DEVELOPMENT OF AN ISOTROPIC UNDERWATER DEVICE FOR COLOR ENHANCEMENT OF GEMSTONES

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ABSTRACT

Over the past years a small-scale Gamma Irradiation Facility of the Nuclear and Energy Research Institute -IPEN has been provided services for color enhancement of Brazilian gemstones. Traditionally the gemstones are placed inside of closed screen steel bags, and then suspended deep inside the water pool by fixed steel cables in front of the Cobalt-60 sources. The processing of this material usually was performed every weekend when the facility was not operating to avoid problems related to the sources movement. The gemstones underwater irradiation is preferred among other reasons because increase their color stability and homogeneity. A hollow cylindrical device, length of 46 cm and external diameter of 38 cm built in perforated aluminum was developed to house approximately 25.0 kg (38.0 l.) of selected standard size quartz gemstones before the final cutting. The cylindrical device was constructed using defined size in order to fit a smaller cylindrical castle placed on the bottom of the pool containing 22 aligned linear radioactive sources totalizing 6.81×10^2 TBq (1.84x10⁴ Ci). A mechanical winch fixed in an aluminum holder together with stainless steel cable and quicklock carabiners were used to descend in safety the loaded cylindrical device until to engage the device outside the smaller castle. Several dosimetric tests were performed using Harwell Red PMMA located in selected positions around the device to study the dose rate and the dose distribution. In most cases, quartz gemstones were irradiated with doses between 2 - 2.5 MGy. To achieve the desired dose values are necessary approximately 30 days of processing. One of major advantages is related to the irradiation treatment using the developed device because this does not interfere with the normal schedule operation of this facility as also the fact that is not necessary manipulate the gemstones by the end of the procedure.

1. INTRODUCTION

Suitable natural geological conditions make the Brazil one of the most important gemstones global producers. Large ore deposits containing several kinds of gemstones have allowed increase and improve alternative color enhancement treatments such as gamma irradiation by cobalt-60. Gemstones gamma irradiation in Brazil has been successful primarily because in the country exist two large gemological colored pegmatitic provinces and greatest number of hyaline quartz occurrences [1].

In 1982, a private Brazilian irradiation facility started providing the irradiation gemstone services using cobalt-60 but only in 2003 it started a large scale production of hyaline quartz coming to lead the South American irradiated gemstones market in 2007 [1]. A small scale Gamma Irradiation Facility of the Nuclear Research Institute – IPEN, São Paulo - Brazil started up in 2004 focusing mainly sterilization treatments [2], then the quartz gemstone underwater irradiation was also implemented.

The gemstones are placed inside of closed screen steel bags, and then suspended deep inside the water pool by fixed steel cables in front of the Cobalt-60 sources. The processing of this material usually is performed every weekend when the facility was not operating to avoid problems related to the sources movement. Another kind of devices to irradiate gemstones were developed [4] but currently only last one cited early was being used. One of the more important facts disclosed in all those research years is related to the irradiation process needs to be carried inside the water to obtain better visual results. Many theories had been proposed to explain it but maybe the better reason is associated to the water moderation of cobalt-60 energy creating X-rays in the Compton region something similar to the sphere of incidence effect [3]. Other parameters such as the temperature irradiation are very important because Bremsstrahlung and heat production are dependent of the atomic number Z.

In this work green–gold and green amethyst quartz samples were used to study the dose distribution inside a cylindrical device developed to achieve isotropic fields of gamma radiation using colbat-60 linear sources.

2. EXPERIMENTAL WORK

A hollow cylindrical device, length of 46 cm and external diameter of 38 cm built in perforated aluminum capable to house approximately 25.0 kg (38.0 l.) of selected standard size quartz gemstones was developed as shown in Fig.1. The cylindrical device was constructed using defined size in order to fit a smaller cylindrical sources holder castle placed on the bottom of the water pool facility at a depth of 7 m. containing 22 aligned linear radioactive sources totalizing 6.81×10^2 TBq (1.84×10^4 Ci). Every linear radioactive source has length of 45 cm and external diameter of 1 cm. A mechanical winch fixed in an aluminum holder together with stainless steel cable and quick-lock carabiners were used to descend in safety the loaded cylindrical device until to engage the device outside the smaller castle containing the radioactive sources on the bottom of the pool.



