against the leak hole). The results of this test (Table 3.2) indicate an increased leakage rate — cesium was detected in the leach water of each of the four capsules at the end of 1/2 hr and in two instances at the end of 5 min.

Table 3.2. Water Leach Tests of Singly Encapsulated Nonradioactive Cesium Chloride Test Sources

Leak Hole Diameter (μ m)	Amount of Cesium Leached from Capsule (μ g)				
	5 min ^a	1/2 hr ^a	2 hr ^a	1 day ^a	5 days ^a
12.7	b	488	938	1698	1,989
22.9	48	2372	2882	3772	4,822
33.0	b	2080	2320	4090	7,830
50.6	35	181	636	1894	10,622

^aElapsed time before sample was taken.

^bSought, but not found.

It was noted during testing that capsule leak holes became plugged. A procedure was developed to determine the amount of plugging which occurred. By measuring the rate of water flow through the leak hole at a constant hydrostatic pressure of 9.25 psig, it was determined that the leak holes were free of plugs at the start of each test, and the degree of plugging could be measured at the end of each test.

The results of several of the leach tests of singly encapsulated nonradioactive cesium chloride sources indicated that the leach rates obtained during the first 3 to 5 days were not reproducible under the test conditions. In all cases but one, including those where cesium was not found in the leach water, examination of the pellets showed that water had leaked into the capsules as evidenced by the formation of pit-holes in the pellets at the point of the leak hole. In a test designed to eliminate any effect of surface irregularities (of pellets or the capsule windows), a thin layer of CsCl powder was placed between the pellets and the capsule

windows. In this test, only a small amount of CsCl was found in the leach water, probably due to contamination of the outer surface of the capsule by the loose powder.

Investigation into the causes of the inconsistent leach rate results obtained indicated that capsule orientation or plugging of the leak hole was at least partially responsible. In order to allow more time for water intake into the capsule void volume, an extended test (16 days) was performed. The amount of cesium found in the leach water of each of the four sources for the various leach times is given in Table 3.3. In this test, the capsules were positioned with the leak holes facing down.

Table 3.3. Water Leach Test Results of Singly Encapsulated Nonradioactive Cesium Chloride Test Sources

Leach Time (days)	Amount of Cesium Leached from Capsule (μg)			
	12.7 μm^a	22.9 μm^a	33.0 μm^a	40.6 μm^a
1	9	5	21	4,540
2	11	8	73	4,974
3	11	57	341	7,012
4	12	1052	394	10,082
7	11	3437	451	13,478
8	13	3833	462	15,388
9	13	3959	446	16,278
14	12	4097	461	18,083
15	13	4220	472	17,458
16	12	4268	477	17,698

^aLeak hole diameter.

Leach rates of the sources with the 22.9- and 40.6- μm -diam holes are comparable with some of the values obtained in previous tests. The decrease in the leach rate after about the fourth day may have been due to a lack of cesium in the vicinity of the leak hole since at the conclusion of the test the part of the pellet (about half) nearest the leak hole was dissolved. The leach rates of the capsules with the 12.7- and 33.0- μm -diam holes were extremely low compared to some of the values from previous tests and illustrate the type of variations experienced under the test conditions.

At the end of this test, the sources were rinsed in water to remove external cesium and were then subjected to the vacuum leach test. The absence of air-bubble evolution from the leak holes under vacuum indicated

that the holes of all four capsules were plugged or that the capsules contained enough water to prevent the evolution of air bubbles. The results of this test (given in Table 3.4) indicate that the total amount of cesium leached from the capsules during the vacuum leach test was related to leak hole diameter.

The vacuum leach test was repeated on the same source capsules after they were cleaned and unplugged, and new CsCl pellets were inserted in order to determine the effectiveness of this test using clean sources. In this test, air bubbles were evolved during the vacuum cycles. After the test, inspection showed that the CsCl pellet had been completely dissolved in each of the sources by the leach water, indicating a positive leak test of each of these sources.

Table 3.4. Vacuum Leach Test Results of Singly Encapsulated Nonradioactive Cesium Chloride Test Sources

Leak Hole Diameter (μm)	Total Cesium Leached (μg)
12.7	None
22.9	3.6
33.0	29
40.6	121

3.4.2 Doubly Encapsulated Test Sources

Four doubly encapsulated test sources were leach tested in 200 ml of distilled water for 16 days (Fig. 3.3). No cesium was detected in the leach water during this time indicating that the water leach test was not reliable under the test conditions. The leak holes (ranging from 12.7- to 57.9- μm diam) were checked before the leak test to be certain they were not plugged. At the end of the test, the leak holes in the inner capsules of the first two source assemblies were plugged. The CsCl pellets in each source were moist and contained a small air hole at the point where the pellet was in contact with the leak hole. The leak hole diameters of the sources are shown in Table 3.5.

Fig. 3.3. Water Leach Test of Doubly Encapsulated Test Source.

