Advanced, Second-Generation Selenium-75 Gamma Radiography Sources.

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Introduction

Gamma radiography using Selenium-75 is now generally acknowledged throughout the world to provide performance benefits relative to Iridium-192 in the working range of 5-30mm steel. Selenium-75 has a softer gamma ray spectrum than Iridium-192 and it has a significantly longer half-life. For these reasons Selenium-75 provides real performance benefits and working life advantages. The general features and benefits of these sources and their performance relative to other techniques has been thoroughly researched and published elsewhere [1,2].

Gamma radiography requires the use of compact, robust, high activity sources with small focal dimensions. Early Selenium-75 source designs were too large to fit many source holders and projectors commonly used by the industry and they failed to optimize the focal geometry. This is because sources contained Selenium-75 in elemental form. This can melt during irradiation and move around inside the capsule; it can even corrode the capsule inside walls at high temperature, potentially with adverse consequences on the focal geometry and the source integrity. The use of elemental selenium in radiography sources has given rise to some safety concerns because of its high volatility and chemical reactivity [4]. As a result, not all countries permit the use of elemental selenium sources for gamma radiography applications.

In this paper, the design of an advanced second-generation source is described. This has been introduced to mitigate safety concerns over the use of elemental Selenium and to provide better control and definition of the focal spot geometry. The advanced design uses highly enriched selenium-74 in conjunction with high flux irradiations, but it replaces elemental Selenium with a thermally stable metal-selenide compound in a quasi-spherical focal geometry. The source is special form approved (pending at the time of writing), USNRC registered and is fully compatible with projectors, cameras and crawler devices commonly used by the industry. As more and more users throughout the world gain experience of Selenium-75, its advantages are becoming more widely appreciated. Consequently, commercial demand for sources is steadily increasing. Selenium-75 is becoming the nuclide of choice in mid-thickness gamma radiography applications requiring high image quality.

Physical & Chemical Properties of Elemental Selenium

Elemental selenium is highly toxic, volatile, reactive and corrosive. It comes in three physical forms: amorphous, monoclinic and metallic with densities of 4.3g/cc, 4.5g/cc and 4.8g/cc respectively and it has a very large coefficient of expansion close to its melting point of 217^{0}C. This is shown in Figure 1. Early source

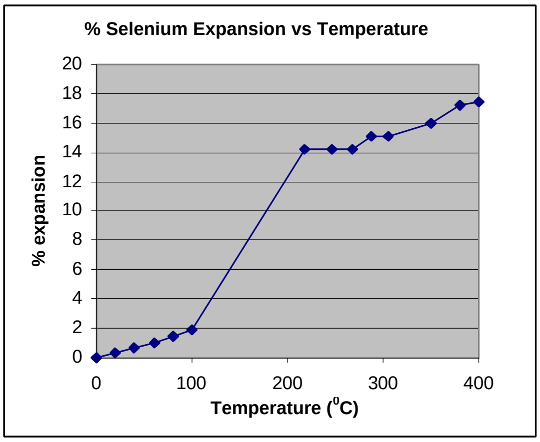


Figure 1
Selenium expansion vs temperature
after Chizhikov et al[3]

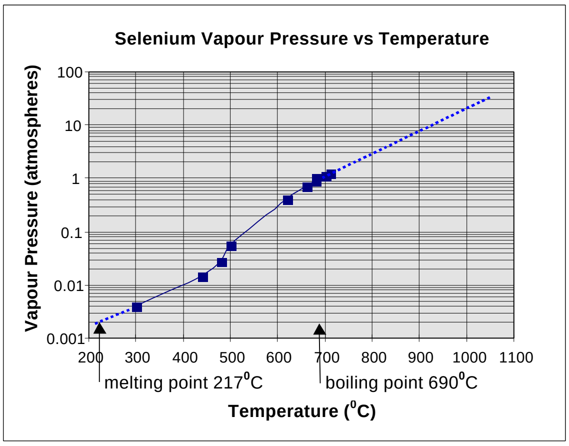


Figure 2
Selenium vapor pressure vs temperature
after Chizhikov et al[3]

designs containing elemental Selenium had to contain some void space inside the capsule to allow for this expansion during reactor irradiation when the Selenium melted. The Selenium vapour pressure increases rapidly with temperature as shown in figure 2. This exceeds atmospheric pressure above the boiling point of 690^{0}C and at 800^{0}C (the temperature of the special-form capsule integrity test) the pressure is about 3 atmospheres. At higher temperatures, the internal pressure continues to rise as extrapolated in the figure.

