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Establishing the irradiation dose for paper decontamination

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ABSTRACT

Museums, libraries and archives are preserving documents that are slowly degrading due to the inherent ageing of the cellulose substrate or to the technological errors of the past (acid paper, iron gall ink). Beside this, large quantities of paper are rapidly damaged by biological attacks following natural disasters and improper storage conditions.

The treatment of paper documents with ionizing radiation can be used for mass decontamination of cultural heritage items but conservators and restaurators are still reserved because of the radiation induced degradation.

We conducted a study for establishing the dose needed for the effective treatment of paper documents, taking into account the biological burden and the irradiation effects on paper structure. We used physical testing specific to paper industry and less destructive analytical methods (thermal analysis).

Our results show that an effective treatment can be performed with doses lower than 10 kGy. Old paper appears to be less affected by gamma radiation than recent paper but the sampling is highly affected by the non-uniform degree of the initial degradation status. The extent of testing for degradation and the magnitude of acceptable degradation should take into account the biological threat and the expected life time of the paper documents.

1. Introduction

Paper is highly susceptible to the fungal, bacterial or insect attack. Due to calamities, lack of funding or care, large quantities of paper where improperly stored in archives, collections or libraries. Radiation processing was not widely accepted by the conservators and restaurateurs because of the well known radiolytic degradation of cellulose (Charlesby, 1955; Calvini and Santucci, 1978–79; Borsa et al., 2003) but there is an increasing interest to support the minimal intervention principle asked for the preservation of cultural heritage (Adamo et al., 2003; Rochetti et al., 2002; Magaudda, 2004; da Silva et al., 2006; Zotti and Calvini, 2008; D'Almeida et al., 2009; Bratu et al., 2009).

Most of the past treatments for paper items where made in an emergency procedure for saving the documents with less or no evaluation of the radiation effects (Hanus, 1985; Sinco, 2000; Dettino, 2007; Rela et al., 2007). Irradiation of archives is sporadically performed at many industrial irradiators and is rarely reported (Ponta, 2008). A related application is the irradiation of mail for prevention of bio-terrorism attacks (Baum, 2002; Smith et al., 2003; Bouchard et al., 2006; Zinov'ev et al., 2006). Here the dose is much higher (>50 kGy) and it is a true concern about the damage of historical documents that accidentally may be included in such treatment. The tests concerning the release of possible toxic volatile organic compounds (Smith et al., 2003) and the resistance of inks (Ramotowski and Regen, 2007) showed no toxic release from irradiated paper and a good radiation resistance of a large variety of inks.

We are reporting the radiation treatment of about 100 m^3 or paper items from archives, libraries and collections. Most of them were not under active biological attack but the treatment was required because the owners decided to improve the storage conditions.

The results presented in this paper was obtained in the frame of a larger study (Moise et al., 2010) initiated in Romania by "Horia Hulubei" National Institute for Physics and Nuclear Engineering

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(IFIN-HH) in cooperation with the Pulp and Paper Research Institute (CEPROHART SA), "Alexandru Darabont" Work Safety Institute and Braila Museum. These results were used for justification the radiation dose range for the treatment of IFIN-HH archive and will be used for the treatment of a museum collection belonging to Braila Museum.

2. Materials and methods

Three categories of papers where tested:

- Whatman paper (pure cellulose) which is widely used as reference in paper degradation studies.
- Contemporary copier type paper, which represents an example of complex mix of wood celluloses, additives and fillers.
- Papers from archives and collections, ranging from about 40 years to 80 years old.

Mechanical tests where performed with the purpose of evaluation of the radiation induced degradation on paper. Two main equipments where used:

- Universal testing machine Zwick-Roell Z005 with 5 kN cell was used for mechanical test of the paper at 25 mm/min – TAPPI T494 (2006) by IFIN-HH, IRASM Physical and Chemical Testing.
- Universal testing machine INSTRON 4411, 5 kN force cell, test speed: 20 mm/min – ISO 1924-2 by CEPROHART S.A., Accredited Pulp and Paper Testing Laboratory. The paper was tested also according to: ISO 534 (2005), ISO 536 (1995) ISO 2471 (2008), STAS 4748 (1995), ISO 2470-1 (2009), ISO 5627 (1995), ISO 2758 (2001), EN 21974 (1994), EN 20535 (1996), ISO 5626 (1993), ISO 11475 (2004).

