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**RELEASE FRACTIONS FOR RADIOACTIVE  
SOURCES IN FIRES**

**FS/99/19**

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HEALTH AND SAFETY LABORATORY  
An agency of the Health and Safety Executive

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## Summary

### Objectives

The purpose of this study is to provide the necessary information to allow third parties to make an informed decision as to whether they must conform with the Radiation (Emergency Preparedness and Public Information) Regulations (REPPIR, 1999). The REPPIR package applies if the inventory of an active material on site exceeds the quantity listed in Appendix 1 of the regulations, or if the total activity arising from a number active materials exceeds a specified level. The quantity listed is that which would give rise to a 5 mSv dose to a member of the public should all the material be dispersed to atmosphere.

At present no recognition is given to the physical form of the materials, their reactivity/volatility if unsealed, or of protection afforded by the use of sealed sources. The aim of this report is therefore to consider the effects of fire engulfment, and to assess the likely release fraction should the integrity of the source fail exposing the active component.

### Main Findings

This review has identified a number of possible release fractions for radioactive materials in fires. Two flow charts have been developed, to help the user identify the relevant issues and hence estimate a possible release fraction. The approach taken is first to establish the likelihood of a sealed source retaining its integrity in a fire and if not to examine, from the stability of the active material, the extent of any release. A similar approach is taken with unsealed sources.

Three methods have been identified to assess possible release from sources:

- Study of UN Transport Classification test details where available;
- use of ANSI/ISO classification details in conjunction with chemical/physical properties identified in this report: and,
- study of NRPB test data for consumer goods containing radioactive sources.

Other findings are as follows:

Most sealed sources subjected to the ISO temperature test class 6 (800 °C for 1 hour) can be regarded as unlikely to fail when heated in a fire, unless the materials used in the construction of the source are likely to melt or oxidise.

The long term stability of many sources appears very good. Care must be exercised, however, in determining the possibility of extending the working life of time-expired alpha emitting sources, since, given too long an extension, the internal Pressure may rise to such a level as to cause the source to fail if heated in a fire.

Discussions with a major source manufacturer suggest that, should a capsule fail, the release fraction for a ceramic/enamel source is likely to be <1 %.

Due to a scarcity of data on the fire performance of many materials used in source manufacture, it has been impossible to identify release fractions for large numbers of potential radioactive isotopes. However, this may not be a problem as many isotopes have no commercial use. In the absence of good data, indicative levels of oxidation and hence possible release have been identified for most metals as follows:

flammable metals - 100 % release for thin/small sections or powders;

metals which melt/boil in a fire - 100 % release; and,

oxidisable metals - varying release depending on the properties of oxides.

In the latter case, metals oxidised in a fire, it is impossible to estimate a release fraction for containment failure. This is due to a lack of information on the extent to which metals are oxidised in a fire, and the rate and the tenacity of the oxide coating thus formed.

### **Main Recommendations**

It is recommended that should release fractions be required for metals susceptible to oxidation in a fire, further work be undertaken to either carry forward a more detailed search, or conduct tests to actually measure losses from heated samples.

It is also recommended that should reliable figures be required to establish the possibility of source failure, fire tests should be undertaken on a range of dummy source types, for example, titanium or beryllium windowed sources.

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## **1. INTRODUCTION**

The purpose of this study is to provide the necessary information to allow third parties to make an informed decision as to whether they must conform with the Radiation (Emergency Preparedness and Public Information) Regulations (REPPPIR). The REPPPIR package applies if the inventory of an active material on site exceeds the quantity listed in Appendix 1 of the regulations, or if the total activity arising from a number active materials exceeds a certain level. The quantity listed is that which would give rise to a 5 mSv dose to a member of the public should all the material be dispersed to atmosphere.