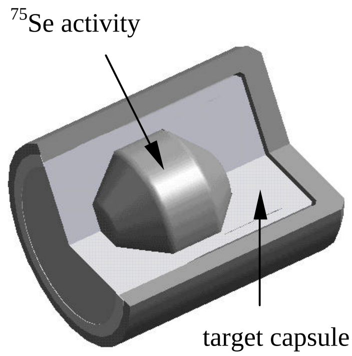
During the development of the advanced, second-generation design, the corrosion reaction between Selenium and Vanadium or Titanium was found to occur above about 400-450^{0}C. Corrosion compounds of the following type were formed: $xM + ySe \rightarrow M_xSe_y$ where M=Ti or V.

Source Design

The source is referred to as the ***$^{75}Se^{ntinel}$*** source and has been designed by Sentinel^{TM} to meet the needs for a more durable, thermally stable, high performance product, in response to market needs. A thermally stable compound of a non-activating metal combined with selenium is used in the capsule. The compound is processed into compact, dense, pellets, which do not melt or interact with the capsule at high temperature. The compound mitigates the risks of corrosion and melting during activation in the reactor and avoids internal pressurisation and potential reactions with the capsule under accident conditions involving high temperatures.

The focal geometry is quasi-spherical and is unique to the ***$^{75}Se^{ntinel}$*** design. The engineered cavity in the capsule determines the shape of the focal geometry. This is octagonal in cross section (in side view) and circular in end view as shown in figure 3. The target capsule has two closure welds for added integrity. The capsule can be made available in either titanium or vanadium.

The ***$^{75}Se^{ntinel}$*** product range provides four activity and focal sizes as shown in Figure 3. The focal dimension is significantly smaller, Curie for Curie, than is achieved using a conventional cylindrical design. The focal dimension is the diagonal of the octagonal cross-section in side view. This is 1.082 times the pellet diameter and is much smaller than the diagonal of a right cylinder of the same diameter, which is 1.414 times its diameter. The ***$^{75}Se^{ntinel}$*** focal dimension is therefore 30% smaller than a cylindrical source of the same diameter. This advantage enables ***$^{75}Se^{ntinel}$*** sources to be used in close proximity shots, in pipeline crawler applications and in contact radiography, with the minimum geometrical unsharpness in the image.



Pellet (diameter)	**Focal Dimension (diagonal)**	**Activity (Curie)**
2.00mm	**2.2 mm**	**~20Ci**
2.50mm	**2.7 mm**	**~40Ci**
2.85mm	**3.1 mm**	**~60Ci**
3.25mm	**3.5 mm**	**~80Ci**

Figure 3
Selenium-75 target capsule

The use of ***$^{75}Se^{ntinel}$*** pellet geometry enables the source to be used up to 30% closer in radiography applications relative to a cylinder of the same diameter and this increases the exposure rate per Curie at the object. Exposure rate is inversely related to the square of the source-to-object distance, so a 30% reduction in focal dimension enables the exposure rate at the object to be increased by up to 70%. Figure 4 compares ***$^{75}Se^{ntinel}$*** exposure rate at an object with exposure rate from a 3mm x 3mm cylinder. In Figure 4 all sources are positioned so that they produce equal geometrical unsharpness in the image. Exposure rates are listed in the figure. The exposure values for all the ***$^{75}Se^{ntinel}$*** designs compare very favourably with the largest size source; the 3mm x 3mm, 80Ci cylinder.

