

# X-ray irradiation induced degradation of cellulose nitrate

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## Abstract

Energy-dispersive X-ray fluorescence (EDXRF) spectrometry was previously proposed to measure the thickness of the cellulose nitrate layer of the commonly used LR 115 solid-state nuclear track detector (SSNTD). The present work is devoted to the investigation whether the X-ray radiation involved in EDXRF spectrometry will induce degradation of the cellulose nitrate. For this purpose, Fourier transform infrared (FTIR) spectroscopy was employed to examine the nitrate functions (at the wavenumber  $1598\text{ cm}^{-1}$ ) and the glycosidic bonds (at  $1146\text{ cm}^{-1}$ ) for various irradiation time involved in the EDXRF spectrometry. No significant changes were observed even for X-ray irradiation up to 3000 live seconds, which was equivalent for 10 separate scans and should be far more than enough for a determination of the cellulose nitrate layer thickness. Therefore, EDXRF remains a fast and non-destructive method to measure the active layer thickness of the cellulose nitrate SSNTD.

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## 1. Introduction

The use of the cellulose nitrate (commercially known as LR 115) solid-state nuclear track detector (SSNTD), which is one of the most commonly used SSNTDs (see e.g., [1,2]), depends critically on the thickness of etched-away cellulose nitrate (CN) layer during chemical etching. However, Yip et al. [3] showed that its bulk etch rate was affected by the amount of stirring so actual monitoring of the thickness of the CN layer is necessary whenever using this SSNTD (e.g., [4]). In particular, so-called a priori non-destructive methods are needed before completion of chemical etching [5].

Yip et al. [5] proposed to monitor the CN layer by using energy-dispersive X-ray fluorescence (EDXRF) spectrometry. For that method, a reference silver nitrate pellet was placed beneath the SSNTD, and the fluorescence X-ray intensity for silver was then measured. A linear relationship between the X-ray intensity and the thickness of the CN layer was found, which provided a fast method to measure the latter. However, there is a risk that the X-ray radiation degrades the CN [6,7]. Such degradation is critical because its occurrence means EDXRF is destructive to the CN layer and will therefore defeat the whole idea in using it to monitor the CN layer.

The present work is devoted to the investigation whether the X-ray radiation involved in the EDXRF spectrometry will induce significant degradation of CN. For this purpose, Fourier transform infrared (FTIR) spectroscopy will be employed to examine the nitrate functions and the glycosidic bonds for various sampling time for the EDXRF spectrometry. These two aspects were examined by Barillon and Yamauchi [8] in order to quantify the chemical damages in LR 115 detectors.

## 2. Experiments

### 2.1. Cellulose nitrate SSNTDs

The CN SSNTDs used in the present study were purchased from DOSIRAD, France (LR 115 film, Type 2, strippable). The SSNTDs consist of a red layer of CN on a  $100\text{ }\mu\text{m}$  clear polyester base substrate. For the purpose of our experiments, only the CN layer is required, so the SSNTD will hereafter refer only to the CN layer stripped from the polyester base.

### 2.2. EDXRF

Our EDXRF experiment follow the setup outlined by Yip et al. [5]. The idea was to detect the EDXRF  $L_{\alpha}$  line intensities for Ag (energy = 2.984 keV) present in a silver nitrate pellet as a substrate placed beneath the SSNTD. The element Ag was chosen so that the characteristic fluorescent X-ray energy is high enough to avoid total absorption by the CN layer but is also low enough to be sufficiently sensitive to the thickness of the CN layer.

The silver nitrate pellet contained 5 g of silver nitrate powder (chemical formula:  $\text{AgNO}_3$ ; purity: 99.8%; manufacturer: Panreac Quimica SA; code:

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131459) and 0.25 g of a blender (from Chemplex<sup>®</sup>, #600) for better binding properties. The materials were homogenized, and then formed into stable pellets using a SpectroPress<sup>®</sup> Systems press (Model no.: Automatic 50 TON PRESS, Chemplex Industries, Inc., USA) with die diameter of 1.25 in. (32 mm) and the following settings: force = 30 t, dwell time = 3 min and bleed time = 4 min. The pellet was placed beneath the CN SSNTD.