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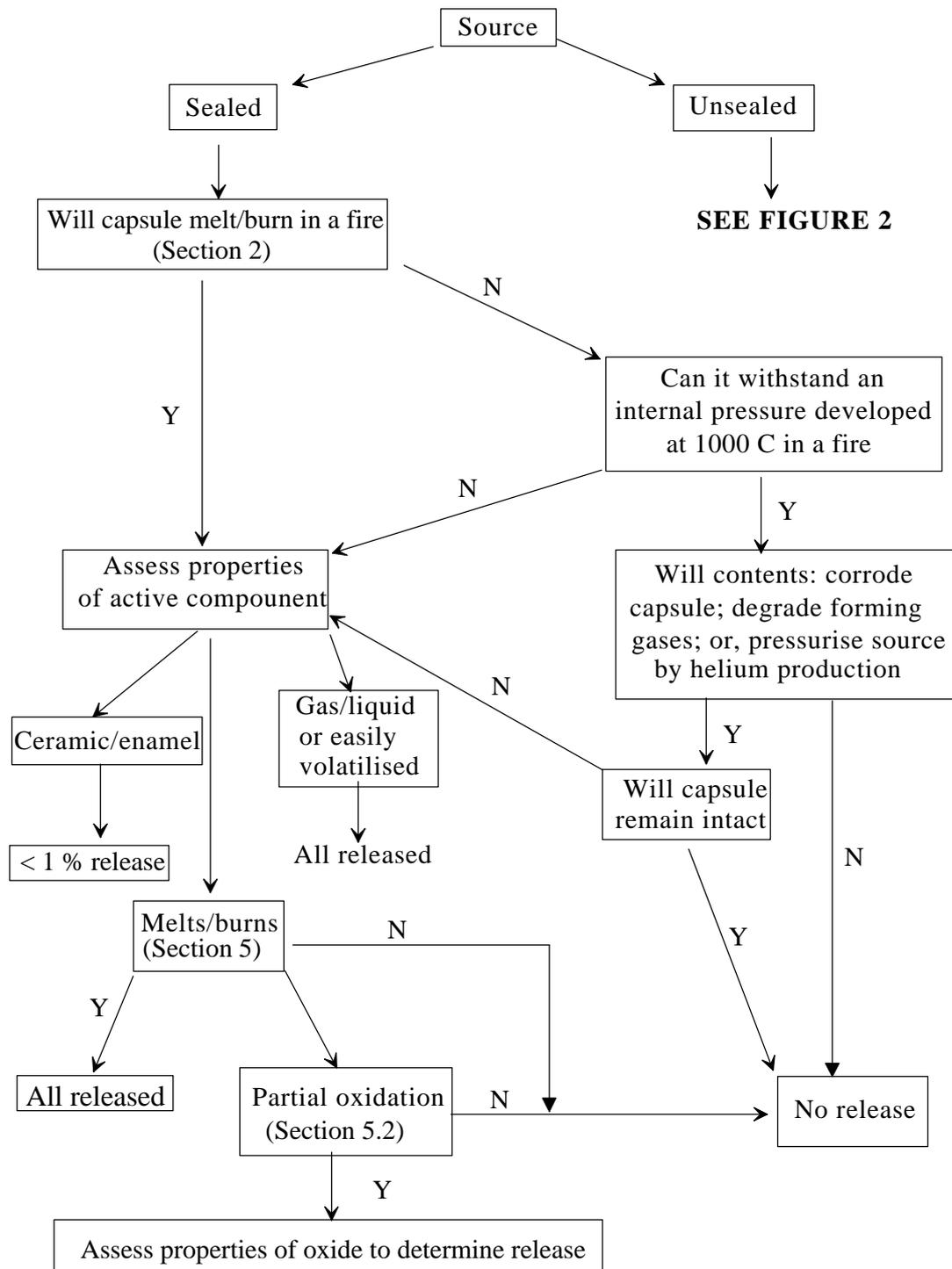
Release fractions from 'lapsed certificate' IAEA Special Form sources are also derived to allow a judgement of the applicability of the REPPPIR package to be made by organisations with such sources.

The use of active liquids, for instance in the medical field, is beyond the scope of this report, since it can be assumed that all the contents of a container will be released.