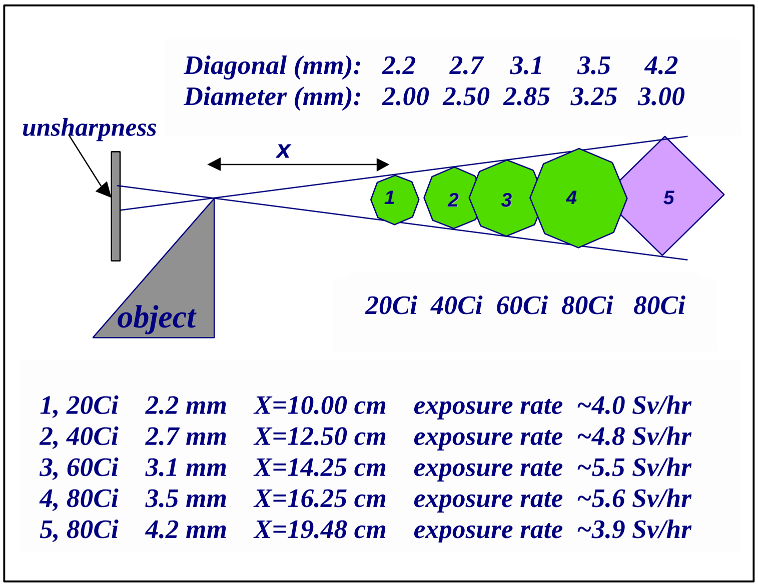


Figure 4, Focal Dimension vs Exposure Rate

The target capsule shown in Figure 3 is encapsulated in a secondary Titanium capsule. This is the outer capsule for the source and is special form approved (pending at the time of writing). The complete capsule assembly is represented in figure 5. The external dimensions of the assembly are 5.15mm diameter x 7.65mm long. This is small and compact, significantly smaller than cylindrical pellet designs, which are currently on the market. The source is robust and durable, meeting all the industry and regulatory requirements for use in source holders, projectors, cameras and crawler devices commonly used by the industry.

The source may be used in conventional projector devices such as the Sentinel 460 and 660 units and the new 880 unit. When used with these projectors, the source is further encapsulated in a stainless steel pigtail assembly as shown in figure 5. In this configuration sources are encapsulated with either three or four closure welds.

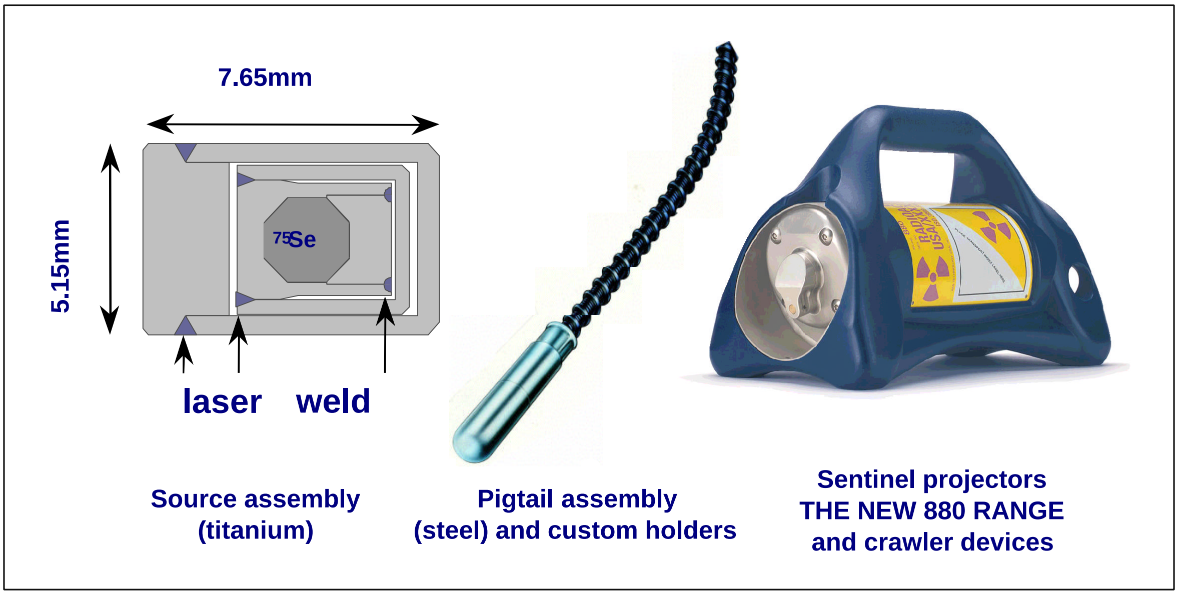


Figure 5
$^{75}Se^{ntinel}$ source assembly and pigtail

Gamma Ray Emissions

MonteCarlo calculations have been carried out using the AEA Technology RANKERN and MCBEND codes to determine exposure rates and self-attenuation of gamma ray emissions from source capsules. The results are summarized in Tables 1 and 2. Attenuation in the capsule removes most of the low energy X-ray emissions below about 20keV. In the energy range above 100keV the gamma ray spectrum is only slightly hardened due to the preferred absorption of lower energy gamma rays. The corrected spectra and gamma ray exposure rates for ^{75}***Se***^{ntinel} sources at 1 meter for a 1Ci and a 1GBq source are summarized in the tables.