In the present experiments, actual measurements of the EDXRF  $L_{\alpha}$  line intensities for Ag are not necessary; only the irradiation part is relevant since we would only like to investigate whether the X-ray radiation involved will induce significant degradation of the CN SSNTDs. However, the irradiation parameters should be the same as those adopted for the actual measurements, with the silver nitrate pellet placed beneath the SSNTD. The EDXRF spectroscopy was carried out under vacuum using an EDAX International DX-95 EDXRF spectrometer with a Mo target, equipped with a liquid nitrogen cooled Si(Li) detector for a specified period of time (in terms of live seconds). The maximum power of the instrument was 25 W. The incident and take-off angles were 45°, with a Be window thickness of 12.5  $\mu\text{m}$ . The distance between the sample (exposed diameter of 22 mm) and the detector was 4.5 cm. The X-ray tube voltage was set as 10 kV while the beam current was set as 200  $\mu\text{A}$ . The irradiation time ranged from 0 live seconds (i.e., without irradiation) to 3000 live seconds, in steps of 300 live seconds.

### 2.3. FTIR spectroscopy

After each X-ray irradiation, the chemical changes in the CN were quantified using a Perkin-Elmer (Model 16 PC FT-IR) Fourier transform infrared spectroscopy system. In each measurement, an SSNTD was scanned for 10 cycles. The wave number range employed was between 1700 and 1100  $\text{cm}^{-1}$ . The scanned diameter was 9 mm so the scanned area was 0.64  $\text{cm}^2$ . A spectrum showing the variation of the infrared transmittance (%) with the wave number ( $\text{cm}^{-1}$ ) would be generated by the FTIR system after scanning. The relative changes in the transmittance at the wavenumber 1598  $\text{cm}^{-1}$  due to changes in the nitrate functions [8–10] and at 1146  $\text{cm}^{-1}$  due to changes in the glycosidic bond (C–O–C) [8] were employed to quantify the chemical changes induced by the X-ray irradiation.

### 3. Results and discussion

The infrared transmittances (%) in the wavenumber range from 1100 to 1700  $\text{cm}^{-1}$  for different X-ray irradiation time are shown in Fig. 1. There are in fact a total of 11 spectra in

