

Radiation induced grafting of acrylic acid onto extruded polystyrene surface

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ABSTRACT

Polystyrene materials with good solubility in liquid scintillation cocktails are used to wipe off different types of surfaces in order to determine the tritium removable contamination with the help of a liquid scintillation counter. This paper analyses hydrophilic surface modifications by radiation induced grafting of acrylic groups onto extruded polystyrene plates. Two grafting methods were used: (a) exposure of extruded polystyrene plates, immersed in aqueous acrylic acid solution, to a gamma radiation of a Co-60 source, and (b) exposure of extruded polystyrene plates to a Co-60 source, followed by the immersion of extruded polystyrene plates in aqueous acrylic acid solution. The grafting of acrylic was proved by IR spectrometry and by radiometric methods using acrylic acid labelled with tritium.

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1. Introduction

Removable surface contamination (RSC) represents the main risk factor for tritium laboratory workers. In case of low β energy radionuclides like tritium, RSC is determined by wiping off contaminated surfaces with absorbent material smears and by measuring the radioactivity collected using a liquid scintillation counter. The polystyrene is recommended for this application because it has a good solubility in liquid scintillation cocktails (Postolache and Matei, 2005). Hydrophobic polystyrene properties imply a low sampling factor, and in order to increase the RSC sampling factor, grafting hydrophilic fragments onto polystyrene surface may be an option.

In these experiments acrylic acid (AAc), which has a pronounced hydrophilic character due to the presence of carboxylic groups, has been grafted onto extruded polystyrene (EPS) by means of gamma irradiation, due to vinyl group of polymeric matrices.

Mainly, the study investigated superficial grafting of AAc fragments onto EPS surface.

The presence of carboxylic fragments grafted in smears may induce a significant decrease of liquid scintillation counting efficiency as a result of the quenching phenomenon. For this reason, it was necessary to study the influence of AAc mass percentage grafted on the scintillation counting efficiency.

2. Materials and methods

2.1. Radiation induced grafting of acrylic acid onto EPS surface

In this experiment, two grafting methods were used:

- Exposure of EPS plates, immersed in acrylic acid aqueous solution, to a gamma radiation source (Co-60);
- Exposure of EPS plates to a Co-60 source followed by the immersion of EPS plates in aqueous acrylic acid solution.

2.1.1. Radiation induced grafting of acrylic acid

2.1.1.1. Irradiation of EPS plates immersed in aqueous acrylic acid solution. The EPS plates (mass $60 \text{ mg} \pm 10\%$) were immersed in the grafting solution having the following composition: 30% AAc, 1% CuSO_4 and 69% purified water (Pietzak, 1995).

Impregnation (intrusion) of grafting solution inside the material's pores was achieved by repeated vacuum-pressure operations. Subsequently, the samples were irradiated with gamma radiation from a Co-60 source at dose rates of 3, 5, and 10 kGy/h. The absorbed doses were up to 250 kGy.

2.1.1.2. Preirradiation of EPS plates before immersion in aqueous acrylic acid solution. The EPS plates were exposed to gamma radiation in the air, to an absorbed dose of 250 kGy. After irradiation, the EPS plates were immersed in acrylic acid solution, with the composition presented in the previous paragraph, for 24 h.

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2.1.2. Radiation induced grafting of acrylic acid labelled with tritium
Tritium labelled acrylic acid was grafted onto EPS plates, using either the simultaneous irradiation or the preirradiation method presented, by replacing the AAC with tritium labelled AAC (Postolache and Matei, 2007).

After grafting, the EPS plates were subjected to the following treatment: three times washing with purified water, storage for 2 h in 1% EDTA solution in order to remove Cu^{2+} ions, washing with purified water, washing with methyl alcohol p.a. grade and finally vacuum drying at 10^{-2} mPa.

2.2. Characterisation of radiation induced grafting of acrylic acid

2.2.1. Grafting yield

Grafting yield is defined as grafted acrylic acid mass percentage on EPS plate and was determined by two methods: gravimetric and radiometric.

Grafting yield (G_{graf}) was determined gravimetrically by measuring the initial and post-grafting mass of EPS plate, using a balance PRECISA XT120A type with ± 0.1 mg precision, by the relation:

$$G_{\text{graf}}[\%] = \frac{m_{\text{AA}}}{m_{\text{EPS}}} 100 = \frac{m_{\text{graf}} - m_{\text{EPS}}}{m_{\text{EPS}}} 100 \quad (1)$$

where: m_{AA} is grafted AAC mass, m_{EPS} is EPS plate initial mass, and m_{graf} is EPS disc grafted mass.

