Studying radiolytic ageing of nuclear power plant electric cables with FTIR spectroscopy

A. Levet\(^a,b\), J. Colomban\(^b,⁎\), L. Duponchel\(^b,⁎\)

\(^a\) IRSN/PSN-RES, Cadarache, St Paul lez Durance 13115, France
\(^b\) LASIR CNRS UMR 8516, Université Lille 1, Sciences et Technologies, 59655 Villeneuve d’Ascq Cedex, France

**ABSTRACT**

Due to the willingness to extend the nuclear power plants length of life, it is of prime importance to understand long term ageing effect on all constitutive materials. For this purpose gamma-irradiation effects on insulation of instrumentation and control cables are studied. Mid-infrared spectroscopy and principal components analysis (PCA) were used to highlight molecular modifications induced by gamma-irradiation under oxidizing conditions. In order to be closer to real world conditions, a low dose rate of 11 Gy h\(^{-1}\) was used to irradiate insulations in full cable or alone with a dose up to 58 kGy. Spectral differences according to irradiation dose were extracted using PCA. It was then possible to observe different behaviors of the insulation constitutive compounds i.e. ethylene vinyl acetate (EVA), ethylene propylene diene monomer (EPDM) and aluminium trihydrate (ATH). Irradiation of insulations led to the oxidation of their constitutive polymers and a modification of filler-polymer ratio. Moreover all these modifications were observed for insulations alone or in full cable indicating that oxygen easily diffuses into the material. Spectral contributions were discussed considering different degradation mechanisms.

**1. Introduction**

Ageing of structure materials and components of Nuclear Power Plants (NPP) is a hot topic considering the willingness to extend their life time operation beyond 40 years. Indeed instrumentation and control (I & C) cables, located in the reactor building, have to endure containment conditions. Moreover they are often difficult, if not impossible, to replace. They are naturally considered as key elements for nuclear safety. During the NPP lifetime, I & C cables have to ensure electricity transmission function. In normal operating conditions, these cables are exposed to about 0.1 Gy h\(^{-1}\) dose rate at a temperature below 50 °C and a humidity level of 70%. In the case of a nuclear accident, they have to withstand a temperature and pressure increases (up to 150 °C and 5.6 bar respectively). Similarly, dose rate increases with the release of fission products. Electrical functionality of these I & C cables is strongly dependent on the ageing of their insulations. The study of the radiolytic ageing of these polymeric materials is therefore a major concern from a nuclear safety point of view. The ageing of polymers affects different properties of the material at the physical, chemical and mechanical level. Many techniques have been used to investigate all these modifications such as tensile tests [1–3], gel fraction measurement [4,5], thermogravimetry [3,4], \(^{13}\)C Nuclear Magnetic Resonance (NMR) analysis [2,6], Differential Scanning Calorimetry (DSC) [3,6]. Chemical modifications are usually studied with mid-infrared spectroscopy because of its ease of use and the richness of molecular information [2,3,5–13]. Thus different strategies can be used in order to extract and observe potential modifications in spectra. It can be as simple as directly comparing spectra of aged materials with original ones. However it is not a very effective method because very small spectral modifications have often observed. A more complicated method is the use of a chemical derivatisation treatment that selectively converts oxidation products into groups with a different infrared (IR) absorption [8]. For the sake of simplicity and in order to have a direct analysis of modified samples we propose in this paper to investigate potential cable modifications with Fourier transform Infrared spectroscopy (FTIR) spectroscopy combined with the well-known Principal Component Analysis (PCA) which is the Swiss army knife of multivariate analysis. This simple but efficient technique has been successfully used in different domains such as for example in medicine [14], food science [15,16], forensic [17], pharmacy [18], polymers [19]. Specifically the present work deals with radiolytic ageing of halogen free cable insulation used in French nuclear power plants. The ageing effect on constitutive materials of cable insulation such as ethylene vinyl acetate (EVA), ethylene propylene diene mono-
mer (EPDM) and aluminium trihydrate (ATH) have been studied for each component separately in different works [2–9,12,13,20,21]. For all these cases, modifications have been caused by irradiation, ionization or thermal effect. To our knowledge, for the first we will explore simultaneously the radiolytic ageing of all compounds presents in insulation cables.

2. Materials and methods

2.1. Samples

The sample of interest is the halogen free insulation of NEXANS instrumentation and control (I & C) cable (2013) (Fig. 1). It is mainly composed of three compounds: EVA, EPDM and ATH filler. EVA is a copolymer of ethylene (86%) and vinyl acetate (14%). EPDM is a copolymer of ethylene (85%), propylene (10%) and 5-ethylene-2-norbornene (ENB) (5%). Aluminium trihydrate is a fire-retardant mineral filler.

