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ORIGINAL PAPER

Radiation processing for cultural heritage preservation – Romanian experience

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Abstract. Radiation sterilization has been considered a mass decontamination technique for biodegradable cultural heritage (CH) since its widespread application in the medical field. Initial experiments have revealed advantages, for example, efficiency and effectiveness, but also disadvantages, namely "side effects" concerning CH materials. More than 50 years later, the adequacy of ionizing radiation for some CH artefacts is still the subject of discussion. The main reason why is that science and industry are not yet able to provide a more efficient technique for treating mass decontamination. For wooden items, there is general agreement that the irradiation dose required for insect eradication is not damaging, even in the case of polychromed wood. For cellulose pulp (paper), there is a reduction in polymerization degree (DP) at the high doses necessary to stop the attack of fungi, but this should be considered taking into account the purpose of the treatment. Emergency or rescue treatments are necessary to mitigate the consequences of accidents or improper storage conditions. In some cases (archives), the value of written information is greater than the historical value of the paper support. For other materials, namely textiles, leather and parchment, less research has been published on the effect of ionizing radiation treatment. As a general rule, irradiation is not necessary when only a few CH elements are present that are affected by biological contamination since restorers can solve the problem by classical means. The need for radiation treatment arises when large collections (hundreds, thousands or even more elements) are heavily affected by the biological attack. In Romania, the IRASM gamma irradiator of IFIN-HH is receiving an increasing number of requests for CH treatment, mainly due to an intensive research programme concerning this topic and close liaison with CH owners or administrators. Besides reviewing the scientific results obtained in Romania and abroad, this paper presents some examples from experiences in Romania.

Keywords: radiation processing • cultural heritage • disinfection efficacy • risk assessment

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Introduction

Radiation sterilization used to be considered a mass decontamination technique for biodegradable cultural heritage (CH) shortly after it was applied in the medical field. Initial experiments [1, 2] exhibited advantages in terms of efficiency and effectiveness, but also disadvantages, namely "side effects" on CH materials. More than 50 years later, the adequacy of ionizing radiation for some CH artefacts is still a topic of discussion. The main reason for this is that science and industry are not yet able to provide a more effective decontamination technique.

Many CH artefacts are biodegradable by nature, for example, wood, paper, leather and textiles can be degraded by several biological agents: insects, fungi or bacteria. Table 1 highlights the main biological threats of cultural heritage, taking into account the frequency of their occurrence.

Table 1. Biological threats to CH materials

Material in CH artefacts	Biological threat
Wood	Fungi, insects
Paper	Fungi, insects
Leather	Bacteria, fungi, insects
Textiles	Bacteria, fungi, insects
Film, photographs	Bacteria, fungi

The worst damage to wooden objects is caused by beetles or wood larvae. Fungi and some xylophagous bacteria cause damage more slowly than worms, usually during the final stage of degradation of the wood. Under high levels of humidity, paper stored in large quantities, for example, in libraries and archives, is easily damaged by fungi. Likewise, old leather is immediately attacked by fungi in the presence of water. Textiles are affected by insects, namely clothes moths, and by fungi in the case of cotton and other cellulose fibres. Due to their gelatin layer, films and photos are easily attacked by fungi. In many cases, biological attack creates a micro-environment with its own ecological system, involving more than one species, making it difficult to assess the effectiveness of a particular decontamination technique.

For individual CH articles, restorers have their own techniques to stop and eliminate biological threats. The problem occurs when large quantities are involved (hundreds of thousands of articles or even tons of material). The use of chemical biocides is limited by residues that may affect users. Freeze drying and anoxia exhibit lower levels of efficacy for resistant forms of the biological agent, namely spores, eggs or pupae. In this context, radiation treatment is fast and efficient, and costs are more closely related to the relocation of large amounts of CH than to the irradiation facility.

After overcoming the concerns about "radiation" or "radioactivity" (it should be noted that none of the radiation processing applications produce radioactivity), reservations remain with regard to possible radiation-induced damage, i.e. side effects.

