

Storage Pool Water Condition of Multipurpose Compact ⁶⁰Co Irradiator of IPEN-CNEN/SP

Yasko Kodama*, Paulo Roberto Rela

Instituto de Pesquisas Energéticas e Nucleares, IPEN – CNEN/SP, Centro de Tecnologia das Radiações, Av. Prof. Lineu Prestes, 2242, 05508-000, São Paulo, Brazil

Abstract. A new type compact gamma irradiator was developed and installed at IPEN-CNEN/SP, it is classified as category IV irradiator according to International Atomic Energy Agency—IAEA. The originality of its design remains on the rotating concrete door, which integrates the shielding system with the product handling system, permitting the input and output of the products in a continuous way, without the necessity to lower the sources and open the irradiator chamber to change the batch. The licensed capacity of this irradiator is 37 PBq (1 MCi) and the water pool liner, manufactured in stainless steel, has 7.0 m deep and 2.7 m diameter. A sealed radiation source may be defined as a quantity of radioactive material sealed within non-radioactive material, which the confining barrier is intended to prevent leakage or escape of the radioactive material under normal handling conditions and also under foreseeable mishaps. The use of cobalt-60 radiation sources in a wet storage environment requires care in the control of that environment to avoid the potential for deterioration of the stainless steel. In the literature, the tests performed demonstrated that stainless steel had excellent performance under controlled environment conditions. The inspection includes assessing the condition of storage pool water and measuring the pool water conductivity. The demineralised water normally used in storage pools is usually an effective and secure medium in which to store stainless steel. However, there are certain constituents and contaminants in the water which can affect this behaviour. For instance, the pH has a small but definite effect on the tendency for pitting corrosion. Nevertheless, there is also evidence that both strongly alkaline and strongly acid environments can encourage pitting corrosion. In general, an around neutral pH is recommended. The conductivity is used as a measure of the extent to which ionic species are present in the pool water. International recommendations states that conductivity must be maintained between 1.0 and 10.0 microsiemens per centimeter ($\mu\text{S}/\text{cm}$). This will reduce the possibility of corrosion of the sealed source. Extreme care shall be exercised to avoid the introduction of contaminants into the water system (e.g. deionizer regenerants, cleaning materials, corrosive fire extinguishing materials, spilled product). However, conductivity may increase to 20 $\mu\text{S}/\text{cm}$ during occasions such as water treatment plant servicing or source loadings. Also, conductivity may occasionally decrease to 0.5 $\mu\text{S}/\text{cm}$. The desired operating conditions for pH ranges from 4.5 or 5.5 to 8.5, depending on the sealed source manufacturer specification. In this work, four points from storage pool samples and one from water treatment system of multipurpose compact irradiator were collected monthly in the last three years. The pH and conductivity measurements were performed on the every collected sample. All the conductivity values were below the stated limit even just before treatment column exchange, which occurred twice in the period considered here. Despite of pH measured values being very close to the inferior limit of 5.5., in practice, when conductivity specifications are met, pH is less important because the water is so pure that very low levels of contaminants may greatly affect the measurements.

KEYWORDS: *Cobalt 60; Water Pool; Gamma Irradiator; Sealed Source.*

1. Introduction

The commercial use of gamma radiation to sterilize health care products began in the late 1950s, and the technology of processing products with gamma radiation is now well entrenched. With increasing experience and confidence in the technology, more applications are being investigated, and more facilities being built. Now several manufacturers offer a variety of designs that are optimized specifically for different applications, whether this is in the field of food irradiation or environmental applications. This in return has given impetus to the radiation processing industry regarding the types of applications and the size of irradiators [1].

A new type of Cobalt-60 compact irradiator was developed and implemented at IPEN-CNEN/SP, it is classified as category IV irradiator according to International Atomic Energy Agency—IAEA. The originality of its design remains on the rotating concrete door, which integrates the shielding system

* Presenting author, E-mail: ykodama@ipen.br

with the product handling system, permitting the input and output of the products in a continuous way, without the necessity to lower the sources and open the irradiator chamber to change the batch. The licensed cobalt-60 capacity load for this irradiator is 37 PBq (1 MCi) and the water pool liner, manufactured in stainless steel, has 7.0 m deep and 2.7 m diameter [2].