Table 1
Abundance of Principal Gamma Emissions of ^{75}Se through capsules.

Principal Gamma Emission (keV)	No capsule Abundance %	^{75}*Se*^{ntinel} source Abundance %	^{75}*Se*^{ntinel} source + pigtail Abundance %
96.7	3.41	3.49	3.76
121.1	17.15	16.47	15.76
136.0	58.82	55.23	51.44
264.6*	59.00	59.00	59.00
279.5	25.00	25.01	24.73
400.6	11.51	11.67	11.70

Capsule abundance is normalized to 59% for the 264.6 keV gamma emission.

Table 2
Gamma-Ray Exposure Rates from 1Ci and 1GBq ^{75}*Se*^{ntinel} Sources

Source	Exposure-Rate at 1m (R/h/Ci)	Exposure-Rate at 1m (mSv/h/GBq)	% Attenuation >20keV
Selenium-75 including X rays < 20keV	0.595	160.6	-
Selenium-75 gamma energies >20keV	0.201	54.3	0%
^{75}***Se***^{ntinel} source	0.186	50.2	~8.4%
^{75}***Se***^{ntinel} source + steel pigtail	0.180	48.6	~11.3%

The approximate half and tenth thickness values for various shielding materials for ^{75}***Se***^{ntinel} source capsules was calculated using the MCBEND code. These values are shown in table 3.

Table 3
Approximate ½ and $^{1}/_{10}$th Thickness Values
for Various Shielding Materials for ^{75}*Se*^{ntinel} Sources

Shielding Material	½ Thickness Value (mm)	½ Thickness Value (inches)	$^{1}/_{10}$th Thickness Value (mm)	$^{1}/_{10}$th Thickness Value (inches)
Aluminum	27	1.06	80	3.15
Steel	8	0.315	27.5	1.08
Lead	1	0.0394	4.75	0.187
Tungsten	0.8	0.0315	4.0	0.157
Concrete	30	1.18	90	3.54

Applications

Selenium-75 provides advantages of longer half-life, improved operator safety, smaller exclusion zone and high image quality, particularly in the 5-30mm steel working range. The advantage of the longer half-life reduces decay loss due to transportation and minimizes the costs associated with managing frequent source exchanges of other isotopes. This is particularly beneficial when sources are used in remote geographical regions and most particularly in offshore applications. Safety compliance, operator dose and storage logistics are made easier using Selenium-75. The operational exclusion zone for Selenium-75 is reduced relative to that of Iridium-192 due to the softer emission spectrum and lower gamma ray constant. Sources can be used in more restricted areas, affording considerable operational and safety advantages. The half-life of Selenium-75 is 120 days; this is almost double that of Iridium-192 and four times Ytterbium-169.

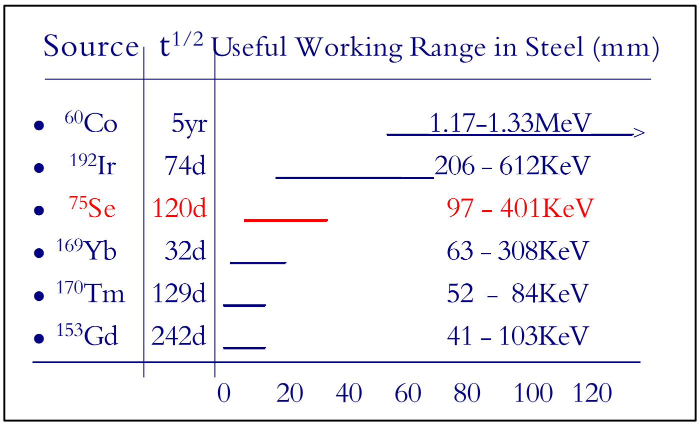


Figure 6
Useful working range of Selenium-75

Selenium-75 has been particularly successful in replacing X-ray devices in some pipeline crawler applications. Operational trials using $^{75}Se^{ntinel}$ sources have been carried out by several pipeline inspection companies. Solus Schall, a division of Oceaneering International in association with Stolt Offshore Ltd have undertaken the first operational trials of Selenium-75 in the "North Sea". They have played an important role in developing and verifying the performance benefits of $^{75}Se^{ntinel}$ sources in offshore pipeline inspection applications.