Table 3.5. Leak Hole Diameters of Doubly Encapsulated Nonradioactive Test Sources Used in Water Leach Test

Source	Leak Hole Diameters (μm)	
	Inner Capsule	Outer Capsule
1	40.6	25.1
2	33.0	57.9
3	22.9	28.7
4	12.7	24.6

Since the leak holes had been exposed to highly corrosive, aqueous CsCl for a considerable length of time during the tests, they were measured to determine if the diameters had increased appreciably. It was found that each of the inner capsule leak holes used had increased $\sim 6 \mu\text{m}$ in diameter. Most of the increase probably occurred during the 16-day leach tests. Since it was not possible to measure the leak hole diameters in the fabricated test sources with a microscope (because of source geometry), the diameters were obtained by comparing standard air flows through the capsule leaks with the flows through several spare leaks of known diameter. This method was used for the remainder of the tests to measure leak hole diameter and plugging.

Four doubly encapsulated CsCl sources were vacuum leach tested in 200 ml of water. Both inner and outer capsules had leak holes ranging from 19.1- to 57.9- μm diameter. When the source capsules were opened after the test, the CsCl pellets had almost completely dissolved and the void spaces in the capsules were nearly filled with water. Flow rates were measured before the test to ascertain that the capsule leak holes were not plugged and after the test to determine if plugging occurred during the test. The leak holes of the outer capsules were found to be free of plugging after the test. The amount of plugging of the inner capsule leak holes and the amount of cesium leached from each source are given in Table 3.6. Positive indications of leaks were obtained for each of the sources.

Table 3.6. Vacuum Leach Test Results of Doubly Encapsulated Nonradioactive Cesium Chloride Test Sources

Leak Hole Diameter (μm)		Cesium Leached from Source (μg)	Amount of Inner Capsule Plugging (%)
Inner Capsule	Outer Capsule		
19.1	24.6	5	98
30.0	25.1	27,800	42
35.6	57.9	10	8
46.7	28.7	37,600	3

The problem of plugging, evident in the test results, was probably not a result of corrosion of the stainless steel by the water solution of CsCl, since the length of the test (~25 min) was too short to allow for corrosion products to build up. The plugging was most likely due to solid CsCl entering the leak holes. The positive leak test results indicate that the vacuum leach test is reliable for water-soluble isotopes under the test conditions.

3.5 Water Leach Tests of Radioactive Doubly Encapsulated Leaking Cesium Chloride Test Sources

Two doubly encapsulated $^{137}\text{CsCl}$ sources were leach tested by immersing each source in 150 ml of water and sampling the water periodically for ^{137}Cs content. The sources were suspended with the leak hole facing down in an enclosed (but not sealed) glass vessel. The results of these tests are shown in Table 3.7. The first source was sampled after 35 days; the leach water contained 4.6 μCi of ^{137}Cs . The second source was sampled after 25 days; a total of 0.37 μCi of ^{137}Cs had been leached from the source. The leak holes were free of plugging before the test was started but were not checked for plugging after completion of the test.

The results of this test further illustrate previously noted discrepancies. Periods of plugging appear to have occurred during the period between the 1- and 7-day sample for the first source and between the 1/2-hr and 4-day sample of the second source. The source with the largest leak holes had the lowest leach rate rather than the highest leach rate as might be expected. The possibility exists that hydrogen and oxygen gases formed by radiolysis of water inside the source capsules might increase the internal pressure enough to prevent leach water from entering the sources as readily as in the case of nonradioactive CsCl. Both test sources showed definite leak indications in 1 day or less (0.005 μCi of transferable activity is regarded as a positive indication of a leaking source).

Table 3.7. Water Leach Tests of Doubly Encapsulated $^{137}\text{CsCl}$ Test Sources

Leak Hole Diameter (μm)		Amount of Cesium Leached from Source (μCi)					
Inner Capsule	Outer Capsule	5 min ^a	1/2 hr ^a	2 hr ^a	1 day ^a	4 days ^a	7 days ^a
35.6	57.9	b	b	b	0.026	c	0.026
19.1	28.7	b	0.011	0.011	c	0.011	0.270

^aElapsed time of test before sample was taken.

^bSought, but not found.

^cNot sampled.

3.6 Leak Test of Nonradioactive Doubly Encapsulated Leaking Cesium Chloride Test Sources Using an IAEA Test Procedure

A partial evaluation of the IAEA Leak Test² for source capsules (for "special form material") was made using doubly encapsulated stainless steel test sources loaded with inert CsCl pellets. Two sources were tested for leakage, and a third source without a CsCl pellet was tested simultaneously as a control. The diameters and the leak flows (measured at a pressure drop of 14.7 psig to atmospheric across the leak hole) are given in Table 3.8.

Table 3.8 Leak Diameters and Leak Flows
of Sources Used in IAEA Leak Test

Source	Capsule	Leak Diameter (μm)	Leak Rate ($\text{atm cm}^3 \text{ air/sec}$)
1	Inner	24.6	0.16
	Outer	17.3	0.09
2	Inner	22.1	0.13
	Outer	21.1	0.12
Control	Inner	27.9	0.19
	Outer	22.4	0.14

The sources were placed with the leak holes facing up in individual glass beakers containing 200 ml of distilled water for testing. The temperature was raised to $50 \pm 5^\circ\text{C}$ and held there for 8 hr, then the sources were removed from the water and stored in air for 7 days. The initial step was repeated using fresh water and clean beakers. The water from each of the two 8-hr immersions was reduced to 10 ml by evaporation and analyzed for cesium by flame photometry (limit of detection $0.003 \mu\text{g/ml}$).

