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Removal naturally occurring radionuclides from drinking water using a filter specifically designed for Drinking Water Treatment Plants

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HIGHLIGHTS

- Filter system (green + silica sand) especially designed to remove radium was characterized.
- Mn in green sand was homogenously distributed (FE-SEM and μ-PIXE).
- Ra adsorption kinetics adsorb 99% from raw water.
- Radium removal efficiency remained unchanged after passing a large water volume.
- It can also remove partially the uranium content.

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ABSTRACT

The occurrence of naturally occurring radionuclides in drinking water can pose health hazards in some populations, especially taking into account that routine procedures in Drinking Water Treatment Plants (DWTPs) are normally unable to remove them efficiently from drinking water. In fact, these procedures are practically transparent to them, and in particular to radium. In this paper, the characterization and capabilities of a patented filter designed to remove radium from drinking water with high efficiency is described. This filter is based on a sandwich structure of silica and green sand, with a natural high content manganese oxide. Both sands are authorized by Spanish authorities to be used in Drinking Water Treatment Plants. The Mn distribution in the green sand was found to be homogenous, thus providing a great number of adsorption sites for radium. Kinetic studies showed that the ^{226}Ra adsorption on green sand was influenced by the content of major cations solved in the treated water, but the saturation level, about 96–99%, was not affected by it. The physico-chemical parameters of the treated water were unaltered by the filter. The efficiency of the filter for the removal of ^{226}Ra remained unchanged with large water volumes passed through it, proving its potential use in DWTP. This filter was also able to remove initially the uranium content due to the presence of Fe_2O_3 particles in it, although it is saturated faster than radium.

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1. Introduction

The content of naturally occurring radionuclides may become an increasingly serious problem for drinking water supplies, due to the growing demand for new water resources (IAEA, 2003). Unless the corresponding water treatment systems are appropriately designed, a significant part of this radioactive content can eventually be incorporated into the population, with the consequent risk to health. In areas unaffected by radioactive contamination events, such as those of Chernobyl or Fukushima, naturally occurring radionuclides are the main contributors to the Total Indicative Dose (TID) for the consumption of drinking water. Their occurrence can be high, mainly caused by the geological environment surrounding the water source (Otton, 1994; Baeza et al., 1995). As the routine procedures in Drinking Water Treatment Plants (DWTPs) have practically no effect on the removal of uranium and radium from drinking water (Baeza et al., 2012), they are ingested by the population as they drink. Anthropogenic causes, such as the

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age and maintenance of pipelines in public water supplies, can also affect the radium content in drinking water, implying a health hazard to population (Fisher et al., 2000). The chemical toxicity of soluble uranium compounds in drinking water can surpass its potential low radiotoxic effects. The exposure to ingested uranium includes changes to bone structure and nephritis (Kurttio et al., 2002, 2005; Ansoborlo et al., 2015). Information on radium content in drinking water is particularly important as radium has a similar metabolic behavior as calcium in the body. An appreciable fraction of the radium acquired through drinking water is present in bones, where it contributes to the internal dose for all population (UNSCEAR, 1982; Tyler et al., 2013).

The use of specific substances for removing uranium and radium in water has often been studied from different approaches. A number of chemical methods were developed by adding substances to raw water in order to produce precipitation or co-precipitation of radionuclides. Such methods work very well for removing uranium (Baeza et al., 2012), but for the radium require very aggressive conditions in practice difficult to apply (Valentine et al., 1985; Baeza et al., 2008). Other methods are based on the use of specific materials, such as resins and adsorbents which produce a large adsorption. Examples of these substances would be the MnO_2-Coated Acrylic Fiber Filters (Moore, 1978), Radium Selective Complexer (DOW, 1986), $BaSO_4$-impregnated alumina (Clifford et al., 1988) and silica sand (Valentine et al., 1985). Silica sand is a material used as a filter bed to retain small solids that are in the water. It is usually employed as a filter bed in water treatment plants. Its ability to adsorb radium depends on working conditions and has many limitations. All methods above described require treatment of the adsorbent material for removal of radium, and in some cases the associated cost is high. Greensand is a natural manganese dioxide, commonly employed in removing iron and manganese from water through a filtration process, and capable to remove radium (Qureshi and Nelson, 2003). Soluble iron and manganese are oxidized and precipitated by contact with major oxides of manganese in greensand granules.