SOLUS SCHALL ^{75}Se PIPELINE DEVELOPMENT & APPLICATIONS

Solus Schall are a global forward-looking company and were eager to work with Sentinel^{TM} to develop applications for the second-generation $^{75}Se^{ntinel}$ design. They were engaged in discussions from a very early stage in the $^{75}Se^{ntinel}$ product development and the project benefited enormously from their experience and knowledge of what was required by their clients for a new product to push forward the boundaries set by existing pipeline radiographic techniques.

The initial discussions while still at the theoretical stage, lead Solus Schall to believe that substantial benefits could be obtained in the 2"- 8" pipeline diameter range. With this in mind Solus Schall approached one of their major clients, Stolt Offshore Ltd and with their like minded forward thinking Pipelay Group it was agreed that major benefits to safety, quality, reliability and cycle times could be possible and they agreed to take an active part in trials.

Once the trial source release date was known, Stolt Offshore provided butt weld samples and released their in-house specialist to plan and finalise an extensive trial program and to support the safety case to allow the first operational application of $^{75}Se^{ntinel}$ in the "North Sea" to commence. The basic scope of the trial covered safety, radiographic quality, flaw sensitivity, film processing, film and screen combinations and cycle times.

Overview of results

There was a substantial reduction in controlled areas c.f. Iridium-192 when used on 2" - 3.5" diameter pipe. It was possible to use the contact method (DWSI) with tungsten collimators to greatly reduce the beam. This contrasted with the problem of shielding directional X-ray units when using the elliptical method (DWDI) on the same diameter range. The resulting output figures enabled a portable exposure bay to be constructed within a container unit to ensure an enclosed controlled area with the inherent safety features of a permanent exposure bay installation. There were also substantial cost savings on the lead shielding required for the portable bay. The amount of lead required for the traditional Iridium-192 methods would have exceeded the weight limits for a portable unit.

Many different film types and screen combinations were tried and the resulting best combination was obtained using Agfa Gevaert Ltd Structurix F6 film and Structurix RCF (Rapid Cycle Film) screens - fluorometallic type. The major benefit from this result is that this combination is specifically designed for rapid automatic film processing in the pipeline business. The ***^{75}Se^{ntinel}*** source output spectrum interacts optimally with the F6/RCF combination giving the expected exposure reduction factors of 6-8 times common to fluorometallic screens; - with the benefit of the film being processed at a rate of 90 seconds (130cm/min) leading edge to leading edge. This process rate was only previously available to the X-ray technique as the radiographic film quality with Iridium-192 could not be considered acceptable. The previous fastest process time for Iridium-192 would have been approximately 4 minutes LE/LE.

The radiographic image quality is superior to Iridium-192 as is the radiographic sensitivity and consequent flaw sensitivity. When compared to X-ray the contrast is not so high, which can be an aid when viewing multi-run capping welds and the sensitivity values expressed are equivalent.

Agfa Gevaert Ltd was brought into the discussions when it was shown that their product had the best results. With the support of their technical representative from the NDT division, new film and screen formats were created specifically to open up the use of the source. The source and film combinations have been processed successfully using both the NDT OS (offshore machines) and the new generation NDT U (universal machines) both in the trials and field conditions including West Africa.

The complete cycle time for this combination is second to none and has substantially increased the production rates for both crawler, reel spooling and piggyback operations when the optimum lay rates are critical.

Conclusions

A new era has opened up in pipeline gamma radiography with the safety features of this source, which without question can reduce dose rates in the long term and the high levels of exposure previously experienced during incidents involving gamma ray sources. It will also provoke questions against the old adage that X-ray is always safer than gamma ray when we already have experience that when used in the correct set up it will be a safer option.

The radiographic image quality has enabled the ***^{75}Se^{ntinel}*** source to be used where once only X-ray could be considered but without the inherent reliability problems associated with X-ray units, this is especially important for overseas and offshore locations where mobilisation of spares is a costly and problematic process. In line with the long working life of the source, cost benefits on change outs to overseas location are an additional benefit.

The ***^{75}Se^{ntinel}*** source has now been used on hundreds of kilometres of onshore and offshore pipelines/pipe reeling and continues to impress client after client with its safety, quality and cycle time properties. This is a product when used in the optimum way has pushed forward the pipeline business to new levels and will continue to do so in the future.

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