The mass of grafted acrylic fragments was also determined by a radiometric method using as support a tritium labelled AAC. In this case, the grafting yield (G_{graf}) has been determined by the equation:

$$G_{\text{graf}}[\%] = \frac{m_{\text{AA}}}{m_{\text{EPS}}} 100 = \frac{A/A_{\text{sp}}}{m_{\text{EPS}}} = \frac{A}{A_{\text{sp}} m_{\text{EPS}}} 100 \quad (2)$$

where A is the Tritium labelled AA grafted disc activity, A_{sp} the masses activity with 37 kBq/mg (1 $\mu\text{Ci}/\text{mg}$) value, m the initial mass of EPS plate.

2.3. Grafted EPS samples characterisation by IR spectrometry

The radiation induced grafting of acrylic fragments onto EPS plate surfaces has been analysed using IR spectrometry. In this study an FTIR ATR spectrometer Bruker TENSOR 27 type was used. The IR spectra for grafted EPS samples and for reference samples (EPS and polyacrylic acid) were collected and compared.

2.4. Evaluation of the quenching effect induced by grafting acrylic acid onto EPS

Quenching was evaluated by measuring the difference between counting rate of tritiated liquid cocktails and grafted EPS plates dissolved into tritiated liquid cocktails.

Counting rates before and after the dissolving of 1 and 5, respectively, grafted EPS plates into tritiated liquid cocktails were determined using the Liquid Scintillation Counter TRICARB 2800 TR.

2.5. Removable surface contamination (RSC) sampling factor determination

The RSC sampling factors were determined for representative surfaces from radiochemical laboratory: stainless steel 316 L type, aluminium, glass, ceramics plates, linoleum, and ALOREX LP 52 epoxy resin.

For each surface type, an area of 100 cm^2 was contaminated, in a controlled manner, with testosterone-1, 2-T toluene solution or

with nucleoside analogue U34-5-T aqueous solution (Postolache et al., 2007).

The contamination was carried out by sampling, using a stainless steel grill, $10 \mu\text{L}$ labelled compound solution for each square centimetre of analysed surface. Determination of the sampling factor was achieved by wiping the contaminated surface with smears moisten in $50 \mu\text{L}$ butyl alcohol, followed by activity measuring using the Liquid Scintillation Counter TRICARB TR 2800 type.

Sampling factor was determined as the ratio between measured activity and real conventional activity initially deposited on the surface.

3. Results and discussion

3.1. Radiation induced grafting yields determination

Quantitative analysis of grafting process were performed by grafting yield (G_{graf}) determination in relation with absorbed doses, dose rate and grafting method (Fig. 1) The results suggest a linear dependence of the grafting yields in relation with the absorbed dose.

No saturation effect has been observed up to an absorbed dose of 250 kGy.

3.2. Sample characterisation by IR spectrometry

The IR spectra of reference and AAC grafted EPS samples are shown in Figs. 2 and 3. In the reference EPS sample, the typical peaks assigned to the aromatic C–H bonds of benzene rings present in the polystyrene were observed at in 3010 , 1600 and 1475 cm^{-1} . In the polyacrylic acid reference sample we have identified: OH characteristic bond in $3000\text{--}3800 \text{ cm}^{-1}$ area, C–H

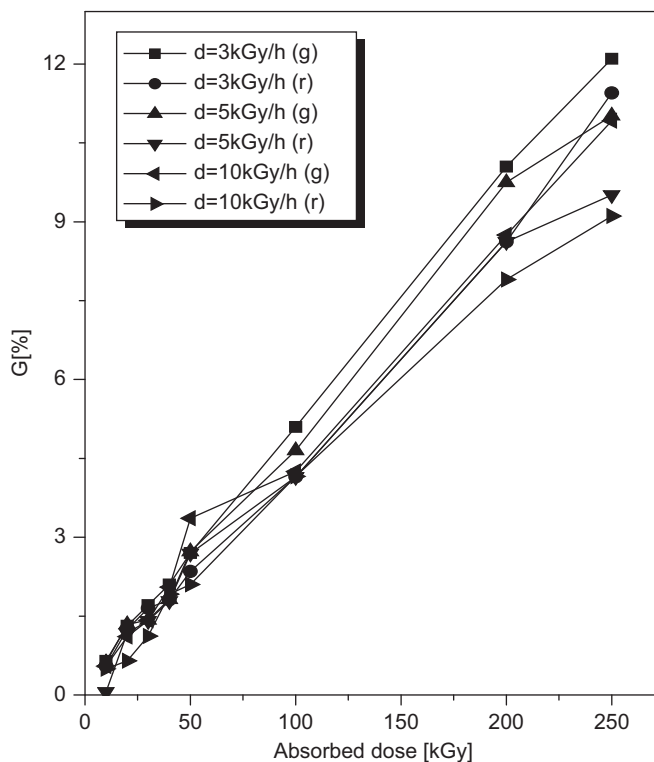


Fig. 1. Gravimetric (g) and radiometric (r) obtained radiochemical yield (G_{graf}) in relationship with absorbed dose (D) and dose rate (d).

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