2.2. Irradiation conditions

The objective of this work is to study degradation of insulation cable after a long time of gamma-irradiation exposition. In general, it is estimated that the insulations are exposed to a cumulative dose of 45 kGy after 50 years of use in normal operating conditions. Obviously, we cannot wait such a long time in order to obtain samples. One can also imagine that removing electric cables in a nuclear power plant is not a trivial task. Furthermore, it is necessary to observe cables with different irradiation levels if we really want to understand potential molecular transformations. Thus an irradiation chamber is used in order to apply accelerated ageing conditions. Typically, dose rates used to accelerate ageing goes from several hundred Gy h⁻¹ up to several kGy h⁻¹. In this study, irradiation was performed at a low dose rate of 11 Gy h⁻¹ as a compromise between the 0.1 Gy h⁻¹ dose rate usually observed in normal operating conditions of a nuclear power plant and the time needed to have a high enough integrated dose. Samples were irradiated by gamma rays from a 60Co gamma-irradiation source at ambient temperature (−35 °C). In order to induce more oxidative degradations and accelerate the ageing by a chemical way, low concentration of acetic acid was introduced in the chamber. Indeed acetic acid is a well-known compound produced by polymer radiolysis which may play a role in ageing phenomena. Irradiation was performed on full cable (i.e. with the insulation irradiated inside of the internal and external jacket) or directly on the insulation alone (i.e. extracted from the cable). These two conditions were studied in order to observe the potential influence of oxygen in direct contact with the insulation or potential limitation due to the full cable design. In this paper, the extent of chemical evolution produced by gamma-irradiation depends on the amount of energy absorbed by the system. The absorbed dose is expressed in Gray (Gy) which corresponds to an absorbed energy of 1 J kg⁻¹ of matter. Thus the samples have been exposed to the doses of 7 kGy, 21 kGy, 48 kGy, 52 kGy and 57 kGy. All these levels of exposition were checked with dosimetry using the alanine EPR method [22] or Perspex films method [23].

2.3. Spectroscopic measurement

Mid-infrared spectra were recorded with a Nicolet ISS50 FT-IR spectrometer using Attenuated Total Reflectance (ATR) mode. The contact between analyzed sample and the diamond of the ATR accessory was ensured by screwing a clamp device. Only the external side of the insulation was explored. A total of 32 scans were co-added to generate a spectrum with a 4 cm⁻¹ spectral resolution on the 400–4000 cm⁻¹ spectral range. Insulation sample was cleaned with ethanol prior FTIR analysis in order to remove potential impurities on its surface. The single beam spectrum of the clean and dry ATR element was used as background. Three spectra of each sample were recorded on three different days. All of these spectra were directly used for chemometric analysis in order to be able to distinguish variance of the triplicates from variances of potential chemical modifications.

2.4. Data analysis

Principal Component Analysis is a chemometric method transforming a dataset with correlated variables (absorbances at various wave-numbers in our case) into a new one with uncorrelated variables called Principal Components (PCs). PCs are known to be linear combinations of initial variables [24]. It is thus possible to represent all spectral information in a low dimensional space. Each spectrum is then represented by scores which are new coordinates in this new space. Samples of a data set are usually observed in scores plot considering two selected principal components. In this way, points (i.e. samples) located together in the graph share same characteristics. On the contrary a high distance between points highlights much dissimilarity. For its part, the observation of a principal component represented in the original space allows to show spectral zones which are taken into account. Spectral preprocessing is a common way to suppress unwanted variances and thus really analyze chemical modifications only. It is never easy to find the optimal combination of preprocessing which is often chosen on a subjective basis. In our case, the lowest observed variance between replicates of all samples in PCA was a good way to select the best suited preprocessing methods. Thus a first order Savitzky-Golay derivative [25–27] (2nd order polynomial, 3-point fitting window) and Standard Normal Variate correction [27,28] where selected. All chemometric calculations were performed using the Eigenvector Solo software, version 8.1.1 (Eigenvector Research Inc, Wenatchee, WA, USA).

3. Results and discussion

Before analyzing the effect of gamma-irradiation on insulation cables, it is important to interpret spectral contributions we can retrieve with mid-infrared spectroscopy considering a non-irradiated sample (Fig. 2). An initial visual inspection allows to highlight the richness of molecular information for the considered multi-compound material. Table 1 summarizes assignment of the main absorption bands of the non-irradiated insulation. Many overlaps are observed mainly due to the complexity of the material and the naturally high bandwidth of mid-infrared spectroscopy. Absorption bands located around 3500 cm⁻¹ are assigned to the mineral filler Al(OH)₃. A specific contribution of EVA is observed at 1740 cm⁻¹ corresponding to the carbonyl function of the vinyl acetate. Various bands can also be assigned to both EVA and EPDM polymers at 1466 cm⁻¹, 2848 cm⁻¹ and 2916 cm⁻¹ corresponding to ethylene chains contributions. The fingerprint region always contains many absorption bands which stay
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