The effects of ionizing radiation on CH materials were studied mainly for cellulosic materials, especially for paper [3–22] and pigments [23–25]. Most authors agree that reducing the degree of cellulose polymerization is the main side effect of paper irradiation. Many reports have observed insignificant levels of change or none at all in terms of the macroscopic properties of the paper, which do not correlate with the significant reduction in the degree of polymerization. Sometimes, on the basis of similar results, some authors encourage the use of radiation treatment for cultural heritage [3, 8, 9] and others disagree [26].

There is general agreement that irradiation for pest control does not produce measurable effects in wood and that most historic panel paintings have not been modified up to radiation absorbed doses of 10 kGy [27–31]. Fewer studies have been published concerning leather or parchment [32, 33], textiles [34] and photographic/film materials [35].

A thorough review of the side effects on most CH materials is presented in the IAEA publication [36].

In Romania, experiments concerning the treatment of radiation in terms of cultural heritage commenced more than 30 years ago [37]. Initial experiments were conducted with low-activity Co-60 sources and pilot irradiators. Since 2000, large-scale treatments have begun at the IFIN-HH IRASM multipurpose irradiation facility. Considering the pressing need for a fast, reliable and cost-effective method concerning CH disinfection, IFIN-HH has developed comprehensive studies addressing the effects of ionizing radiation on various materials, for example, wood, polychromed wood, paper, paintings, leather and textiles, funded by R&D projects in national competition.

Some of our recent work is geared towards finding a reliable method of quantifying the benefits of radiation treatment that could be used in a cost-benefit analysis preceding the decision to take action on CH materials strongly affected by a biological attack.

Risk assessment is a current tool utilized by the nuclear field amongst other technical fields, namely medical devices, pharmaceuticals, food products, etc. The current edition of the ISO 9001 standard includes risk management in terms of quality management and will apply risk assessment techniques to many other activities.

In this paper the methods used to demonstrate the efficacy of radiation treatment in terms of multiple radiation treatments with large amounts of CH materials and a model risk assessment analysis are presented.

Materials and methods

The gamma irradiation of CH collections was performed at the IRASM Radiation Processing Center of IFIN-HH. For the irradiation of small samples a Gamma Chamber 5000 self-shielded irradiator (BRIT, India) was used. For the treatment of large CH collections, a SVST-Co-60/B tote-box irradiator (pictured in Fig. 1) was used. Items received in cartons (paper) or sacks (mixed collections) were irradiated in batches, up to 2000 kg per batch. The large wooden elements were manually transported inside the irradiation room and were irradiated whilst stationary. In order to improve the dose uniformity ratio, irradiation was discontinued and the objects were manually turned 180 degrees.

The absorbed dose was measured by a calibrated Ethanol-Chlorobenzene Dosimetry System (ISO/

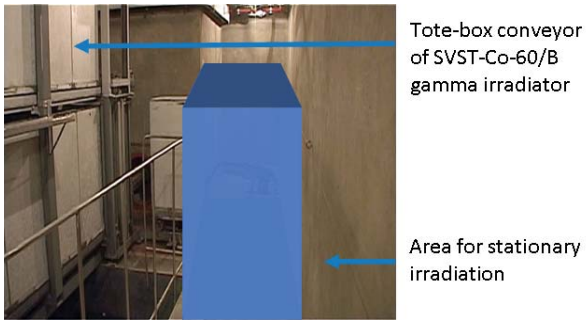
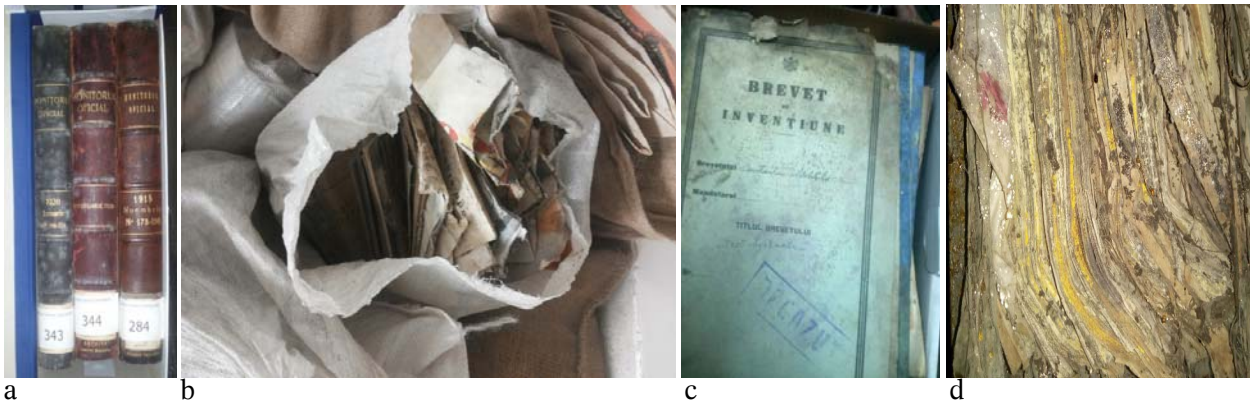