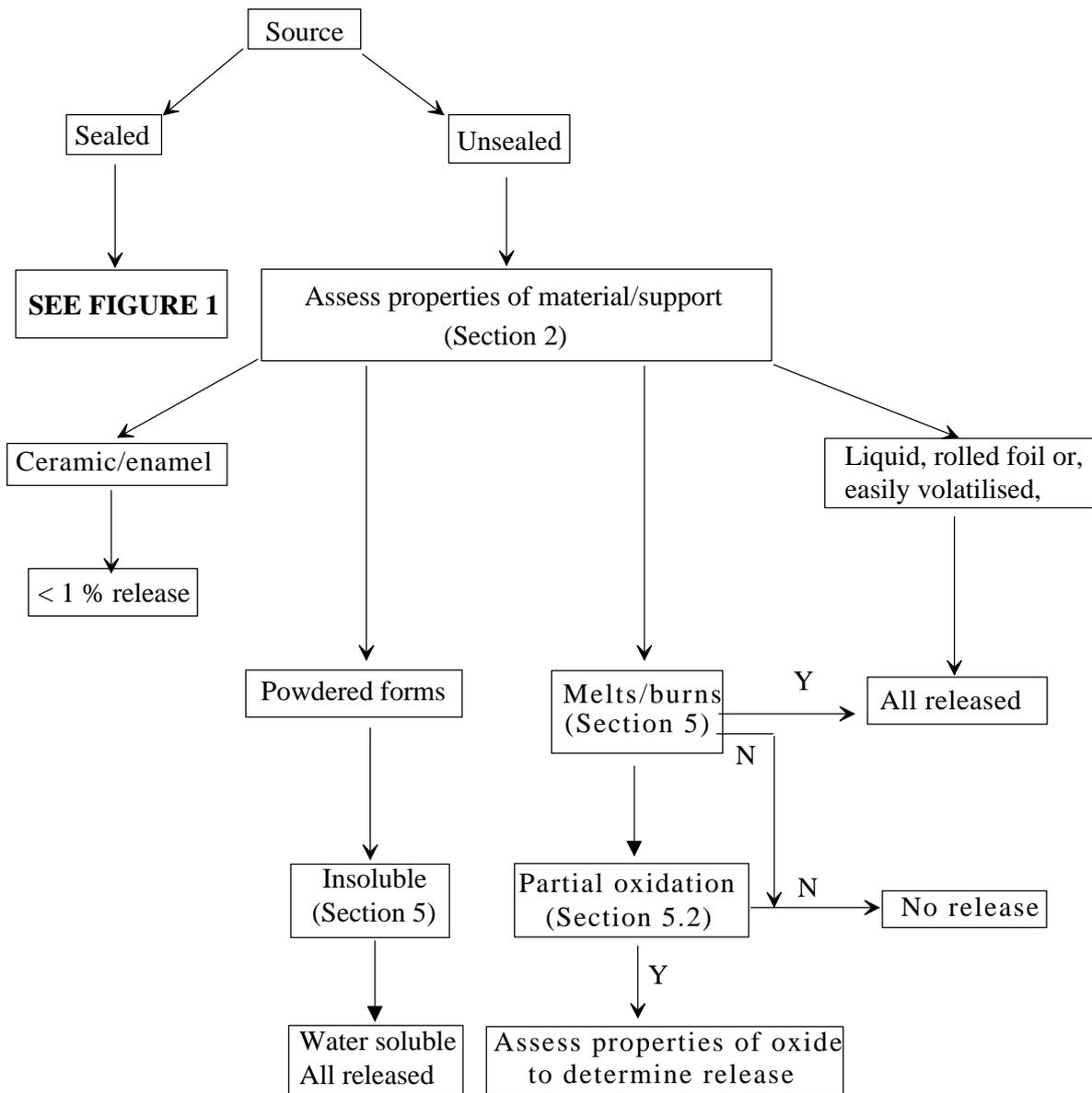
## **2. TESTS TO ASSESS SOURCE SAFETY**

### **2.1. Initial assessments**

Possible routes by which material can be released in a fire are given in Figures 1 and 2, applicable to sealed and unsealed sources respectively. The release fraction is assumed to be a function of several factors examined in detail hereafter in this report. They include the melting point and combustibility of the material forming the source, the melting and boiling point of the radioactive constituent, and the effects of internal pressurisation of a capsule amongst others.



**Figure 1**  
**Flow chart to assess likelihood of release of active materials from sealed sources**



**Figure 2**  
**Flow chart to assess likelihood of release of active materials from unsealed sources**

## 2.2. Standard tests to assess sealed source safety

### 2.2.1 Sources for commercial use

## ISO 2919

All sealed sources for commercial use are subject to an ANSI/ISO classification scheme to ensure they are safe for use and can withstand the environment in which they will be used, (ISO 2919, equivalent to BS5288: 1976). The tests to which sources are subjected are listed in Table 1.

**Table 1**  
**Classification of sealed source performance standards**

Test	Class					
	1	2	3	4	5	6
<b>Temperature</b>	No test	-40°C (20 min) +80°C (1h)	-40°C (20 min) +80°C (1h)	-40°C (20 min) +400°C (1h) and thermal shock 400 to 20 °C	-40°C (20 min) +60°C (1h) and thermal shock 600 to 20 °C	-40°C (20 min) +800°C (1h) and thermal shock 800 to 20 °C
<b>External pressure</b>	No test	25 KPa absolute to atmospheric pressure	25 KPa absolute to 2 MPa absolute	25 KPa absolute to 7 MPa absolute	25 KPa absolute to 70 MPa absolute	25 KPa absolute to 170 MPa absolute
<b>Impact</b>	No test	50 g from 1 m or equivalent imparted energy	200 g from 1 m or equivalent imparted energy	2 kg from 1 m or equivalent imparted energy	5 kg from 1 m or equivalent imparted energy	20 kg from 1m or equivalent imparted energy
<b>Vibration</b>	No test	3 times 10 min 25 Hz to 500 Hz at 49 m/s <sup>2</sup> (5 g <sub>n</sub> peak acceleration amplitude)	3 times 10 min, 25 Hz to 50Hz 49 m/s <sup>2</sup> (5 g <sub>n</sub> ) and 50 Hz to 90 Hz at 0.635 mm amplitude peak to peak and 90 Hz to 500 Hz at 98 m/s <sup>2</sup> (10g <sub>n</sub> )	3 times 30 min, 25 Hz to 80 Hz at 1.5 mm amplitude peak to peak and 80 Hz to 2000 Hz at 196 m/s <sup>2</sup> (20 g <sub>n</sub> )	Not used	Not used
<b>Puncture</b>	No test	1 g from 1 m or equivalent imparted energy	10 g from 1 m or equivalent imparted energy	50 g from 1 m or equivalent imparted energy	300 g from 1m or equivalent imparted energy	1 kg from 1 m or equivalent imparted energy

Each test can be applied in several degrees of severity, with the test results expressed as a five digit code denoting the tests applied. These figures are preceded by the letter C or E to show if the source activity was less or greater than certain limits for the test. These limits depend on the toxicity, solubility and reactivity of the active source component.

A C designation indicates that the activity level did not exceed the prescribed limit and E that the limit was exceeded during a test.

A typical code may, therefore, be C 64444 denoting that the capsule had been subjected to a maximum temperature of 800 °C, a maximum external pressure of 7 MPa, the impact of a 2 kg weight dropped from 1 m, etc..

From the point of view of fire engulfment the most important of the tests in Table 1 are those for temperature, impact, and puncture, with the ability to withstand elevated temperatures being the primary factor, and the latter two providing an indication of mechanical robustness of the item. Thus the source ISO classification number can be used as a screening tool to give a preliminary assessment of the likelihood of release of active material during a fire. It would be sensible therefore for any item tested at a low temperature or at a low impact loading to be subject of further screening to establish both the properties of materials used in manufacture to assess the possibility of failure and subsequent content dispersal.

The chemical/physical properties of common active materials and materials used to manufacture sealed sources are examined in the remainder of this report.

