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THERMAL DECOMPOSITION OF RADIO-OXIDIZED POLYMERS

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Introduction

Many polymers are used in nuclear industry and are then stored in nuclear waste packages. In case of accidental conditions of transportation, the temperature of packages may increase and reach 150°C. The accumulation of the gases formed may affect the integrity of the waste container. Therefore, for safety purpose, it is important to predict the gas quantity released by different organic materials and measure the kinetic of thermal decomposition of radio-oxidized polymers [1] [2] [3].

The purpose of this work is to present the results of a multi-year study performed on four polymers (polyethylene, cellulose, polyurethane, polyvinyl chloride). The effect of dose, atmosphere and degradation temperature on the quantity of gas release was investigated.

Experimental

High density polyethylene (Sigma), cellulose (commercial cotton), polyurethane (Plastunion) and plasticized polyvinyl chloride (Plastunion) were gamma-irradiated under air atmosphere at room temperature at different doses (0, 1, 2 and 4 MGy). After irradiation, the thermal degradation of polymers at different temperatures (60°C, 80°C, 100°C, 120°C and 150°C) for 48 hours in air or inert conditions was analyzed by thermogravimetric analysis (TGA) and infrared spectroscopy. In addition, in order to quantify the volatiles species a known amount of polymer was conditioned in a small vessel (made on stainless steel or glass) equipped with a pressure gauge. The pressure build-up was recorded as a function of the time during the thermal treatment. An example of the pressure measurement is shown in Figure1 for the pre-irradiated polyvinyl chloride, at 4 MGy, heated at different temperatures for 48 hours under air. Identification and quantification of gases were performed by high resolution mass spectrometry and gas chromatography coupled with mass spectrometry (GC-MS). For the experiments under air, oxygen consumption was also measured.

Results and discussion

Even at 150°C, non-irradiated polymers were rather stable. The results are different for radio-oxidized materials. A rapid increase of the pressure is observed during the first hours after heating followed by a stabilization or a slow increase. For all polymers, the production of volatile species is almost proportional to the dose and is thermally activated. The main degradation products are water and carbon dioxide. The production of hydrogen and inflammable gases is rather limited. Under air atmosphere, the pressure increase is partially compensated by oxygen consumption.

Conclusions

A comprehensive study performed on different polymers representative of organic materials present in nuclear waste shows that the thermal decomposition of pre-oxidized polymers is a rapid phenomenon. The majority of the gases is released in the first hours. Water and carbon dioxide are the predominant species. Although irradiation reduces the thermal stability of the polymers, our results show that the risk of inflammation is not exacerbated.

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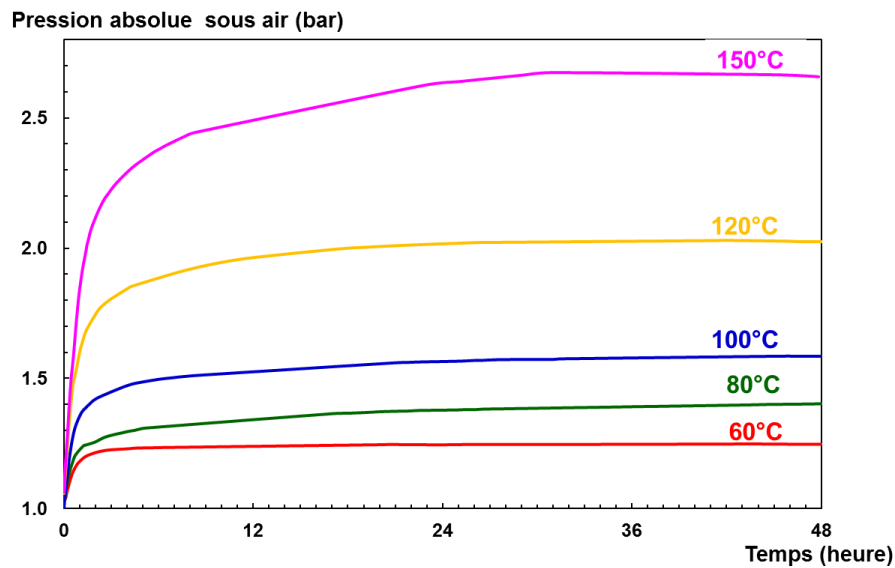


Figure 1 – Pressure increase measured for polyvinyl chloride gamma-irradiated at 4 MGy under air heated at different temperatures for 48 hours under air.

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