Fig. 1. SVST Co-60/B Multipurpose Gamma Irradiator: tote-box conveyor and the area for stationary irradiation.



a b c d

Fig. 2. Documents highly affected by fungi: (a) volumes with leather covers from the "Official Journal Collection", (b) bags containing items from the "Museum Inventory", (c) patents from the "Royal Patents Collection", (d) documents from the "Sahia Film Archive".

ASTM 51538) [38]. In the case of collections packed in carton boxes or bags (paper, mixed), the ECB ampoules were placed in the maximum and minimum positions (known from operational qualification of the irradiator) of each tote box. ECB dosimeters were measured by oscillometric readout method (Radelkis reader – Institute of Isotopes).

Microbiological tests were performed in the microbiological laboratory at IRASM. Two types of sampling were used, depending on the availability of the material at the time, sampling conditions and the compatibility of the substrate: surface sampling and material mass.

Three methods were used for tests applied to surfaces:

- Surface contamination test using a dry swab: the surface of the tested item was swabbed and the collected microorganisms were inoculated in Petri dishes with culture media.
- Surface contamination test using a wet swab: similar to the dry swab technique but the swab was soaked in a buffer solution.
- Surface contamination test using the contact plate method: contact plates with culture media on a convex surface were applied to the surface of the tested items and the microorganisms were collected directly.

The surface tests were always performed on equally sized areas of similar, highly contaminated adjacent surfaces, before and after irradiation.

A second type of test was performed on masses of paper material. Because of its destructive nature, bioburden testing (total count) was performed on samples of highly degraded paper, which could not be restored. The samples were vigorously mixed in a sterile buffer and aliquots were incorporated in liquefied culture media (Malt Extract Agar), then incubated between 19 and 20°C for 6 to 7 days. The composition of the medium and the temperature of incubation were selected to promote the growth of filamentous fungi, including slow growing isolates.

The risk assessment was performed in a simple way, taking into account only two parameters, namely probability and severity, for each hazard that was identified. Each parameter was assigned a numerical value from 1 to 3. Probability was assessed as follows: 1 – unlikely, 2 – rare and 3 – probable. Severity was classified as: 1 – negligible, 2 – moderate and 3 – critical. For each hazard, its priority index was calculated: P.I. = probability × severity.

Their global risks were evaluated as an average of each probability index: $(1/n)\Sigma_i^n p_i \times s_i$, and the magnitude of each risk was assigned based on the following scale: low risk: 1–3, medium risk: 4–6, and high risk: 7–9.

Results and discussion

Four gamma irradiation treatments of paper documents are analysed in this paper that were carried out at the IRASM Center between 2014 and 2016 (photos of samples can be found in Fig. 2) in addition to a gamma irradiation treatment performed in 2014 on a collection of six large wooden sculptures (Fig. 3).

The treatment of each collection of paper documents and mixed collections was performed over several irradiation runs (batches) and the dose values in Table 2 represent the minimum and maximum achieved for each collection.