**UN Transport Classification Testing**

Packing for radioactive sources, such as radiography containers manufactured from depleted uranium, are subject to testing under the UN Transport regulations. Package testing includes a number of tests dependant upon the nature of the packaging. Those applicable to radiography containers manufactured using (non-fissile) uranium are:

- a 9 m drop test onto an unyielding surface for the worst case impact angle;
- penetration test involving a drop onto a steel spigot; and,
- an 800 °C fire test.

The fire test involves subjecting the package to a controlled fire where the test package must be completely engulfed in flame at a temperature of 800 °C for a minimum of 30 minutes. After this period the fire is allowed to burn out and the package removed for testing to measure surface contamination and loss of contained activity. An increase in surface activity of 20 % is allowable along with an overall loss of activity of  $10^{-6} A_2$  per hour. Where  $A_2$  is a quantity defined in the Transport Classification regs. dependant upon the isotope involved.

**2.2.2 Sources for domestic use, e.g. smoke detectors**

Consumer goods containing radioactive sources are tested on a voluntary basis subject to procedures laid out by the NRPB. (NRPB 1992). Items covered and maximum allowable release fractions under fire attack (where appropriate) are given in Table 2 below.

**Table 2**  
**Release fractions for consumer goods containing radioactive sources**

	Maximum allowable release fraction, or assumed fraction %
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Ionisation chamber smoke detectors containing americium foils;	600 °C for 1 hr: 200 Bq 1200 °C for 1 hr: < 1 % total activity
Radioluminous time measurement instruments;	100
Tritium light sources;	100
Compasses containing gaseous tritium light sources; and,	100
Thorinated gas mantles	Quantity release varies with age: see NRPB publication

Allowable release fractions for ionisation chamber smoke detectors based on an americium oxide coated foil are given in Table 2. Two tests are performed, one simulating the detectors involvement in a domestic house fire, and the other simulating the effects of incineration arising through disposal of detectors in domestic waste.

### **3. MATERIALS USED IN SOURCE FABRICATION AND LIKELY STABILITY IN A FIRE**

Study of a range of typical sources detailed in a major radiochemical suppliers catalogue shows that sources are fabricated using a range of metals. Typical examples are listed in Table 3, along with their melting point and stability towards oxidation.

The information given in Table 3 shows that the majority of materials used have good thermal and chemical stability, and therefore should not be adversely affected by fire. Some metals such as titanium, however, are likely to be damaged by fire, and may even ignite if used as a thin window. Such behaviour will expose the source and, may lead to a significant release should the active material be in a dispersible form.

**Table 3**  
**Typical materials used as components in sealed sources and likely stability if heated by a fire**

<b>Metal</b>	<b>Melting point °C</b>	<b>Use</b>	<b>Stability</b>
Stainless steel	1427 (304 stainless)	Capsule and windows.	Good stability towards aerial oxidation at elevated temperatures.
Monel	1332 (monel K)	Capsule.	Good stability towards aerial oxidation at elevated temperatures.
Titanium	1660	Capsule and windows.	Autoignition temp for large samples 1200 °C and 250°C for powders. By inference thin windows will have an AIT of < 1000. Ignites in steam at 740 °C, and in CO <sub>2</sub> between 470-900 °C
Aluminium	< 660	Capsule and windows.	Oxidised slowly in fires, but has a low melting point.
Beryllium	1278	Windows.	Oxidation rapid above 850 °C, volatilisation of oxide significant above 800°C.
Tungsten alloys	≈ 3000	Backing material for active component in sealed source.	Very high.
Silver	962	Foils for high contact area Beta sources.	Poor in thin foils.
Nickel	1453	Tubular capsules, foils and electroplate over previously electrodeposited radionuclides.	Thin wall tubes and foils may be stable, very thin electroplate over active deposits is likely to fail.
Brazing spelter 65 Cu 35 Zn 60 Cu 40 Zn	915 900	Brazing windows in some sources.	Good but will melt at high temperatures.
Epoxy resin cement	< 200	Sealing active component in some sources.	Will melt and burn in a fire.

#### **4. INVESTIGATIONS INTO THE POSSIBILITY OF CAPSULE FAILURE AT ELEVATED TEMPERATURES**

One possible failure mode of sealed sources is excessive internal pressurisation caused by the expansion of trapped gases. Such gases may be present as:

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air trapped inside during manufacture;  
 a pressurised radioactive gas as the radiation source; or,  
 helium produced by alpha decay.

Of these, trapped air is unlikely to pose a problem as the maximum room temperature pressure will be atmospheric. Thus at an ambient temperature of 20 °C, the maximum internal pressure is likely to be ~ 1 bar. The starting pressure in the latter two cases, may, however, be up to 10 bar or greater for a pressurised source, or one which may become pressurised by helium. Due to the wide range of source activities and configurations found it is difficult to exactly quantify any pressure rise over time, especially as a fraction of the helium may be retained in the ceramic matrix and thus behave as a gas.

A number of simplified calculations have been undertaken, (ignoring deformation of the capsule under pressure), in an attempt to quantify the possible pressure rise for a source at the end of its recommended working life. For instance, a 300 mCi Am<sup>241</sup> source with an ullage space of 20 mm<sup>3</sup> may be subject to a pressure rise of around 10.9 bar after 15 years (given that all helium produced escapes and is present as a gas and that the window does not deform increasing the ullage), whilst a different source type with a higher nominal activity of 5000 mCi but with a larger ullage of 879 mm<sup>3</sup>, may only develop a pressure of 4.8 bar over a similar period. Calculations supplied by a manufacturer indicate that the pressure reached in these sources may rise to 39.5 and 17.5 bar respectively at 800 °C, (again assuming no window deformation), At worst this is still 70 % below the estimated burst pressure based on the assumption that the window does not deform and is not adversely weakened by fire attack. In practice the window could deform by between 1 - 3 mm leading to a considerable decrease in internal pressure, and hence an increased safety margin. The subject of material properties at elevated temperature is now discussed further.