Analysis of the immersion water from each 8-hr immersion period (Table 3.9) indicated that both test sources were leaking. The control source showed leakage in the first immersion but not in the second immersion. This leak test procedure, although lengthy (~ 9 days), appears to work well with sources having leak holes $\sim 25 \mu\text{m}$ in diameter. The fact that the control source showed leak indications points out the need for cleanliness in the test area to maintain a low background contamination.

Table 3.9. Results of Leak Test of Doubly Encapsulated Nonradioactive Cesium Chloride Test Sources Using IAEA Procedure

	Amount ^a of Cesium in Immersion Water (μg)		
	Source 1	Source 2	Control Source
First immersion	1.06	6.3	0.83
Second immersion	0.18	1.38	0.0

^aOne microgram of fission-product cesium contains ~32 μCi of ¹³⁷Cs. An amount of transferable activity 0.05 μCi or greater constitutes a leaking source under the IAEA regulation.

3.7 Smear Tests of Radioactive Leaking Cesium Chloride Test Sources

3.7.1 Singly Encapsulated Test Sources

Smear tests were made on leaking, singly encapsulated CsCl test sources to obtain data on reliability and sensitivity of the smear test. These tests were made at 7-day intervals to provide data directly related to the 7-day smear test used in leak testing commercial sources. The capsule was covered with aluminum foil during loading to prevent contamination of the outer surfaces. A moist paper wad was placed immediately below the leak hole to collect any activity which might fall through the leak hole during the pellet loading operation. Immediately after loading, the capsule was smear tested for initial activity, and the activity on the paper wad was measured. The individual capsules were placed under glass jars to prevent contamination from external sources, with capsules positioned so that the leak holes would be facing up in all cases except one in which the leak hole was facing down. The results of the smear tests are shown in Table 3.10. The results show that activity can fall through the leak holes during the loading operation without leaving transferable activity on the exposed surfaces of the capsule windows. The leak holes in these sources were large (30.0- to 46.7-μm diam) yet only two of the seven sources showed positive leaks in the 7-day smear test. Only four of the sources exceeded the allowable activity on smears taken the 14th day, and the smear tests of one of the sources remained at background for 21 days. For the three sources where wet smears were used exclusively, the 7-day smear test did not reveal the presence of the leak holes. In the three cases where dry smears were used exclusively, the 7-day smear test revealed two of the leaks, indicating that wet smears may cause temporary plugging of the leak holes. The possibility of leak hole position (facing up or down) being a factor is inconclusive based on the single test made.

The sources used in these tests were not sealed by welding, and decontamination was not necessary. It is possible that either of these procedures could have an effect on both the initial and 7-day smear tests.

Table 3.10. Results of Smear Tests of Singly Encapsulated Leaking $^{137}\text{CsCl}$ Test Sources

Test	Amount of Activity on Smears (μCi)						
	30.0 μm^a	30.0 μm^a	30.0 μm^a	30.0 μm^a	30.0 μm^a	46.7 μm^a	46.7 μm^a
Leak hole position	down	up	up	up	up	up	up
Paper wads	0.16	d	0.00012	0.000036	b	0.039	0.20
Initial	b,c	b,c	b	b	b,c	b,c	b
7 day	b,c	b,c	0.24	0.18	b,c	0.00017	b
14 day	0.00013 ^c	0.073 ^c	0.32	0.064 ^c	b,c	0.028	0.00011
21 day	d	0.021 ^c	d	d	b,c	d	0.055

^aLeak hole diameter.

^bBackground

^cWet smears.

^dNot determined.

3.7.2 Doubly Encapsulated Test Sources

Smear tests were also conducted on a doubly encapsulated source (Fig. 3.4) using the same procedure as for the singly encapsulated sources. The capsule was positioned so that the leak hole was facing up, and only dry smears were taken. The leak hole diameters of the inner and outer capsules were 19.9 and 24.6 μm , respectively. During the pellet loading operation, $\sim 0.03 \mu\text{Ci}$ of $^{137}\text{CsCl}$ fell through the leak hole of the inner capsule. Initial smear tests of both the inner and outer capsules showed that the external surfaces were free of transferable contamination. Dry smear tests of the capsule taken at 7-day intervals over a period of 11 weeks did not reveal any transferable activity. In an effort to cause activity to pass through the leak holes, the source was alternately evacuated and vented to atmosphere five times. An additional smear test of the window did not reveal any activity. The leak holes were checked and found to be free of plugging at the conclusion of the testing.

Fig. 3.4. Smear Test of Doubly Encapsulated Test Source.

3.8 Elevated Temperature Tests of a Doubly Encapsulated Cesium Chloride Test Source

Elevated temperature tests were conducted on a doubly encapsulated $^{137}\text{CsCl}$ source made from the test capsules. The source was heated for 1 hr at 400°C and for 1 hr at 925°C . The leak hole diameters in the inner and outer capsules were 35.6 and 57.9 μm , respectively.

The source used for the elevated-temperature tests had previously been used for a water leach test. Vacuum leak tests indicated that the leak hole of the inner capsule was plugged. The O-rings were removed from both inner and outer capsules, and the outer capsule was decontaminated to 35,000 dis/min. For testing, the source was placed inside a 250-ml stainless steel beaker (with a loose fitting lid) and heated in a crucible furnace for 1 hr at the desired temperature. Smear tests (Table 3.11) were used to observe the extent of contamination resulting from the tests.