Microbiological tests (before and after treatment) were only conducted on paper documents and for items in the mixed collection. The results are presented in Tables 3 to 6. Microbiological tests were only conducted to estimate the decrease in the size of the actual microbial population after treatment. To meet the general goal of the treatment, the worst case scenario was taken into consideration; as such, the most contaminated specimens were sampled according to visual examination. One should be aware that

Fig. 3. Large wooden sculptures affected by woodworms from the "Nicăpetre" collection.

Table 2. Gamma irradiation treatments and the minimum number of logarithmic reductions in terms of surface contamination

Treatment	Minimum dose [kGy]	Maximum dose [kGy]	No. of log. reductions
"Official Journal Collection" (Romanian Parliament Archives, 2014) ~4 tons of documents	5.6 ± 0.4	7.9 ± 0.5	3
"Museum Inventory" (Bucharest National Theater, 2015) ~6 tons of documents, stage props, photos, etc.	5.2 ± 0.4	9.0 ± 0.6	3
"Royal Patents Collection" (Romanian State Office for Inventions and Trademarks, 2016) ~4 tons of documents	5.8 ± 0.4	8.5 ± 0.6	3
"Sahia Film Archive" (Romanian Ministry of Culture, 2016) ~3 tons of documents	5.8 ± 0.4	7.5 ± 0.5	5

Table 3. Surface contamination on samples from the "Official Journal Collection" – wet swab technique over a surface area of 50 × 50 mm

No.	Sample	Number of colony-forming units (CFU)	
		Before irradiation	After irradiation
1	Box no. 1 (vol. 167)	480	0
2	Box no. 1 (vol. 206)	>1000	2
3	Box no. 2 (vol. 106)	48	0
4	Box no. 3 (vol. 343)	134	0
5	Box no. 1 (vol. 260)	>1000	1

Table 4. Surface contamination on samples from the "Museum Inventory" – contact plates technique (24 cm^2)

No.	Sample	Number of colony-forming units (CFU)	
		Before irradiation	After irradiation
1	Box no. 1 (Photo album)	1200	1
2	Box no. 1 (Photo album)	1312	0
3	Box no. 1 (Photo album)	432	5
4	Box no. 2 (Photo album)	240	3
5	Box no. 2 (Photo album)	1600	1
6	Box no. 2 (Photo "Tablou Th. Popescu")	120	0
7	Bag no. 3 (Photo "Caleașcă cu sultan")	312	2
8	Bag no. 3 (Photo "Portret Nataşa Alexandra – Medalia Muncii")	240	1
9	Bag no. 4 (Photo "Portret Ag. Macri Eftimiu")	144	0
10	Bag no. 4 (Photo "Portret Lazăr Vrabie")	288	1
11	Bag no. 4 (File cover "Caragiale")	1360	1
12	Bag no. 5 (File cover "Cremer Robert")	480	0
13	Bag no. 6 (Old documents, heavily degraded)	1288	0
14	Bag no. 6 (Old documents, heavily degraded – "Letter Maria Filotti")	1840	0
15	Bag no. 6 (Old documents, heavily degraded – book for visitors)	1264	1

microbiological tests do not provide exact results concerning the overall microbial contamination of the entire collection, due to very high degrees of heterogeneity. This decrease in microbial contamination is expressed in terms of the logarithmic decrease in the size of the initial population (Table 2).

Since the main purpose of the treatment of wooden CH material was the eradication of insects, namely woodworms, no microbiological tests were performed on the "Nicăpetre" collection. There are practically no analytical methods for the evaluation of insect activity, only that of monitoring specimens over many years. In terms of wood there is general agreement that no measurable side effects result from even very high doses that exceed 50 kGy with no maximum dose limitation. The maximum dose measured for each wooden item resulted from the geometry of the irradiation. For massive wooden items each of 100–200 kg in weight the maximum dose exceeded 20 kGy. The minimum and maximum doses were measured in the accessible areas of the wooden items (Table 7).

The following tables (8–10) summarize the risk assessments for three models covering the four collections of documents and a collection of wooden sculptures that were examined: a collection significantly degraded by fungi, a collection exposed to early or low levels of fungal degradation, and a collection damaged by woodworms.