In this paper, the characterization of a patented filtration system for the removal of radium in water for human consumption is presented (patent publication number ES2564566A1). This system uses two types of natural substances, silica sand and greensand, which are permitted for use in drinking water. Thus, the elemental composition of the green sand was mapped using qualitative FE-SEM (Field Emission Scanning Microscopy) and quantitative μ-PIXE (Micro Particle Induced X-ray Emission). The radium adsorption was analysed by means of different experiments focused on: a) studying its kinetics on green sand; b) the variables affecting its adsorption in silica sand. Finally, experiences aimed to determine its use for treatment of large water volumes were also carried out.

2. Material and methods

2.1. Design and characterization of the filter

The filter was designed using adsorbing materials authorized by the Spanish Health Ministry to be used in water purification processes (BOE, 2003; BOE, 2009). It consists in a mixture of silica and green sand in a sandwich structure (1-4-2), as shown in Fig. 1. The green sand has a high Mn content, thus providing adsorption sites for radionuclides, especially for radium. The difference of this filter with other existing in the market are: i) it is radium specific, ii) its components are already approved by Spanish legislation to be used in DWTP, while some other resins are not authorized, iii) it is based in a sandwich structure, while other filters use only one component. The silica sand in the filter has a double utility. It holds the green sand and provides an extra filter bed in order to prevent drinking water from taking any colour and avoiding obstructions.

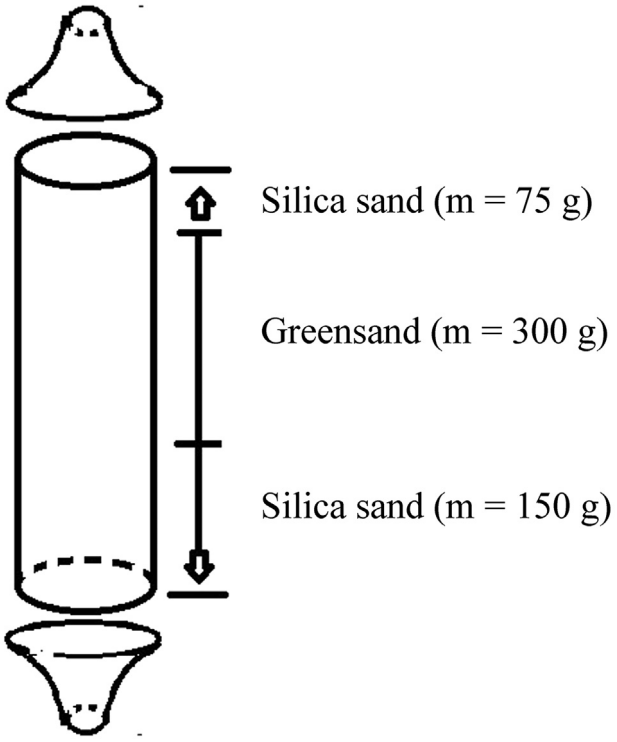

Fig. 1. Schematic diagram of the filter, with three layers: silica sand, green sand, and silica sand in 1-4-2 proportion.

The elemental composition of the green sand was analysed in detail by a qualitative analysis by FE-SEM, using 30 keV electron beam. Nevertheless, quantitative μ-PIXE analysis was also performed. For the preparation of the samples, about 0.2 g of sample pressed into pellets of 11 mm of diameter on a boric acid substrate about 0.4 g using a pneumatic press (10 Ton). The measurements were performed at the Centro Nacional de Aceleradores micro-PIXE facility (García-López et al., 2000). A proton beam of 3.0 MeV energy, a current of 100 pA, and a spot size of $4 \times 4\ \mu m^2$ was produced normal to the sample. A Si(Li) detector (area 80 mm^2, resolution 145 eV) was mounted at 135° for X-ray detection. For all measurements, a 50 μm thick Mylar filter was used between the sample and the Si(Li) detector improve the sensitivity of trace elements. Proton backscattered spectra (BS) were collected simultaneously with a surface barrier detector of an active area of 300 mm^2 at an angle of 37° to the beam. Samples were sprayed with low-energy electrons originated from a hot tungsten filament in order to avoid sample charging during the analysis. All signals produced during the proton beam irradiation were recorded together with the beam position using the OM_DAQ data acquisition system (Grime and Dawson, 1995). Elemental maps up from $25 \times 25\ \mu m^2$ to $150 \times 150\ \mu m^2$ were generated using the scanning mode in order to determine element distribution and to select the most convenient areas of analysis. Quantification was performed with the GUPIX-WIN V2.1 software package (Campbell et al., 2010).

2.2. Design of laboratory experiences

The efficiency of the filter to remove naturally occurring radionuclides, mainly uranium and radium, was tested in laboratory controlled conditions, by passing a known quantity volume of water, whose radium content was known, through the filter. Two different kinds of water with different physico-chemical

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