Table 3.11. Smear Test of $^{137}\text{CsCl}$ Source Environment After Heating for One Hour

Area Smeared	Smear Count. (μCi)	
	400°C^a	925°C^b
<u>Outside Surface</u>		
Beaker Lid	c	2.27
Beaker walls	c	0.45
Capsule end cap	0.00018	1.36
Furnace walls	d	0.0055
Capsule window	0.035	—
<u>Inside Surface</u>		
Beaker lid	0.00086	0.045
Beaker walls	c	2.27
Furnace lid	d	0.045
Furnace walls	d	0.0045
Beaker bottom	0.00086	d
<u>Hot Cell Wall</u>		
Near furnace	d	c

^aCounted using a shielded GM counter (end window type).

^bCounted using a GM survey meter.

^cBackground.

^dNot smear tested.

After the tests at elevated temperatures were completed, the source was rinsed with water, scrubbed with a cotton applicator, and rinsed again with water. The resulting solution contained 1.44 μCi of ^{137}Cs .

This test illustrates the method of smear testing the source surroundings to detect activity leakage. It also provides an indication of the amount of activity that might leak from this type of source under the test conditions. In the test at 400°C only one smear (capsule window) is significant. At 925°C the $^{137}\text{CsCl}$ is a liquid (melting point 645°C) and the smear values are, as expected, much higher. Figure 3.5 shows the source after the tests.

PHOTO 88023

Fig. 3.5. Doubly Encapsulated Test Source After Exposure to Elevated Temperature Tests.

3.9 Discussion

3.9.1 Water Leach Tests

In the water leach tests, the amounts of cesium found in the leach water appeared to be related to leak hole diameter and leach time. This relationship was often obscured by other factors, such as leak hole plugging, capsule orientation, and atmospheric changes. Water entered the test sources through the leak holes as evidenced by the formation of pit holes in the CsCl pellets at the point of the leak hole, but in many instances CsCl was not detected in the leach water.

During testing, the leak holes became plugged with what appeared to be CsCl in some cases and corrosion products in others. The amount of this plugging was uncertain, since the methods of measurement might have increased the amount of plugging by compacting solids into the plugged areas or might have decreased the amount of plugging by moving solids out of the plugged area. Microscopic examinations made before these measurements clearly revealed solid material in the leak holes. In doubly encapsulated test sources, plugging was often found in the inner capsule leak holes but not in the outer capsule leak holes, indicating that the plugging problem was internal rather than external to the source.

Capsule orientation during the water leach testing appeared to be important. The water leach test was least effective when the test capsule leak holes were facing up. Apparently a continuous water path between the CsCl pellet and the leach water bath did not always exist. Perhaps water entering a test source through a leak hole at the top of the capsule could flow away from the leak hole due to gravity or absorption by the pellet, thereby not always forming a continuous leak path until the available void space became filled with water. When water entered a leak hole at the bottom of the

test capsule, a relatively small pool of water between the pellet and the leak hole could provide a direct leak path from the pellet to the leach water bath.

In those instances where the water leach test indicated leaks, changes in barometric pressure or ambient temperature might have occurred before plugging sealed the leak holes. Such changes are normal and can produce pressure drops across the leak holes which greatly exceed that produced by the liquid head of the water bath. The sensitivity and the reliability of this test may therefore depend heavily on atmospheric changes.

The limit of detection for the water leach test was not determined. For the singly encapsulated nonradioactive sources, leaks ranging from 12.7- to 40.6- μm diameter were detected in 1/2 hr. Leaks in four doubly encapsulated nonradioactive sources, ranging from 12.7- to 40.6- μm diameter for the inner capsules and 24.6- to 57.9- μm diameter for the outer capsules, were not found when leached for 16 days. For two doubly encapsulated radioactive sources having leak holes of 35.6- and 19.1- μm diameter in the inner capsules and 57.9- and 28.7- μm diameter in the outer capsules, the leaks were detected in 1 day and 1/2 hr, respectively.

Due to the observed effects of leak hole plugging, capsule orientation, and atmospheric changes, the use of the water leach test procedure on CsCl sources does not appear to have the desired reliability.

3.9.2 Vacuum Leach Test

The vacuum leach test detected leaks in the test capsules in all cases where it was known that the leak holes were not plugged. Air bubbles, evolved during the first vacuum cycle, indicated the presence of the leaks and their location on the surface of the test capsule windows. When these sources were disassembled, the CsCl pellets were completely dissolved. When bubbles were not evolved during the vacuum cycles due to known leak hole plugging, three out of four leaking test capsules were identified by analysis of the leach water. No evidence of corrosion was found during the vacuum leach tests, probably because the time required to perform the test was very short (~ 24 min). Over the leak hole size range tested (12.7- to 57.9- μm diam) no significant differences were found between singly or doubly encapsulated test sources.