Thermal analysis was performed for establishing correlations between the mechanical properties and parameters of paper thermal decomposition (Bratu et al., 2009). Netzsch STA 409 PC Luxx Simultaneous Thermal Analyzer with TG/DSC sample carrier was used for testing under inert atmosphere (IFIN-HH, IRASM Physical and Chemical Testing laboratory) Table 1.

Microbiological testing was performed in two experiments:

- Determination of bioburden (total aerobes by disintegration, filtration and culture on TSA), isolating representative strains and testing their radiation resistance (IFIN-HH)
- Evaluation of initial bioburden by two methods: total count (ISO 8784-1, 2005) and surface colony count (SR 13451, 2000).

The paper items used for the microbiological testing (Tables 2 and 4) where chosen randomly from 2 batches of IFIN-HH archive, excepting two books from 1952 and 1962 which was chosen from a very contaminated library collection of INCDPM.

The samples where irradiated in the SVST Co-60 gamma irradiator of IRASM Radiation Processing Center (IFIN-HH). The irradiator is a tote-box industrial type (10 m^3 maximum load) but experimental arrangements (static irradiation) allows dose rates between 0.3 and 10 kGy/h. Samples for physical tests where irradiated in the dose range: 0–30 kGy (Table 1). The range was chosen to overlap the dose range of interest (0–15 kGy) for the irradiation of cultural heritage items (Adamo et al., 2001; Gonzalez et al., 2002; D'Almeida et al., 2009). The dose was measured with ECB (ethanol–chlorobenzene) by oscillometric method.

Samples for evaluation of microbial radiation resistance where irradiated in the same irradiator in a lower dose range (for example 343 ± 11 Gy, 686 ± 22 Gy, 1029 ± 33 Gy, 1372 ± 44 Gy at 1.7 kGy/h). The dose measurements where made with EPR/alanine dosimetric system.

3. Results

The physical tests where performed on samples non-irradiated and irradiated in the three experiments (Table 1). The evolution with absorbed dose for tensile strength (force at break and elongation at break) are shown in Fig. 1. The evolution of folding endurance, tear and burst resistance is given in Fig. 2.

From Figs. 1 and 2 it can be seen that below 15 kGy it is difficult to quantify the irradiation effects. There is a net decrease of the most of the paper properties only for recent copy paper types and less obvious decrease for recent Whatman paper. For older and lower quality papers, the large spread of the results arises from the non-homogenous character of the paper amplified by the non-uniform degradation of the sampled material. The residual lignin content, which is present in certain sorts of wood pulp (Klemm et al., 1998), may contribute to this spread if lignin may act protectively in the process of cellulose radiolysis (Leliwa-Kania et al., 1998).

Table 1
Irradiation experiments.

Dose, kGy	6.0 ± 0.5	11.8 ± 1.0	24.5 ± 2.0	1.7 ± 0.1	4.2 ± 0.2	5.9 ± 0.4	9.9 ± 06	13.7 ± 0.8	2.9 ± 0.2	5.7 ± 0.5	8.4 ± 0.8	14.1 ± 1.2	28.2 ± 2.2
Paper samples	Copier paper 1 (80 g/m^2) Copier paper 2 (80 g/m^2)			Whatman paper (1 CHR) Copier paper 1 (80 g/m^2) Official Journal of Romania - 1968 (from IFIN-HH archive)					Whatman paper (1 CHR) Copier paper 1 (80 g/m^2) Forms printed in 1984 (not filled, from IFIN-HH archive) Notes-book, hand written in 1977 (from IFIN-HH archive)				
Dose rate, kGy/h	0.6			0.3					9.3				

Table 2
Bioburden on items from IFIN-HH archive.

Paper type	Journal 1968	Notes-book 1981	Forms 1983	Forms 1984	News-letter, 1983	Page from file 1	Page from file 2	Page from file 3	Average
CFU/g	25	115	20	60	140	140	95	125	**90**
CFU/100 pgs	9.5×10^3	4.4×10^4	7.6×10^3	2.3×10^4	5.3×10^4	6.7×10^4	5.0×10^4	6.1×10^4	$\mathbf{3.9 \times 10^4}$
CFU/1 m shelve	2.8×10^5	1.3×10^6	2.3×10^5	6.8×10^5	1.6×10^6	2.0×10^6	1.5×10^6	1.8×10^6	$\mathbf{1.2 \times 10^6}$
Cardboard type			File cover 1965			File cover 1965			Average
CFU/g			880			235			**558**