International recommendations state that the manufacturer and user shall take account of the possible effects of fire, explosion, corrosion and any aspects related to the continuous use of the sealed source. The outer capsule material shall be such that it does not significantly corrode under the conditions of storage of the sealed source in the pool. So, one of the factors that should be considered is the environment in which the source is stored, moved and used. Significant radiation exposure may result from loss of control over or damage to the radiation source. In extreme cases the exposures may be sufficient to cause serious injury or even fatalities in the short term. Damage to the source can also lead to widespread contamination. The source itself should be substantially insoluble in water so that the consequences of a breach in the containment are kept to a minimum. In this context it should be noted that cesium chloride is highly soluble in water and the use of ^{137}Cs in this form is not recommended [3].

All *permanent* pool components shall be made of corrosion resistant materials, because corrosion products may affect the integrity of the sealed source. Where practical, stainless steel components (brackets or pulleys) should be passivated, particularly after fabrication. The pool shall be equipped with a water conditioning system capable of maintaining the water in a clean condition and at a level of conductance not exceeding 10 $\mu\text{S}/\text{cm}$. This will reduce the possibility of corrosion of the sealed source. Extreme care shall be exercised to avoid the introduction of contaminants into the water system (deionizer regenerants, cleaning materials, corrosive fire extinguishing materials, spilled product) [3].

Copell [4] has mentioned that the interaction between stainless steels and aqueous environments remains an uncertain science. Within a cobalt storage pool, the situation is made more complex by thermal cycling and by the radiolysis products from irradiation both of water and of materials dissolved or suspended in it. The demineralised water normally used in storage pools is usually an effective and secure medium in which to store stainless steel. However, there are certain constituents and contaminants in the water that can affect this. The radiation fields that surround Co-60 radiation sources generate ozone that is a strong oxidising agent and, when combined with other materials, is capable of causing oxidation of stainless steel. Peroxides are formed by irradiation of water. They are powerful oxidising agents and can lead to oxidation of stainless steel when combined with other materials. Halides (for example, chloride ions), sometimes present as contaminants in pool water, are oxidising agents and can also act as de-passivating agents. They are the most common initiators for pitting corrosion. Nitrates can be formed from the radiolysis of dissolved nitrogen and oxygen. Nitrates are oxidizing agents and, though they do not initiate pitting corrosion, will accelerate its progress once initiated by some other factor. Ferric ions occasionally arise as corrosion products from steel. They act as additional oxidising agents and can accelerate pit growth once pitting has been initiated. Silica and other insoluble materials can be deposited from water as surface layers on the radiation sources. This may have two effects; first it masks the real surface appearance of the steel; second and more significantly, it can create a micro-environment trapped within the surface layer or between it and the steel. Insoluble carbon has been identified as a potential initiator for crevice corrosion. Organic material degrades in the presence of high radiation fields and also in the presence of ozone and can create deposits of insoluble carbon. pH has a small but definite effect on the tendency for pitting corrosion. In most circumstances, alkaline environments (especially those at $\text{pH} > 9$) increase the effectiveness of the passive layer on the surface of stainless steels whilst acid environments can damage or break down the layer in some cases. However, there is also evidence that both strongly alkaline and strongly acid environments can encourage pitting corrosion. In general, an approximately neutral pH is recommended. Conductivity is used as a measure of the extent to which ionic species are present in the pool water. Though there is no exact correlation, it may be assumed to be an indication of the potential presence of chloride or other halide species which would be detrimental [4].

In this study, samples of four points of ^{60}Co sources storage pool and one from water treatment system of multipurpose compact irradiator were collected monthly in the last three years (from Jan., 2005 to June, 2008). pH, conductivity and radioactivity measurements were performed on the every collected samples. The results of pH and conductivity will be presented and discussed.

2. Experimental

2.1 Water sampling points

500 mL of water samples were collected in PE flasks from four points. One point from a tap on the water treatment system and three different levels from the pool: surface, medium and deep.

2.2 Conductivity

Conductivity measurements were made using a portable conductivity analyzer Quimis, model Q405B2.

2.3 pH measurement

pH measurements were made using a Quimis, Q 400MT model, glass electrode model 338-EW. At least three readings for each sample.