3.9.3 Smear Tests

The results of the smear tests of the test capsules indicate that singly encapsulated sources having large leak holes of 30.0- to 46.7- μm diameter might not deposit enough activity on the source surfaces to be judged leaking sources over a period of 7 to 21 days. Smear tests of a doubly encapsulated test source having inner and outer leak hole diameters of 19.1 and 24.6 μm , respectively, did not reveal any transferable activity over a period of 11 weeks when it was smeared at 7-day intervals. There were some indications that the wet smears may have caused temporary plugging of the leak holes. The smear tests following the elevated temperature tests illustrate the method of smear testing the source surroundings to detect leakage of activity from a source.

3.10 Evaluation of the Vacuum Leak Test

An evaluation of the vacuum leak test⁴ was performed in 1964 as a part of the ORNL Source Safety Testing Program. Leak rates were evaluated in terms of both leak diameter and differential pressure required to initiate bubbling in order to correlate the test sensitivity with the size of the leak hole.

Experimental leaks were made from glass capillary tubing using a heating and drawing technique which reduced the capillary diameter to the desired size range. This method resulted in a continuous taper to a point of minimum diameter at which the capillary was broken cleanly.

A McLeod gage system was used to calibrate the leaks in terms of atm cm³/sec (air at 25°C) leaking into an evacuated chamber held at less than 0.5 psia. The upstream pressure of the leak was maintained at 14.7 psia.

The leak diameters (microns) were measured at the tips (point of minimum diameter) using a microscope equipped with an oil immersion lens. A beam of light was reflected through the capillary to the tip to improve visibility.

The differential pressures (psi) required to initiate bubbling in isopropyl alcohol were measured using vacuums to regulate the downstream pressures. The upstream pressures were maintained at 14.7 psia except for the higher differential pressures which required increasing the upstream pressures to initiate bubbling and to prevent serious outgassing of the isopropyl alcohol. The data obtained from the above measurements are plotted in Figs. 3.6 and 3.7.

Comparison of the leak diameter and the differential required to initiate bubbling for any given flow indicates that the experimental data agree closely with those predicted from the theoretical relationship for round leak holes, $\Delta P = 4T/D$, where ΔP is the differential pressure required to initiate bubbling, D is the leak diameter, and T is the surface tension of the liquid.

This equation was developed by equating the surface tension force acting on the periphery of the leak to the force due to differential pressure acting on the cross-sectional area of the leak as follows:

$$F = \pi DT$$

where

- F = total force due to surface tension (dynes)
- D = diameter of capillary at smallest point (cm)
- T = surface tension of liquid (dynes/cm)

and

$$F' = (\pi/4)D^2\Delta P$$

where

- F' = total force due to differential pressure (dynes)
- D = diameter of capillary at smallest point (cm)
- ΔP = differential pressure across leak (dynes/cm²)

Fig. 3.6. Correlation of Leak Rate with Leak Diameter.

Fig. 3.7. Relationship Between Leak Rate and Differential Pressure Required to Initiate Bubbling in Isopropyl Alcohol.

Neglecting the effect of hydrostatic pressure and equating F with F' ,

$$\Delta P = 4T/r$$

where

ΔP = differential pressure (dynes/cm²)

T = surface tension of liquid (dynes/cm)

D = diameter of capillary at smallest point (cm)

The practical limit at which serious outgassing became a problem occurred at a vacuum of ~ 3 to 4 psia for the liquids commonly used in vacuum leak testing, and for this reason a maximum vacuum of 3.4 psia (12.3 psi ΔP) was recommended. The sensitivity of the test is obtained by referring to Fig. 3.7; a ΔP of 12.3 psi corresponds to a measured leak rate of $\sim 4 \times 10^{-6}$ atm cm³/sec. The measured leak diameter for this leak rate is ~ 1.0 μ (Fig. 3.6).

To increase the sensitivity of the test, a study of pressurization prior to leak testing was included. Calibrated containers of known volume were subjected to an external gage pressure of 1000 psi, and the internal pressure was measured as a function of time. Both helium and nitrogen were used as the pressurizing gas with identical results as shown in Fig. 3.8.

A study was also conducted on the effect of internal void volume on leak test sensitivity. Calibrated experimental leaks with leak rates of 1×10^{-7} to 1×10^{-5} atm cm³/sec and internal void volumes of 0.1 to 0.5 cm³ were tested. Positive results were obtained in all cases, and there was no indication that the test sensitivity was affected by void volume over the range studied.

Upon completion of the basic studies, the following procedure was developed for the testing of radioisotope source capsules:

- A. Establish desired leak test sensitivity as a function of the particle-size distribution of the material to be contained.
- B. Estimate pressurizing requirements.
 1. Determine the theoretical differential pressure requirement for the smallest leak to be detected (Figs. 3.6 and 3.7).
 2. Establish an actual differential pressure requirement by applying a safety factor of +25% to the theoretical requirement.
 3. Calculate the amount by which the internal container pressure must be increased. (Assuming that the container is initially at atmospheric pressure, subtract 12.3 psi from the actual differential pressure requirement to account for the vacuum created during vacuum leak testing.)
 4. Estimate the capsule internal void volume.

Fig. 3.8. Rate of Void Pressure Increase.

5. Calculate the pressurization requirement in terms of hours of exposure to 1000 psig, based on the calculation of the required pressure increase, the estimation of internal void volume, and the pressurization data in Fig. 3.8. If the container cannot withstand 1000 psig, a lower external pressure must be used with a corresponding increase in the time required for testing.