Available information on changes in yield and tensile strengths with temperature for materials commonly used in source manufacture is limited, with the maximum temperature studied often being well short of that seen in a fire. The temperature ranges studied along with the initial and final tensile strengths of the metals of interest are given in Table 4.

**Table 4**  
**Effects of temperature on tensile strength of metals commonly used in the**  
**manufacture of sealed sources**

<b>Metal</b>	<b>Temperature range studied °C</b>	<b>Melting point °C</b>	<b>Tensile strength at room temp N.mm<sup>-2</sup></b>	<b>Tensile strength at maximum temperature studied N.mm<sup>-2</sup></b>
Aluminium	23 - 371.1	660	468.84	27.58

Titanium *	25 -537.8	1,660	880	468
Beryllium	25 - 800	1,278	494.13	48.26
Stainless steel (Annealed type 316)	≈ 60 - 982.2	1427 (Type 304)	620	53 (≈ 172 at 800°C)

\* Mean tensile strength of two Ti alloys used, Ti - 5 Al - 2.5 Sn annealed, and Ti - 8 Mn mill annealed.

The weakest points in a capsule are likely to be either the window, as this must be thin to maximise the radiation emitted from the source, or the weld securing the window to the body of the capsule. The calculation of failure pressure is complex, as the window cannot be treated as a simple flat plate, since it is substantially reinforced by the surrounding metal. As a result it is 'stiffer' than would be expected from a consideration of its material properties. Thus any prediction using standard properties would considerably underestimate failure pressure. A more detailed approach could be to start from a flat plate solution, but with a modified tensile strength to allow for the increased window stiffness, and with a series of iterative calculations where both internal pressure and window deformation are calculated with temperature.

Such a detailed approach has not been used here due to the large range of possible source configurations. Instead it is proposed that the possibility of source failure can be approximately estimated from the following:

- the limited material data available;
- the melting point of materials used in source construction;
- the ISO classification number for the source; and,
- a knowledge of the material propensity to burn/oxidise in a fire.

Unfortunately the temperature range for which material property data is available is very limited for many of the metals commonly used in source manufacture, as seen from Table 4. However, from Table 4 it can be seen that 316 Stainless still retains a significant percentage of its original tensile strength even when heated to nearly 1000 °C.

On this basis the likelihood of the metals in Table 5 failing when heated to 800 - 1000 °C in a fire has been estimated qualitatively and given in Table 5.

**Table 5**  
**Predicted survivability of sealed sources in fires**

Window/capsule material	Oxidised in fire or likely to melt	Possibility of failure
316 stainless steel	No	Low if passed ISO temp. test 6
Titanium	Yes especially when thin	May fail if thin titanium window used, or carbon / carbonates used inside due to Ti igniting in CO <sub>2</sub> atmosphere
Beryllium	Significant oxidation above 800 °C	May fail if used as a thin window

Aluminium	Melts	Fails
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Also it would be conservative to propose that any source not tested to Class 6, (800 °C), should be regarded as likely to fail, irrespective of the material of manufacture.

## 5. RELEASE FRACTIONS FOR ACTIVE MATERIALS WITH DIFFERENT PHYSICAL FORMS

There are many hundreds of individual isotopes, of which a considerable number have no commercial application. Those used are found as metals, compounds, gases, or as ceramics /enamels. Typical examples are listed in Table 6.

**Table 6**  
**Examples of typical isotopes, their physical form and stability**

Source material	Physical form	Stability on heating	Long-term stability
Iridium	Metallic wires	Oxidised slowly at red heat	Excellent
Gadolinium oxide	Pressed pellets	Stable	Excellent
Calcium sulphate	Pressed pellet	Melts at 1010 °C	Good
Caesium chloride or sulphate	Pressed pellets	Good, but non-alkali metal sulphates form SO <sub>x</sub> on heating	Good
Promethium carbonate	Crystalline powder	Will decompose at high temps. liberating CO <sub>2</sub>	Good
Cobalt	Metal pellets	Good	Good
Americium	Ceramic	Excellent	Good stability for ceramic, but Am suffers from long term helium production
Strontium	Ceramic	Excellent	Excellent
Copper	Ceramic	Excellent	Excellent
Iodine	Absorbed on charcoal	Charcoal substrate will react with oxygen in source forming CO and CO <sub>2</sub>	Excellent

This table shows that a number of materials possess very good thermal and long term stability. However, it also highlights difficulties in a number of areas:

- the use of metallic salts which may decompose liberating gases on heating;
- the use of possibly corrosive metal salts, and,
- the previously mentioned helium production by all alpha emitters.

The first two of these factors are covered in Section 5.3, and the third, that of helium production, has already been discussed in Section 4.

## 5.1 **Ceramics/enamels**

Ceramics used in source manufacture are produced by heating a mixture of a refractory material and the radioactive constituent in an electrical or radio frequency oven. The exact composition of the ceramic is varied depending on the nature of the source radiation, but it usually contains low atomic mass elements such as beryllium or silicon.