The assignment of specific values for the risk assessment parameters, namely probability and severity, is specific to each field of activity, and depends on the knowledge and experience of the analyst and the existence of a known history of similar cases. In the case of the CH collections, the availability of additional analytical data concerning the materials involved may also be considered.

Table 5. Bioburden of samples (destructive analysis of fragments of paper) from the "Royal Patents Collection"

No.	Sample	Number of colony forming units (CFU/g)	
		Before irradiation	After 5 kGy of irradiation
1	Shelf no. 6 – sample 1	2.0×10^3	<67
2	Shelf no. 6 – sample 2	1.0×10^4	<70
3	Shelf no. 12 – sample 2	2.3×10^6	$<2 \times 10^3$
4	Box no. 3	1.4×10^4	<70

Table 6. Surface contamination on samples from the "Sahia Film Archive" – dry swab technique over a surface area of 50 × 50 mm

No.	Sample	Number of colony forming units (CFU)	
		Before irradiation	After irradiation
1	Pallet no. 1 – sample 1	5.1×10^5	0
2	Pallet no. 1 – sample 2	9.5×10^5	0
3	Pallet no. 1 – sample 3	3.1×10^6	2
4	Pallet no. 2 – sample 1	1.0×10^5	0
5	Pallet no. 2 – sample 2	9.5×10^4	0
6	Pallet no. 2 – sample 3	6.3×10^5	0
7	Pallet no. 3 – sample 1	5.0×10^5	5
8	Pallet no. 3 – sample 2	7.0×10^5	0
9	Pallet no. 3 – sample 3	7.5×10^5	3
10	Pallet no. 4 – sample 1	1.0×10^6	0
11	Pallet no. 4 – sample 2	2.5×10^5	0
12	Pallet no. 4 – sample 3	4.5×10^5	0
13	Pallet no. 5 – sample 1	3.5×10^5	0
14	Pallet no. 5 – sample 2	4.6×10^5	0
15	Pallet no. 1 – sample 3	1.4×10^5	0

Table 7. Gamma irradiation treatment of the "Nicăpetre" collection of wooden sculptures

Treatment	Minimum dose [kGy]	Maximum dose [kGy]
Item no. 1 (Inv. 238)	5.7 ± 0.4	22.5 ± 1.0
Item no. 2 (Inv. 239)	5.4 ± 0.4	21.0 ± 1.0
Item no. 3 (Inv. 240)	5.7 ± 0.4	25.0 ± 1.0
Item no. 4 (Inv. 242)	5.2 ± 0.3	22.3 ± 1.0
Item no. 5 (Inv. 244) Item no. 6 (Inv. 245)	5.5 ± 0.4	16.8 ± 0.8

For example, the likelihood of side effects on wooden articles was assigned "1" (unlikely) based on data from the literature. The likelihood of side effects on paper was assigned "2" (rare), also based on data from the literature as well as the results of some non-destructive tests performed on selected samples of paper collections, namely colour and Fourier-transform infrared spectroscopy (FTIR) tests, which are not reported here.

In this way, values concerning the probability of each selected hazard were assigned for each of the three models presented in Tables 8 to 10. According to the microbiological results, the "collection of documents significantly degraded by fungi" corresponds to the "Sahia Film Archive" (Romanian Ministry of Culture, 2016). The "Official Journal Collection" (Romanian Parliament Archives, 2014), the "Museum Inventory" (Bucharest National Theater, 2015) and the "Royal Patents Collection"

Table 8. Risk assessment for a collection of documents significantly degraded by fungi

No.	Hazard	Probability (P)	Severity (S)	Priority index (P.I. = P×S)	Initial risk	Actions*)
	I. Initial risk assessment					
1	Total loss of physical integrity of documents and/or total loss of written information	2	3	6	High	Radiation treatment
2	Documents are not accessible due to health hazards	3	3	9		
	II. Residual risk assessment					
1	Total loss of physical integrity of documents and/or total loss of written information	1	3	3	Low	
2	Documents are not accessible due to health hazards	2	1	2		
3	Side effects of the radiation treatment	2	2	4		

*) Proposed actions to reduce the risk.