C. Make vacuum leak test in isopropyl alcohol.

1. Evacuate the leak testing chamber for 1 min or longer to lower the dissolved air content of the isopropyl alcohol.
2. Transfer the capsule in question to the leak testing chamber within 10 min after termination of the pressurizing operation.
3. Evacuate the leak testing chamber slowly and observe for the positive indication of leaks through the evolution of bubbles; do not decrease pressure below 2.4 psia.

The validity of the procedure was confirmed by actual testing. Two simulated isotope containers, with leaks of 8.0×10^{-7} and 4.5×10^{-7} cm³/sec, were subjected to the vacuum leak testing procedure with positive results.

The conventional leak testing procedure, without prepressurization, is applicable to the detection of container leaks down to ~ 1.0 μ m in diameter. The new procedure can be used to detect holes as small as 0.1- μ m diameter in any container that can be prepressurized to about 125 psig. Much smaller leaks can be detected if there is time and apparatus to carry out the extended prepressurization procedure; the smaller the leak, the greater the prepressurization required. This study did not reveal any limit to the test sensitivity.

3.11 Evaluation of Large Experimental Leaks

The evaluation of the vacuum leak test was limited to the range of the smaller leak diameters (~ 0.05 to 5 μ m). In order to evaluate the various leak tests performed on the cesium chloride test sources, data were needed for the larger leaks. It was also desirable to be able to correlate activity losses with leak diameters under various leak test conditions and to determine test sensitivity.

Both glass and metal leaks were fabricated and measured. The glass leaks were made using the heating and drawing technique discussed earlier, and the metal leaks were made using the virtual electrode technique. About 100 glass leaks and 50 metal leaks were fabricated and measured; no significant differences were found in the results obtained from these two types of leaks.

The leak rates were measured with the pressure upstream of the leak at 29.4 psia and the pressure downstream of the leak at 14.7 psia. The resulting leak rates were corrected to atm cm³ air per sec at 25°C.

The differential pressures required to initiate bubbling in water, ethylene glycol, and isopropyl alcohol were measured with the pressure downstream of the leak at 14.7 psia. The pressure upstream of the leak was varied to obtain the required ΔP .

The data showing the relationship between leak rate and leak diameter are shown in Fig. 3.9, and the data showing the relationship between leak rate and differential pressure required to initiate bubbling are shown in Fig. 3.10. Only a portion of the points are shown; these points were selected to accurately represent a complete plot of the data. Comparison of the leak rates obtained in this study with those obtained in the evaluation of the vacuum leak test shows a considerable difference in the leak rate obtained for a given leak diameter. For example, the leak rate of a 5- μ m-diam leak taken from Fig. 3.9 is 1.03×10^{-2} atm cm³/sec, whereas the leak rate obtained in evaluating the vacuum leak test is 1×10^{-4} atm cm³ air/sec (Fig. 3.6). This difference in leak rates is attributed to the compressibility of gases, the leak rate being dependent not only on the differential pressure across the leak but also on the mean absolute pressure within the capillary. The leak rates measured in this section will accordingly be greater since the mean absolute pressure within the leak is $(29.4 + 14.7)/2 = \sim 22$ psia, whereas the mean absolute pressure was $(14.7 + 0.5)/2 = 7.6$ psia for the leak rate method used in evaluating the vacuum leak test.

For comparison, an additional set of three curves was drawn (Fig. 3.11) showing the relationship between leak diameter and the ΔP required to initiate bubbling in isopropyl alcohol. Data for the first curve were taken from the measurements of the larger leaks described in this section. The second curve was drawn from the data presented in the evaluation of the vacuum leak tests (Figs. 3.6 and 3.7). The third curve was drawn by calculating the ΔP 's required to initiate bubbling for various diameter; using the theoretical relationship $\Delta P = 4T/D$.

The curve drawn from the vacuum leak test evaluation follows the theoretical equation reasonably well over most of the range covered, but the curve drawn from the measurements for the evaluation of the large leaks made in this section varies considerably from both of the other curves. For example, for a leak diameter of 0.33 μ m, a ΔP of 44 psi was required to initiate bubbling in isopropyl alcohol for the vacuum leak test, whereas only 10.2 psi was required in the current study of large leaks. The reason for this difference is not known, but it is probably due to the effect of the kinetic energy of the gas flowing through the leak hole during the ΔP measurement. The kinetic energy of the air flowing through a given leak varies directly with the square of the velocity. Much of this energy is available to overcome the force of surface tension and to initiate bubbling. The velocity of the air passing through a given leak size depends largely on the ΔP and the mean absolute pressure of the air in the leak. Taking the mean absolute pressure ($\bar{P}$) in the leak to be the sum of the inlet and outlet pressure divided by two, then the mean absolute pressure in the leak when using the vacuum leak test is (for values of $0 < \Delta P < 14.7$ psi)

$$\bar{P} = (14.7 + 14.7 - \Delta P)/2 = 14.7 - (\Delta P/2) .$$

Fig. 3.9. Relationship Between Leak Rate and Leak Diameter.

Fig. 3.10. Relationship Between Leak Rate and ΔP Required to Initiate Bubbling in Various Liquids.