Sources are often manufactured in two stages. Firstly the ceramic mixture is heated to its melting point to produce beads. The activity of these beads is determined and they are then either used as a discrete source, or a number, depending on the required activity, are placed in a die and re-melted producing a pellet of the desired dimensions.

Typically, the ceramic will remain solid to around 700<sup>o</sup> C, will become soft / tacky at around 1000<sup>o</sup>C, and will melt at about 1400<sup>o</sup>C. Discussions with a source manufacturer suggest that possible losses of the radioactive constituent during manufacture would be very slight, and may well be due to loss of the active component before incorporation into the ceramic matrix. Thus the release fraction from a heated ceramic product in a fire can be regarded as being very low, probably < 1 %.

## 5.2. **Metals**

Releases from metallic sources will only be possible if they are unencapsulated, or if the capsule fails exposing the active constituent within. Should a metal be exposed the rate of oxidation will be a function of:

- chemical reactivity;
- oxygen concentration; and,
- thickness/surface area of metal.

Most metals will be subject to some degree of oxidation when heated in a fire and a small number, in particular the so-called light metals, will burn even in supposedly inert carbon dioxide or nitrogen atmospheres. Due to the high thermal conductivity of metals, the most likely forms to support combustion are thin sections or high surface area forms such as foils, wires or powders. Foils and wires are often used as unencapsulated sources. The variation in ignition temperature with surface area is discussed in Section 5.2.3 which gives possible releases from metallic uranium.

Any powdered metals should be regarded as flammable or potentially explosive and assigned a release fraction of 100 % if the capsule could fail. Larger samples (> 1-2 mm thick) must be heated to their melting point for ignition to occur, unless the metal is subject to slow oxidation causing self-heating or if water is present, when highly exothermic reactions can occur causing ignition at much lower temperatures.

As well as these forms, it is also common to electroplate thin layers of an active material onto a supporting substrate. In this case the mass of the support is such that the entire item would

behave as a comparatively thick section, from the point of view of ignition, but surface oxidation would lead to a high release of activity.

An indication of the properties of metals used in the manufacture of sealed sources has been given in Table 3.

### **5.2.1. *Flammable metals***

As stated above any powdered metal should be regarded as flammable or potentially explosive and a release fraction of 100 % assigned.

Those metals known to be flammable are indicated in Table 7, along with possible release fractions.

**Table 7**  
**Indicative release fractions for potentially flammable/readily oxidised metals**

<b>Metal</b>	<b>Form (foil, wire pellet etc.)</b>	<b>Release fraction / %</b>
Alkali metals	Any form	100
Alkaline Earths	Foils, wires, powder	100
Magnesium - radium	Thicker samples	25
Titanium, hafnium, zirconium	Foils, wires, powder	100
	Pellets/thicker samples	25
Zinc	Any form	100
Aluminium	Foils, wires, powder	100
	Thicker samples	< 5
Bismuth	Any form	100
Lanthanides	Any form	100
Uranium	Large samples	5
	Pellets/thicker samples	10
	Foils, wires, powder	100
Other Actinides	Review properties for individual metals	

The derivation of the release fractions given in Table 7 is complex and is a function of many factors.

The propensity of the alkali metals to burn is well known and will not be discussed, a release fraction of 100 % is therefore fully justified.

The reactivity of the Alkaline Earth or Group II elements increases as the group is descended. Magnesium is moderately stable in air and subject to slow oxidation. The bulk metal will not react with water, whether hot or cold, but it will react vigorously with steam. Barium, is far more reactive, ignites spontaneously in air and reacts violently or explosively with water. Radium can be expected to be even more reactive. This again justifies the choice of a unit release fraction for large surface area or thin form of the material.

The transition metals, titanium, hafnium and zirconium are all potentially flammable, especially when finely divided, or in thin sections. The release fraction for combustion of powders or thin sections can again be set as 100 %, as much of the oxide is likely to be dispersed in the fire plume. For thicker sections, a more mechanically robust oxide coating will form and a smaller proportion of the oxide will become airborne.

Zinc burns readily in air when heated and boils at 908 °C. Metal turnings burn when heated in a flame, and thicker sections burn if heated more strongly in a crucible, forming a white oxide cloud which settles out in the form of woolly flocks, (Partington, 1950). Thus the combination of a boiling point comparable with temperatures typically found in a fire and the strong convective flow would result in a large release fraction.

Aluminium is a reactive metal, but is normally protected by a tenacious oxide coating. The metal powder has been the cause of many explosions and thin foils and wires burn/oxidise forming an oxide/metal mixture which will be readily become airborne in a fire plume. A fire will melt or partially oxidise thicker sections and it is likely that some of this oxide will become airborne, however, the release fraction is likely to be low.

The comparatively low melting point of bismuth of 271 °C, combined with its reactivity towards air (flammable when exposed to flame, Sax and Lewis 1989), again suggests that a high release fraction is appropriate for this metal.

The lanthanides react slowly with cold water, tarnish in air and burn easily forming oxides. Thus it must be assumed that should a metallic lanthanide be used and exposed to air in a fire it will all be lost to atmosphere.

### **5.2.2 Metals susceptible to oxidation**

Many metals are partially oxidised when heated in a fire. The extent of oxidation and nature of the oxide for 'non-flammable' metals is given in Figure 3, along with a possible release fraction assuming the metal is in an unencapsulated form.