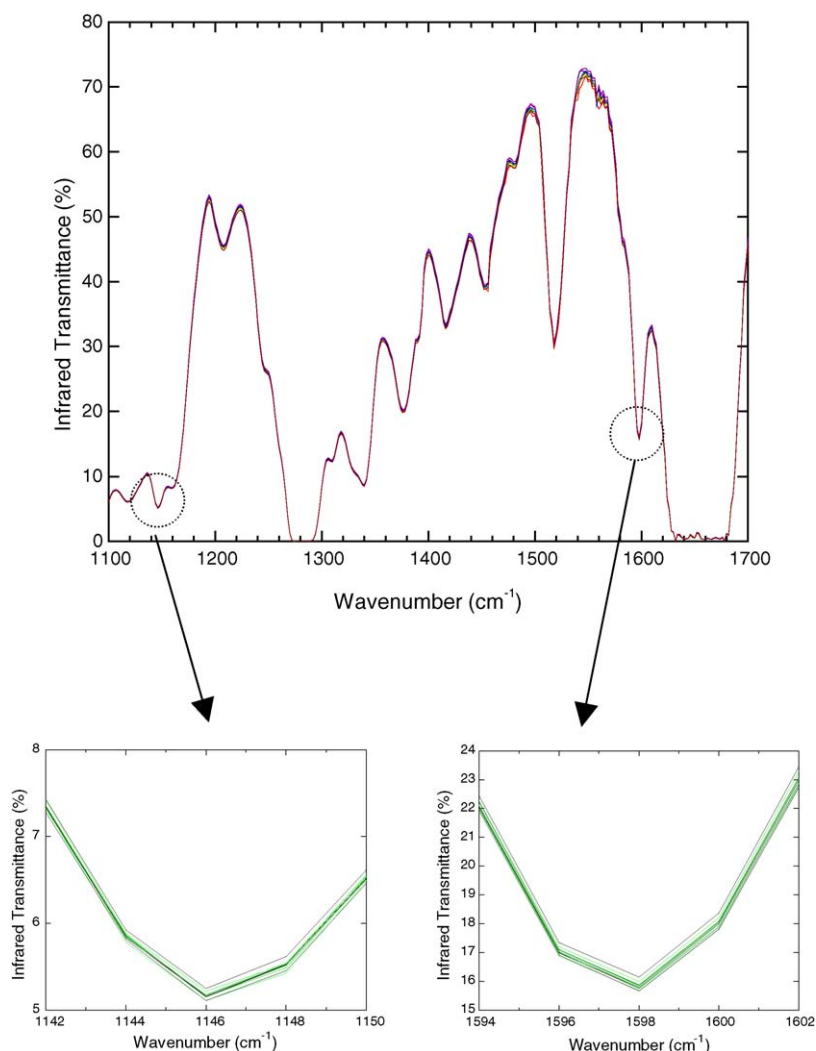


Fig. 1. *Upper*: Infrared transmittances (%) in the wavenumber range from 1100 to 1700  $\text{cm}^{-1}$  for different X-ray irradiation time. There are a total of 11 spectra corresponding to irradiation time ranging from 0 live seconds (i.e., without irradiation) to 3000 live seconds, in steps of 300 live seconds. *Lower left*: Enlargements of the region around the wavenumber 1598  $\text{cm}^{-1}$ . Dotted lines represent curves for irradiation time ranging from 0 to 1500 live seconds while solid lines represent curves for irradiation time ranging from 1800 to 3000 live seconds. *Lower right*: Enlargements of the region around the wavenumber 1146  $\text{cm}^{-1}$ . Dotted lines represent curves for irradiation time ranging from 0 to 1500 live seconds while solid lines represent curves for irradiation time ranging from 1800 to 3000 live seconds.

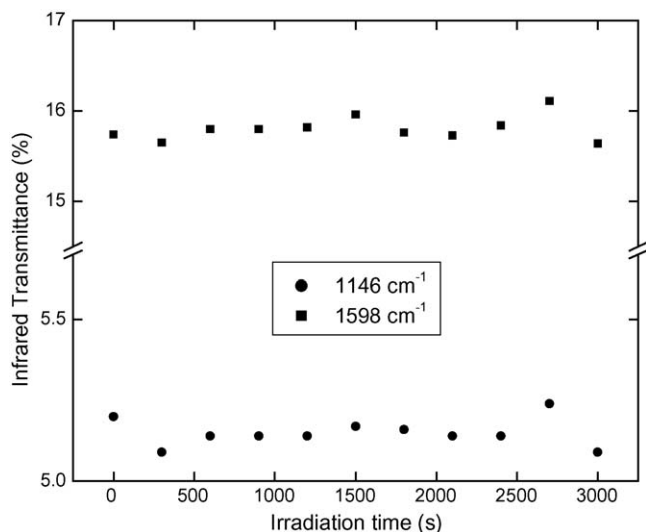


Fig. 2. The infrared transmittances (%) at the wavenumbers 1598 and 1146  $\text{cm}^{-1}$  for different X-ray irradiation time.

Fig. 1, corresponding to irradiation time ranging from 0 live seconds (i.e., without irradiation) to 3000 live seconds, in steps of 300 live seconds. However, the curves are almost identical. Even enlargements of the region around the wavenumbers 1598 and 1146  $\text{cm}^{-1}$  do not show significant differences among the curves. For easier comparison, the transmittances (%) at the wavenumbers 1598 and 1146  $\text{cm}^{-1}$  for different X-ray irradiation time are shown in Fig. 2. From Figs. 1 and 2, we can see that the X-ray irradiation involved in the EDXRF spectrometry will not induce degradation of CN even up to 3000 live seconds. Considering that each EDXRF spectrometry to determine the CN layer thickness requires only 300 live seconds [5], there will not be observable chemical changes in the CN SSNTD even after 10 scans, which is far more than enough for a determination of the CN layer thickness.

In conclusion, the method proposed by Yip et al. [5] using energy-dispersive X-ray fluorescence spectrometry remains a fast and non-destructive a priori method to measure the thickness of the cellulose nitrate layer of the LR 115 detector.

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