3. Results and Discussion

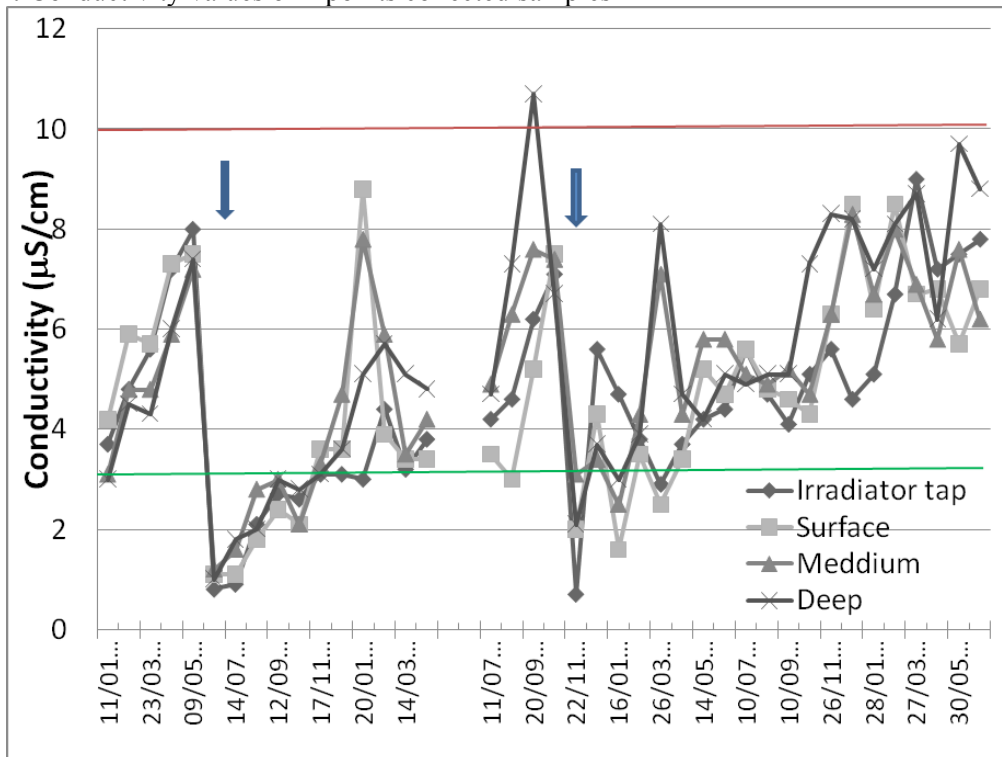
Malkoske [5] has cited that tests and analyses of sealed sources included review of thermal stresses and strains due to cyclic quenching, affects on weld joint design, materials selection and tests, corrosion tests, temperature tests, impact tests and vibration tests. Laboratory tests were initially used to show that there was a need to control water quality in storage pools. The tests demonstrated that AISI 316L stainless steel had excellent performance under controlled environment conditions. The focus of early source surveillance activities was mainly to look for evidence of corrosion and none was found. There were very few examples of poor water quality available at that time and results showed that conditions in the field should limit the risk of corrosion. [5].

During the time on site, the opportunity is usually taken to make a safety inspection of the equipment, sources, and operating conditions under which they are used. The inspection includes assessing the condition of storage pool water and measuring the pool water conductivity [5]. It is possible to observe in Fig. 1 the values of conductivity of the samples collected from the 4 points from 11/01/2005 to 16/06/2008. Arrows indicate deionizing resin exchange dates, that were on May, 2005 and Nov, 2006.

Most of the results of conductivity analysis, Fig. 1, are in concordance with the literature that says that the conductivity must be maintained between 1.0 and 10.0 microsiemens per cm ($\mu\text{S}/\text{cm}$). However, conductivity may increase to 20 ($\mu\text{S}/\text{cm}$) during occasions such as water treatment plant servicing or source loadings [5]. Coppell found that more than 25% of the 169 samples analysed showed conductivity levels in excess recommended upper limit of 10 $\mu\text{S}/\text{cm}$. After closer examination, corrective action was taken in those cases where the high conductivity had arisen from the presence of undesirable impurities [4]. Even though, Krzaniak [6] cited that in a practical sense, conductivity specifications are the result of targets for halides, 1 mg/L of chloride is roughly equivalent to a conductivity of 300 $\mu\text{S}/\text{m}$ in the absence of any other ionic impurities, conductivity may be used as a measure of the overall concentration of ionic species in the water environment; though not as specific as monitoring for individual species (eg chloride), it has been shown to be well correlated with risk factors in water quality, In addition, water should be analysed regularly (at least every six months) to determine what impurities and trace materials it contains. If unusual impurities are identified, expert advice should be sought to determine whether these might constitute a hazard to the integrity of the radiation sources and other components of the irradiator system. If we consider that conductivity values of all samples are only due to chloride presence in the collected water samples from pool of

IPEN's facility, in this study we have found that the values of concentration were between 1 and 3 $\mu\text{g}/\text{mL}$, only one sample presented 4 $\mu\text{g}/\text{mL}$. Specifications attributed to halides by PURIDEC is $< 10 \mu\text{g}/\text{mL}$ and by MDS Nordion is 3 $\mu\text{g}/\text{mL}$ [6]. Then our results meet both specifications.