Table 9. Risk assessment for a collection of documents with a low level of incipient or inactive fungal attack

No.	Hazard	Probability (P)	Severity (S)	Priority index (P.I. = P×S)	Initial risk	Actions*)
	I. Initial risk assessment					
1	Total loss of physical integrity of documents and/or total loss of written information	3	2	6	Medium	Radiation treatment
2	Documents are not accessible due to health hazards	2	2	4		
	II. Residual risk assessment					
1	Total loss of physical integrity of documents and/or total loss of written information	1	3	3	Low	
2	Documents are not accessible due to health hazards	2	1	2		
3	Side effects of the radiation treatment	2	2	4		

*) Proposed actions to reduce the risk.

Table 10. Risk assessment for a collection of wooden CH material severely attacked by woodworms

No.	Hazard	Probability (P)	Severity (S)	Priority index (P.I. = P×S)	Initial risk	Actions*)
	I. Initial risk assessment					
1	Total loss of physical integrity of wooden items	2	3	6	High	Radiation treatment
2	Contamination with xylophagous insects from other wooden collections of the museum	3	3	9		
	II. Residual risk assessment					
1	Total loss of physical integrity of wooden items	1	2	2	Low	
2	Contamination with xylophagous insects of other wooden collections of the museum	1	3	3		
3	Side effects of the radiation treatment	1	2	2		

*) Proposed actions to reduce the risk.

(Romanian State Office for Inventions and Trademarks, 2016) are categorized by the "collections of documents exposed to low levels of incipient or inactive fungal attack" model.

The accuracy of the risk assessment is influenced by a more accurate assessment of the initial state of the CH collection. For example, in the case of documents, there are two models of evaluation available in the literature, namely Havermans *et al*. [39] for archives, and Capiau *et al*. [40] for library books, which could be extended to any other type of material.

Conclusions

Recently in the literature, there have been numerous reports concerning ionizing radiation treatments for disinfecting cultural heritage, which show that this technique is gaining more and more acceptance from the owners and administrators of cultural heritage. The benefits of treatment are indisputable and most side effects have been studied and reported. At the time of writing, the IRASM facility of IFIN-HH has treated over 500 m^3 of wood, paper and mixed collections over the previous 15 years.

To evaluate the efficacy of ionizing radiation treatment as far as paper is concerned, the first reported method was the inactivation of isolated fungal strains from documents and their cultivation on culture media [1, 41]. Since it is not directly related to the level of artefact contamination, this method can lead to a high dose of treatment, resulting in undesirable side effects. In previous works [19, 42], a method based on the evaluation of the bioburden before irradiation and the testing of the radiation resistance of isolated dominant paper strains without their identification was applied. This method requires a lower dose of treatment but is destructive, relatively expensive and time-consuming in terms of radiation resistance studies (D_{10} determination). In the present paper, a different approach was investigated: the treatment dose was limited to less than 10 kGy for reasons related to the side effects and an attempt to evaluate the efficacy of the treatment by non-destructive testing of surface contamination was made. All microbiological tests performed in this paper showed a reduction in initial contamination by a factor of 10^3 or more.

All three of the aforementioned methods possess some inconveniences, namely high costs, time-consuming, failure to take into account the non-cultivable strains, and imbalanced competitiveness in the *in vitro* culture. Further developments to improve the accuracy of determining the effectiveness of radiation treatment will focus on establishing a library of common fungal strains specific to any particular type of CH material.

The risk assessments presented in this paper concluded that the overall residual risk is "low", but different circumstances may lead to the acceptance

of a higher residual risk. It is important to state that the failure to implement any interventions will maintain high levels of risk, therefore, any action to reduce risk is beneficial to the CH collection.

Risk assessment could become a useful tool for choosing a specific intervention to save cultural heritage collections under biological attack. The risk assessment models presented in this paper can be applied to other methods of decontamination or preservation, namely chemical treatment as well as environmental control, and a comparative analysis as part of a cost-benefit study could objectively justify such an intervention.

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