Figure 1: Schematic view of the hollow cylindrical device.

The 22 radioactive cobalt-60 sources were distributed around the holder. Each source had very similar activity (\sim 30 TBq) with the mean value of the set was 30.9±1.6 TBq (835,3±42.8Ci) and variation coefficient of 5.1 which means a good uniformity of the radiation field.

Several experimental measurements were carried on using Harwell Red PMMA dosimeters placed in desired positions around and inside the device (16 per level), to study the dose rate and the dose distribution on fifth different levels, totalizing 80 dosimetric measurement points as shown in Fig. 2 . All measurements were carried into the water pool with the empty and the quartz gemstones filled device.



Figure 2: (a) Cylindrical device top view and dosimetric measurement points; (b) Dosimetric measurement levels

The PMMA commercial dosimeters were analyzed using a UV-VIS spectrophotometer at wavelength 640 nm and the producer calibration curves [5,6,7,8]. These kinds of dosimeters change the absorbance or transmittance at defined light wavelength in proportion with the absorbed dose (5-50kGy). The total exposition time for all dosimeters in all experiments was 6h. The dose rate results were processed to obtain the standard deviation for each level and for each radial position [9]. Statistical errors and residuals were calculated and presented in all results.

In order to obtain additional radial and axial dosimetric measuring data points inside the cylindrical device, the MNCP (Monte Carlo simulation code for the transport of the radiation) was used to compute another dosimetric measurements [10,12]. In the first step every twodimensional level was simulated and studied to create several radial data then the MCNP code was used to create numerous axial dosimetric points. The image results were reconstructed using 30x30 pixels matrix [11].

3. RESULTS AND DISCUSSION

3.1. Two-dimensional absorbed dose distribution results

The cross-sectional absorbed dose radiation distribution images sometimes called isodoses diagrams for the levels 1 to 5 are showed in Fig.3 to Fig.7 respectively. Higher doses are presented with red color while low doses with blue color. In general terms, the radial distribution of the absorbed dose was very uniform, however exist a little difference between the axial planes as will be shown later.



Figure 3: Cross-sectional dose distribution at Level 1



Figure 4: Cross-sectional dose distribution at Level 2



Figure 5: Cross-sectional dose distribution at Level 3



Figure 6: Cross-sectional dose distribution at Level 4



Figure 7: Cross-sectional dose distribution at Level 5

The absorbed dose and dose rate results of the external and internal rings for each measuring level are showed at Table 1. The minimal dose rate at level 4 (2,77 kGy.h⁻¹) at the external ring position is determinant to the process because this value allows to calculate the total irradiation time (between 30 to 38 days) necessary to reach 2 - 2.5 MGy. These results put in evidence that inside the device at the final of the process, some portions of material have been irradiated with doses four times higher than the minimal required value but this is not represents a problem for this kind of quartz because once the material achieves a specific color at defined dose (ex. 2 MGy) the increase of this propriety cannot improve the final color of the gemstones. For this reason is not necessary the rotation of the device around the sources.

	External Ring		Internal Ring	
Level	Dose	Dose rate	Dose	Dose rate
	(kGy)	$(kGy.h^{-1})$	(kGy)	$(kGy.h^{-1})$
1	17.93 ± 0.27	2.99 ± 0.04	39.31 ± 0.59	$6.55 \hspace{0.1cm} \pm \hspace{0.1cm} 0.10 \hspace{0.1cm}$
2	16.11 ± 0.28	2.68 ± 0.05	35.31 ± 0.62	5.88 ± 0.10
3	17.33 ± 0.31	2.89 ± 0.05	37.98 ± 0.68	6.33 ± 0.11
4	16.62 ± 0.14	2.77 ± 0.02	36.43 ± 0.32	$6.07 \hspace{0.1in} \pm 0.05$
5	16.65 ± 0.69	2.78 ± 0.11	36.50 ± 1.51	6.08 ± 0.25

Table 1: Radial absorbed dose and dose rate

3.2. Three-dimensional absorbed dose distribution results

The results of the 3D absorbed dose distribution images for the cylindrical device containing the quartz gemstones are showed in Fig. 8 as an axial section to make easy the visualization. A two-dimensional section is also showed in Fig. 9. In this work only was studied the gemstone filled region without sources volume located in the middle of the device.