Fig. 3.11. Relationship Between Leak Diameter and ΔP Required to Initiate Bubbling in Isopropyl Alcohol.

and, for the method used in this section to measure the larger leaks, the mean absolute pressure in the leak during the ΔP measurement is (for values of $\Delta P > 0$)

$$\bar{P} = (14.7 + \Delta P + 14.7)/2 = 14.7 + (\Delta P/2) .$$

With large leaks (low ΔP) the effect of ΔP on $\bar{P}$ is small, but as the leak diameter decreases the ΔP to initiate bubbling increases and $\bar{P}$ becomes much more important. As the ΔP increases in the vacuum leak test method $\bar{P}$ decreases, whereas for the method used in this section $\bar{P}$ increases. The contribution of $\bar{P}$ to the kinetic energy of the air passing through progressively smaller leak holes becomes considerably different for the two methods used for measuring ΔP . For example, the velocity of air exiting from a 1- μm -diam leak hole (based on air at atmospheric pressure at 25°C) is 510 cm/sec when using the vacuum leak test method and 85,400 cm/sec for the method used in this section. Data are not available for the vacuum leak test for leak diameters greater than 5 μm , so comparison of the curves in this area is not possible.

3.12 Evaluation of the Liquid Nitrogen Leak Test

In this test, the source is immersed in liquid nitrogen (-195°C) for a predetermined time during which cooling of the source causes a considerable reduction of the gas pressure inside the capsule void volume. If a leak exists, liquid nitrogen can be drawn into the capsule or it can evaporate and be drawn into the capsule void as a gas. The source is next removed from the liquid nitrogen bath and immediately placed in a bath of hot (90°C) water. Any liquid nitrogen in the source will be rapidly vaporized and escape through the leak hole leaving a stream of bubbles in the water. If nitrogen is drawn into the source capsule as a gas, rather than a liquid, it will expand and produce a stream of bubbles in the hot water. If no nitrogen is drawn into the source capsule during immersion, bubbling can still occur when the source is placed in the hot water due to expansion of air that is initially present in the source at the time of manufacture.

In order for this test to have a high sensitivity, ($\sim 10^{-6}$ atm cm^3/sec or better) nitrogen, either as a liquid or a gas, must be drawn into the void volume during immersion in the liquid nitrogen bath. If this does not occur the advantage of cooling the source to -195°C will not be realized, and the sensitivity of the test will be the same as that of the hot-water bubble test ($\sim 10^{-4}$ atm cm^3/sec). If sufficient gaseous nitrogen enters the capsule to raise the pressure in the void space to ~ 1 atm, or if an equal weight of liquid nitrogen enters, then a differential pressure of ~ 3.6 atm (~ 53 psi) can develop across the leak hole when the source is heated to 90°C in the hot-water bath.

3.12.1 Tests Using Glass Ampuls

The liquid nitrogen test was evaluated because of its potentially high sensitivity resulting from the large differential pressure developed when the sources are heated.

The initial tests were performed using glass ampuls, made by the heating and drawing technique, for evaluating the vacuum leak test. The open end of the ampuls, opposite the leak holes, were sealed in a flame. The internal void volume of the ampuls was $\sim 0.3 \text{ cm}^3$. The leak hole diameters ranged from 2.5 to 56 μm .

Each of 9 ampuls under test was immersed individually in a dewar of liquid nitrogen for varied lengths of time. The ampul under test was then quickly transferred to a container of hot (90°C) water. In all of the tests performed, no liquid nitrogen was observed inside the ampuls even when they had been immersed for more than 1 hr. Immersion times of 5 to 10 min in the liquid nitrogen were sufficient to give maximum bubble duration in the hot-water bath. Several of the leak holes became plugged during the tests, probably due to moisture freezing in the leak holes during immersion in liquid nitrogen. Baking the ampuls in a furnace for several hours usually unplugged these leak holes.

3.12.2 Tests on ^{90}Sr Sources

Tests were also performed on 72 encapsulated ^{90}Sr sources, some of which were known to be leaking activity. The sources, in three separate groups, were completely immersed in liquid nitrogen for ~ 1.5 hr, then removed individually from the dewar, and immediately and completely immersed in a large glass graduate cylinder filled with water at $\sim 90^\circ\text{C}$. The entire length of each capsule was scanned with a monocular for a stream of bubbles which would indicate a leak; seven of the 72 capsules were observed to leak. These seven were retested using the same procedure with the exception of immersion of the capsules in liquid nitrogen for ~ 2 hr. After completing the test the second time, only four of the original seven were observed to leak. Water might have entered the capsules during the first test and prevented liquid nitrogen from entering the capsules during the second test.

A vacuum leak test using ethylene glycol was then conducted on the seven capsules; only three were observed to leak in this test.

The tests indicate that the liquid nitrogen test is unreliable due to the possibility of ice plugging the leak holes.