### **5.2.3. Depleted uranium**

Commercial applications of depleted uranium arise through its high density, viz:

armour piercing munitions; and,  
radiation shielding in portable radiography containers.

It has also found use as counterweights / ballast in some aircraft by virtue of its extreme density.

The first of these groups, munitions, are not covered by this report.

Metallic uranium may be found either in a bare unencapsulated form, or in an encapsulated form such as a radiography container. The properties of the bare metal are given below.

The variation in ignition temperature of uranium with surface area is evident in the following example (Baker et al. 1966):

	Ignition temperature °C
10 x10 x10 mm cube	700
8.5 x 8.5 x 8.5 mm cube	650
0.47 mm dia. wire	475
bundled wires	~275
foil	350-390
stacked foils	290

It is evident from the above that thicker sections require stronger heating to ignite, but are still be susceptible to surface oxidation on exposed surfaces at rates up to a maximum of 5 g.cm<sup>-2</sup>hr<sup>-1</sup> (A Thyer, private correspondence). The oxide coating thus formed is semi-stable giving about a 5 % release as respirable/dispersible material in the fire plume for large samples. Release fractions for articles with a range of sizes are given in Section 5.2.1, Table 7. To minimise such releases, articles are usually plated with a non-combustible metal, or used in a steel or otherwise fire resistant container, as is the case with radiography containers. Where such treatments are applied the release fractions given in Table 7 can be

reduced where it is proven that the treatment is effective, for instance through examination of test UN Transport Classification test data.

As detailed earlier in Section 2.2.1. radiography containers manufactured from depleted uranium are subject to testing under the UN Transport regulations. Tests applied to radiography containers manufactured using (non-fissile) uranium are:

a 9 m drop test onto an unyielding surface for the worst case impact angle;

penetration test involving a drop onto a steel spigot; and,

an 800 °C fire test.

The fire test involves subjecting the package to a controlled fire where the test package must be completely engulfed in flame at a temperature of 800 °C for a minimum of 30 minutes. After this period the fire is allowed to burn out and the package removed for testing to measure surface contamination and loss of contained activity. An increase in surface activity of 20 % is allowable along with an overall loss of activity of  $10^{-6} A_2$  per hour. Where  $A_2$  is a quantity defined in the Transport Classification regs. dependant upon the isotope involved.

										<i>Al</i>			
Sc	<b>Ti</b>	V	Cr	Mn	Fe	Co	Ni	Cu	Zn Boils at 908 °C 100 % release	Ga Melts at 30 °C 100 % release	Ge Melts at 937 °C		
Y	<b>Zr</b>	Nb	Mo	Tc	Ru	<b>Rh</b>	<b>Pd</b>	Ag	Cd Boils at 767 °C 100 % release	In Melts at 156 °C. 100 % release	Sn Melts at 232 °C	Sb Melts at 630 °C	
<b>La</b>	<b>Hf</b>	Ta	W	Re	Os Volatile oxide, release 100 %	<b>Ir</b>	<b>Pt</b>	<b>Au</b>	Hg Boils at 357 °C, 100 % release	Tl Melts at 303 °C	Pb Melts at 327 °C	<b>Bi</b>	Po Boils at 962 °C 100 % release

**Key**

Metals in Bold Italicised text will burn readily or on strong heating, see Table 5 for release fractions.

	Some oxidation but stable coating - low release		Slight oxidation low release		Unaffected by fire or very slight oxidation - no release
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**Figure 3**

**Metal oxide formation and release fractions for metals heated to 1000 °C in fires**

### 5.3. Inorganic chemicals in sealed sources

Many sulphates and carbonates have a relatively poor thermal stability and breakdown on heating forming CO<sub>2</sub> or oxides of sulphur. This process could potentially produce a significant pressure rise in a source far in excess of that expected from simple thermal expansion of trapped air.

Calculation of the volume production of these gases is complex, as any reaction would occur in a capsule of fixed volume, and any increase in pressure would tend to stop dissociation at an equilibrium pressure. For instance, 1 g of calcium carbonate could theoretically liberate 280 cm<sup>3</sup> of CO<sub>2</sub>. However, any decomposition in a sealed vessel would result in a dissociation equilibrium level well below this. Table 8 illustrates the temperatures at which the Group II metal carbonates reach a dissociation pressure of 1 atmosphere CO<sub>2</sub> (Greenwood and Earnshaw, 1986).

**Table 8**  
**Temperatures at which Group II metal carbonates reach a dissociation pressure of 1 atmosphere of CO<sub>2</sub>**

Carbonate	Temperature °C
Beryllium BeCO <sub>3</sub>	250
Magnesium MgCO <sub>3</sub>	540
Calcium CaCO <sub>3</sub>	900
Strontium SrCO <sub>3</sub>	1,289
Barium BaCO <sub>3</sub>	1,360

Table 8 indicates that the extent of dissociation decreases down the group, with calcium carbonate producing 1 atmosphere pressure at 900 °C and barium carbonate at 1360 °C. Such a level of additional pressurisation is unlikely to be problematical for the source designs considered here. No information could be found listing the dissociation constants for the lighter elements, beryllium and magnesium, and thus it is impossible to predict the source pressure developed at typical fire temperatures for these metals.

There are two further possible ignition hazard with some metal carbonates:

an incandescent reaction has been reported between titanium and fused alkali-metal carbonates, (Bretherick, 1990); and,

titanium itself will burn in a carbon dioxide atmosphere.