Figure 8: 3D Axial section dose distribution



Figure 9: 2D Axial section dose distribution

3.2. Final cutting products

Several tests were performed using two kind of quartz material green-gold and green amethyst (prasiolite) as showed in Fig. 9. Sometimes are necessary additional treatments with heat and UV radiation to stabilize or rectify the color (photostability). In all cases first the select quartz material was irradiated and then it was cut.



Figure 9: In order from left to right: Green-gold and green amethyst (prasiolite) final cutting products, samples after the irradiation process and original quartz before the irradiation process

4. CONCLUSIONS

The developed device is an excellent tool to process and to continue the studies of the Brazilian gemstones. The radiation field inside was very uniform in relation to the radial and the axial positions. For the green-gold and green amethyst quartz samples used in the experiments, the increasing of the absorbed dose (>2 MGy) when the final color was achieved does not change the gemstones final color, then in this case the rotation movement of the device around the sources is not necessary. One of major advantages is related to the irradiation treatment using the developed device because this does not interfere with the normal schedule operation of this facility as also the fact that is not necessary manipulate the gemstones by the end of the procedure.

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REFERENCES

- 1. CGEE, "Estudo da Cadeia de Suprimento do Programa Nuclear Brasileiro Relatório Parcial Irradiadores e Aplicações Irradiação de Gemas (pedras preciosas), CGEE, Brasilia, Brazil (2010).
- 2. W.A.P. Calvo, P. Rela, L.G.A. Silva, A small size continuous run industrial gamma irradiator, *Radiation Physics and Chemistry*, **71**, pp.561-563 (2004).
- 3. N. Omi, P. Rela, "Underwater versus dry irradiation with ⁶⁰Co, a qualitative analysis", Proceedings of the *International Nuclear Atlantic Conference INAC*, Santos Brazil, September 30 to October 5, 2007.
- 4. N. Omi, P. Rela, "Gemstone dedicated gamma irradiator development", Proceedings of the *International Nuclear Atlantic Conference INAC*, Santos Brazil, September 30 to October 5, 2007.
- 5. Harwell Dosimeters, "Characteristics of Harwell Gammachrome YR PMMA Dosimeters", Culham/Harwell Design Services, Oxfordshire, United Kingdom (2000).
- 6. Harwell Dosimeters, "Harwell PMMA Dosimeters for Radiation Processing", Culham/Harwell Design Services, Oxfordshire, United Kingdom (2000).
- 7. K.M. Glover, M.E. Plested, M.F. Watts, B. Whittaker, "A Study of some parameters relevant to the response of Harwell PMMA dosimeters to gamma and electron irradiation", Process Service Division, AEA Technology, Harwell Laboratory, Oxfordshire, United Kingdom (2000).
- 8. Harwell Dosimeters, "Uncertainties in absorbed dose as measured using PMMA dosimeters", Culham/Harwell Design Services, Oxfordshire, United Kingdom (2000).
- 9. P.A.S. Vasquez, M.M. Hamada, Methodological analysis of gamma tomography system for large random packed columns, *Applied Radiation and Radioisotopes*, **68**, pp.658-661 (2010).
- 10. P.A.S. Vasquez, M.M. Hamada, "Simulated study of parallel-beam gamma ray tomography and image reconstruction", Proceedings of the *International Nuclear Atlantic Conference INAC*, Santos Brazil, September 30 to October 5, 2007.
- 11. P.A.S. Vasquez, M.M. Hamada, "Phantom study using a first generation gamma tomography system", Proceedings of the *International Nuclear Atlantic Conference INAC*, Santos Brazil, September 30 to October 5, 2007.
- 12. M.J. Berger, J.H. Hubbell, S.M. Seltzer, J. Chang, J.S. Coursey, R. Sukumar, D.S. Zucker, and K. Olsen, XCOM: Photon Cross Sections Database, *NIST, PML, Radiation and Biomolecular Physics Division*, <u>http://www.nist.gov/pml/data/xcom/index.cfm</u>