Figure 1: Conductivity values of 4 points collected samples



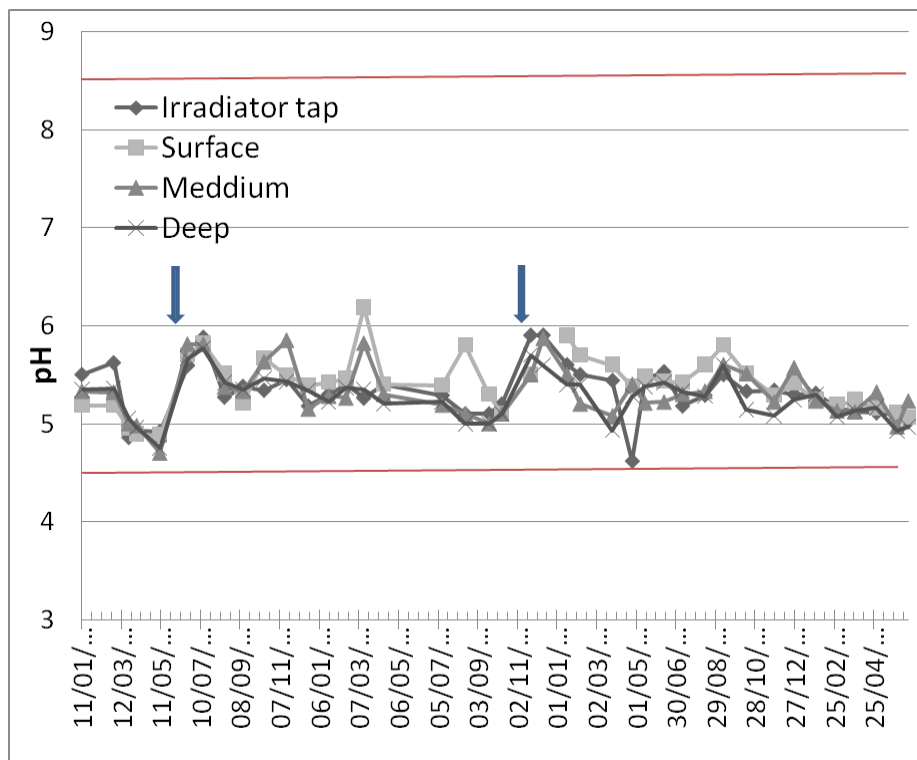
Malkoske [5] had cited that experience have had shown that pitting corrosion had been found on sources examined in the field and this was attributed to the poor environment in which the sources were stored in the field. Being detected at an early stage, the sources were reencapsulated and the situation was corrected when improved control of water quality was exercised. For sources to maintain their integrity over long periods of twenty years or more, it is critical that the conditions under which they are used or stored be maintained according to the conditions for which they were designed. Stainless steels of the 316L or 317L families have been designed to be able to cope in environments such as storage pools and are not prone to general corrosion in such situations. Under normal circumstances, the steel operates in the passive region of its electrochemical polarization curve. It is covered by a thin but dense layer of passive mixed metal oxides which provide it with protection. There are, however, certain stimuli which can cause this passivity to fail [5].

Coppell [4] has cited that the presence and concentration of chloride and other chlorine-containing species is the most common and often the most important factor in the initiation of pitting corrosion. Chloride concentrations as low as 50 mg/L at the metal surface, can be significant in the initiation of pitting. At levels of 50mg/L and above, chloride ions compete for positions within the passive layer on the surface of the steel. They create sites at which the layer ceases to have the structure of a hydrated metal oxide "gel" but, instead, contains regions of metal chloride which then dissolves leaving localised access through the passive layer direct to the metal surface. The overall effect of chloride is to act as a local depassivating agent as well as an oxidising agent. Pitting corrosion occurs where a steel surface is preferentially attacked in one or more local positions. It is a particularly unwelcome type of corrosion because it can lead to perforation of the steel in a relatively short period of time. Pitting corrosion is usually initiated by either halides (eg chloride at a level of 50 mg/L or above) or by imperfections in the steel surface (eg sulphide inclusions or carbides formed during a welding process). Following initiation, pitting can be self-sustaining. For example, if pitting is initiated by a local presence of excess chloride ions, the process may continue even after the chloride has been reduced to trace quantities. The reason

for this is that the high current density in the base of the pit results in a strong positive charge which, in turn, attracts negatively charged chloride ions and other anions and so concentrates them in the area of the pit. Pitting is one of the most common causes of failure of stainless steel components in environments similar to those found in cobalt storage pools.