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APPENDIX A

Approximate Leak Test Sensitivities

Test	Leak Rate ^a (atm cm ³ /sec)	Leak Diameter (μm)	Other
Smear	Unknown	Unknown	
Immersion	Unknown	Unknown	0.0002 μg lithium/ml 100 dis/min.ml
Hot water bubble	2.8×10^{-4}	8.5	
Weight gain	10^{-5} to 10^{-7}	1.7 to 0.17	
Helium "sniffer"	10^{-4} to 10^{-6}	5 to 0.5	
Vacuum leach	Same as vacuum leak test if bubbles are evolved Same as immersion test if bubbles are not evolved		
Vacuum leak (with pressurization)	4×10^{-6} 4×10^{-7} b	1 0.1 ^b	
Helium mass spec- trometer with bell jar	10^{-8} to 10^{-10}	0.05 to 0.005	
Krypton-85	10^{-8} to 10^{-10}	0.05 to 0.005	

^aThe leak rates refer to gas flowing through leaks having an upstream pressure of 14.7 psia and a downstream pressure of <0.5 psia.

^bOr smaller.

APPENDIX B

Examples of Typical Leak Test Procedures*

Immersion Leak Test

1. Immerse the source capsule for 24 hr in a liquid which will dissolve the source material but not attack the source capsule.
2. Remove the source capsule slowly, allowing the liquid to drain back into the bath.
3. Reduce the volume of the liquid by evaporation to obtain the maximum count rate.
4. If the liquid contains more than 0.005 μCi , the source is regarded as leaking.

Vacuum Leak Test

1. Evacuate the leak test chamber for 1 min or longer to lower the dissolved air content of the leak test fluid (reagent-grade ethylene glycol or isopropyl alcohol).
2. Place the source capsule in the leak test chamber, making certain that it is completely submerged to a depth of 2 in. below the fluid level.
3. Evacuate the leak test chamber slowly to 2.5 psia, while observing for the indication of leaks by bubbles rising from the source capsule surface. If no leaks are observed over a period of 5 min, the source capsule passes this leak test.

Vacuum Leach Test

1. Evacuate the leak test chamber for 1 min or longer to lower the dissolved air content of the leak test fluid.
2. Place the source capsule in the leak test chamber, making certain that it is completely submerged to a depth of 2 in. below the liquid level. The liquid should be one which will dissolve the source material but not attack the source capsule.

*The leak test procedures are stated in general terms and are not applicable to all leak test situations. Changes in the procedures may be necessary to meet the requirements of source geometry, test reliability and sensitivity. The applicable parts of Sections 2.0 to 3.9 should be considered when applying these test procedures to a particular source design.

3. Evacuate the leak test chamber slowly to 2.5 psia and look for leaks (indicated by bubbles rising from the source surface). Maintain at this pressure for 3 min. Vent the chamber to atmospheric pressure and maintain at this pressure for 3 min. Repeat this vacuum-venting procedure four times. If leak indications are present during any of the vacuum cycles, the source fails this test.
4. Remove the source capsule slowly, allowing the water to drain back into the vacuum chamber.
5. Reduce the volume of the water by evaporation to obtain the maximum count rate.
6. If the water contains more than 0.005 μCi , the source is regarded as leaking.

Hot Water Bubble Test

1. Boil a container of distilled water to drive off the dissolved air.
2. Cool the water to $\sim 90^\circ\text{C}$ and maintain at this temperature during the leak test.
3. Immerse the source capsule in the container of water to a depth of at least 2 in. and observe for a stream of bubbles, emanating from a point on the capsule, indicating a leak.

Weight Gain Test

1. Clean the source capsule to remove any solids which might later become dislodged during the test.
2. Weigh the source to the nearest 0.0001 g.
3. Place the source in the water-filled pressure vessel and raise the pressure to 300 psig for 1 hr.
4. Remove the source from the pressure vessel and dry it.
5. Weigh the source to the nearest 0.0001 g. A weight gain of more than 0.0005 g indicates a leak.

Helium Mass Spectrometer Leak Test

1. Determine the leak detector sensitivity using a standard helium leak.
2. Place the welded, cleaned source capsule inside the leak test chamber and evacuate the chamber to the extent that the helium leak detector will operate in test condition with the throttle valve wide open.

3. Observe the leak detector output for helium indications for 5 min.
4. Repeat step 1.
5. In order for the test to be valid, both the leak detector sensitivity and the system background should not have changed significantly during the test.
6. Calculate the leak flow based on the ratio of helium to total gas in the source capsule during the test.

Krypton-85 Leak Test

1. Place the welded, cleaned source inside the pressurization chamber.
2. Evacuate the chamber and introduce a known amount of ^{85}Kr . Pressurize the chamber with argon to ~45 psia.
3. Allow the system to stand for the precalculated period required for the desired test sensitivity.
4. Vent the chamber and place the source in a clean test chamber.
5. Purge the chamber with argon at atmospheric pressure and allow the system to stand for the precalculated collection period.
6. Draw a fraction of the chamber atmosphere into an evacuated, calibrated ^{85}Kr counting chamber.
7. Calculate the leak rates from the count rate.

Smear Test

1. Thoroughly wipe all accessible surfaces of the source with a piece of filter paper, a cotton applicator, or other suitable material of high wet strength and absorptive capacity. If wet smears are used, select a liquid which will not attack the capsules, but will effectively remove the radioisotope involved.
2. Count the total activity on the smear using a procedure which has been demonstrated to be effective in counting the radioisotope involved. A smear having $>0.005 \mu\text{Ci}$ indicates a leaking source.