The ignition temperature of titanium in CO<sub>2</sub> is dependent on particle size/sample thickness, with published figures ranging from 470 to 900 °C, (Jacobson et al., 1964). Thus it is assumed

that a 100 % release fraction is applicable should alkali metal carbonates be used in sources with titanium windows.

Sulphates of the Group I and II elements are stable on heating except at very high temperatures. The majority of other metal sulphates dissociate on heating, forming the metal oxide, SO<sub>2</sub> or SO<sub>3</sub>, and oxygen. As for carbonates, the degree of dissociation in a sealed vessel is dependent on the individual equilibrium constant for the element. Thus it is likely that metal sulphates will behave in a similar manner to metal carbonates, where reactivity increases down a group.

The final area of possible concern is the use of potentially corrosive inorganic salts such as caesium chloride. Many molten salts are highly corrosive and could weaken thin metal sections to such a degree that they could fail, especially if combined with internal pressurisation and external oxidation of the window or other thin section by fire. In this event much of the active material would be released, when the source failed, and there is a strong possibility of the remainder escaping should firewater be applied to the source during fire fighting.

#### 5.4 Inorganic chemicals in unsealed sources

A 100 % release fraction should be assigned to any water soluble radiochemical used in an unsealed source, as packaging (bottles etc.,) will fail in a fire releasing the material enabling it to be dissolved in water applied for fire fighting. More robust packing should be assessed on an individual basis to determine its survivability in a fire, and if it is found suitable a nil release assigned.

Non water soluble chemicals should also be assigned a 100 % release if packaging will fail in a fire, as whilst the chemical may not dissolve, a large portion will be carried into drainage systems by fire water.

Airborne releases into the fire plume will also occur with both the above cases to varying extent depending on the nature of substrate chemicals are supported upon. Table 9, lists maximum published airborne release fractions for radioactive (or simulated radioactive) materials from fires involving contaminated materials, (Quintiere 1998)

**Table 9**  
**Maximum airborne release fractions for contaminated materials**  
**(Quintiere 1998)**

Fuel type	Total release fraction	Release on filter system	Contaminant/fuel	Reference
Cellulosics, cotton	0.18 - 0.55	0.10 - 0.38	U oxide or nitrate on thin materials	Mishima & Schwendiman
Perspex (PMMA)	0.7	0.02 - 0.06	Ce or Eu on Perspex, 600 kW pool fire	Buijs et al.
Kerosene	0.2 - 0.9	10 <sup>-4</sup> - 10 <sup>-3</sup>	Ce in kerosene	Malet et al.

Two release fractions are quoted above. A total release based on the original inventory, and a downstream release measured on filter systems on the fire test chamber. This later figure is lower as a substantial amount of activity can plate-out on walls/surfaces in the test chambers.

The airborne fraction from a bulk, rather than contaminated material as above, is dependent on a number of factors including:

- the chemical involved;
- method of containment;
- the height the container falls from;
- the particle size distribution;
- powder density; and,
- the local air velocity.

Due to the complexity of the issue a generic release fraction cannot be assigned and each case should be considered individually, or a worst case 100 % release assigned.

## **6. CONCLUSIONS AND RECOMMENDATIONS**

This report proposes a number of possible release fractions for radioactive materials exposed to fires. Two flow charts have been developed to help aid the user in the identification of relevant issues, and hence possible release fractions. The approach taken is first to establish the possibility of a sealed source retaining its integrity in a fire, and should the source fail, the stability of the active material is examined to determine the extent of any release. A similar approach is taken for unsealed sources.

Three methods have been identified to assess possible release from sources:

- Study of UN Transport Classification test details where available;
- use of ANSI/ISO classification details in conjunction with chemical/physical properties identified in this report: and,
- study of NRPB test data for consumer goods containing radioactive sources.

Other findings are as follows:

Most sealed sources which have been subjected to ISO temperature test class 6 (800 °C for 1 hour) can be regarded as being unlikely to fail when heated in a fire, unless the materials used in fabrication of the source are likely to be oxidised or melt in a fire.

The long term stability of many sources appears very good. Care is needed, however, when determining the possibility of extending the working life of time-expired alpha emitting sources, as it is possible that, given too long an extension the internal pressure may rise to a level sufficient to cause source to failure in a fire.

Discussions with a major source manufacturer suggest that should a capsule fail, the release fraction for a ceramic/enamel source is likely to be <1 %.

Due to a scarcity of data on the fire performance of many of the materials used in source manufacture, it has not been possible to identify release fractions for a large number of radioisotopes. However, this may not be too great a problem as have no commercial use. In the absence of reliable numbers, indicative levels of oxidation and hence possible release fraction have been identified for most metals as follows:

flammable metals - 100 % release for thin/small sections or powders;

metals which will melt/boil in a fire - 100 % release; and,

oxidisable metals - varying release depending on properties of oxides.

In the latter case, for metals oxidised in a fire following containment failure, it has not been possible to determine a release fraction. This is due to the lack of data on the extent to which metals are oxidised in a fire, the rate of oxidation, and the tenacity of the oxide coating thus formed. It is recommended that, should release fractions be required for such cases, further work be undertaken to either perform a more detailed literature review, or carry out a number of tests to attempt to measure the response of heated samples.

It is also recommended that, should reliable figures be required to establish the possibility of source failure, fire tests should be undertaken on a range of dummy source types, for example, titanium or beryllium windowed sources.

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