The Fig. 2 presents the pH of the four points collected water samples from the storage pool of IPEN. Both PURIDEC and MDS Nordion specification for pH is between 4.5 and 8.5 [6,7]. Results found in this study meet the specification limits. By keeping the pH in this range minimises the risk of damage to the passive oxide layer on the surface of the stainless steel source capsules. Coppell [4] had analyzed more than 70 samples of water from a variety of cobalt storage pools from around the world which contained sources supplied by PURIDEC and other manufacturers during consecutive three years. He found that 53% of the samples showed a value for pH which was beneath the PURIDEC recommended lower limit of 5.5. The great majority of samples which fell outside the recommended range were not grossly acid. In these cases, the effects of pH on steel passivity are not great, especially where the water is fairly pure as in these cases. There were no other significant indications of risk and it was concluded that, whilst there may be benefits in raising the pH, the risk represented by the current levels is tolerable.

Figure 2: pH values of 4 points collected samples



Coppell [4] have mentioned that crevice corrosion may occur at positions where the environment is not typical of the general environment in the pool. The features of the bulk pool water and its effect on the behaviour of steel may be significantly changed if there are other factors at work which create localised, atypical environments. An example of this would be crevice corrosion although both cobalt radiation sources and the hardware in which they are stored and used are designed so as to minimise the potential for crevice corrosion. A similar effect can be created by the deposition of insoluble layers on the surface of steel during service. An example of this is the deposition of silica layers from colloidal SiO₂ in the water. Silica has been found to form hard, strongly-adhering coatings which are built up in strata. The deposition of carbon on the surface of the steel may have a similar effect; the carbon can be generated by the irradiation and degradation of organic material (for example oil or grease) in the pool water. Periodically, parts of the coating may break away (probably as a result of differential thermal expansion

during thermal cycling) and the result can be the creation of a rough or layered surface topography. This may have a similar effect to a crevice by providing the opportunity for localised concentration of species such as chloride. In addition, the deposition process which results in the formation of such insoluble layers may combine with the thermal cycling and drying process to trap and concentrate trace materials within the matrix; for example chloride ions.

The results of IPEN's pool water analysis report of the PURIDEC surveillance program is presented in Table 1.

Table 1: Results of IPEN's water analysis reported by PURIDEC

| Chemical attributes | mg/L | Physical attributes | |
|-------------------------------|-------------|----------------------------|-------------|
| SiO ₂ | 15 | pH | 4.82 |
| F ⁻ | 0.06 | Conductivity | 5.74 |
| Cl ⁻ | 0.47 | ⁶⁰ Co | < 0.2 Bq/L |
| NO ₃ ⁻ | < 0.05 | ¹³⁷ Cs | < 0.09 Bq/L |
| SO ₄ ²⁻ | < 0.05 | | |

Coppell [4] had cited that more than 30% of the samples contained silica levels in excess of 10 µg/mL. At present, there is insufficient data available for PURIDEC to recommend a specification for maximum silica levels but it is clear that, in several examples, there was sufficient silica available in the pool water to lead to the formation of deposits on the sources with the attendant risk of concentration effects. Even though our silica result is in agreement with those in excess, all the results meet specification.

In the early 1970' s, a small number of C- 188 had been stored in non-de-ionized water environments. Source surveillance activities concentrated on corrosion investigations with no positive corrosion occurrence. Further laboratory tests demonstrated that there was a need to control water quality in the storage pools. Although no material is totally corrosion resistant, tests and experience demonstrated that AISI 316L stainless steel has excellent performance under controlled environmental conditions. There have been two instances of a small amount of micro-pitting due to poor water quality in the early stages of use. In both cases deeper corrosion development was arrested due to the adoption of pool water deionizers and the sources in question were removed from service for re-encapsulation or disposal. No C-188 surveillance source has showed any loss of leak tightness integrity or any weld crack initiation [6]. So it is very important to control the water quality periodically.

4. Conclusion

Maintaining source integrity and safety is of paramount importance to the radiation processing industry. The credibility of the gamma processing industry requires a commitment to quality in design, manufacture and use. A controlled environment is critical to the integrity of sealed sources and manufacturers have defined suitable operating conditions for their sealed sources. In general, manufacturers define temperature requirements for transport and use, controls on materials in contact with the sources, air and pool water quality specifications. These are designed to ensure that the encapsulation maintains its corrosion resistance, and to ensure that the sealed sources operate within known thermal cycling conditions. [8].

Even though, the water quality of IPEN's pool meets the requirements of REVISS' Supplementary Conditions of Sale, more effort on implementing other chemical analysis has started. For instance, the chemical analysis for determination of chloride instead of the use of conductivity as the only parameter.

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