



Determination of oil reservoir radiotracer ($S^{14}CN^-$) in a single step using a plastic scintillator extractive resin

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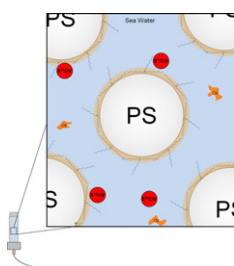
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HIGHLIGHTS

- ▶ A new procedure for $S^{14}CN^-$ radiotracer determination using PS resin was established.
- ▶ The minimum detectable activity for a 100 mL sample is 0.08 Bq L^{-1} .
- ▶ The minimum quantifiable activity for a 100 mL sample is 0.31 Bq L^{-1} .
- ▶ PS resin is capable to quantify $S^{14}CN^-$ radiotracer samples with errors lower than 5%.
- ▶ PS resin is also capable to quantify complex matrices obtained from oil reservoirs.

GRAPHICAL ABSTRACT



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ABSTRACT

The analysis of radiotracers is important in the study of oil reservoir dynamics. One of the most widely used radiotracer is $S^{14}CN^-$. Prior to activity measurements by Liquid Scintillation (LS), routine determinations require the pretreatment steps of purification and concentration of the samples using anion exchange columns. The final elution media produces samples with high salt concentration that may lead to problems with phase separation during the LS measurement. Plastic Scintillation (PS) is an alternative technique that provides a solid surface that can be used as a platform for the immobilisation of selective extractants to obtain a PS resin. The proposed procedure unifies chemical separation and sample measurement preparation in a single step, serving to reduce the number of reagents needed and manpower required for the analysis while also avoiding mixed waste production by LS.

The objective of this study is to develop a PS resin for the determination of ^{14}C -labelled thiocyanate radiotracer in water samples.

For this purpose, the immobilisation procedure was optimised, including optimisation of the proportion of PS microspheres:extractant and the use of a control blank to monitor the PS resin immobilisation process. The breakthrough volume was studied and the detection and quantification limits for 100 mL of sample were determined to be 0.08 Bq L^{-1} and 0.31 Bq L^{-1} , respectively. The established procedure was applied to active samples from oil reservoirs and errors lower than 5% in the sample determinations were obtained.

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1. Introduction

The use of radiotracers is a common option for studying oil reservoirs dynamics [1,2]. When the initial pressure of a reservoir decreases, it is necessary to inject sea water to extract additional oil. Currently, a radiotracer is injected (dissolved in the sea water),

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distributed in the reservoirs and samples are taken from the production well as a function of time. The activities measured provide information about preferential flow directions and communication between wells. The activities also allow for a better understanding of the reservoirs that can help the need for well operations and optimise oil recovery.

Several molecules with beta emitter radionuclides are used as passive radiotracers, including tritiated water (HTO), ^{14}C -labelled thiocyanate (S^{14}CN^-) and tritiated methanol (T-MeOH). A cobalt hexacyanide complex ($^{60}\text{Co}(\text{CN})_6^{3-}$) and sodium salts ($^{22}\text{Na}^+$) are also used, both including gamma emitter isotopes. Among them, thiocyanate is one of the most commonly used. Depending on the radiotracer chosen, each type of sample follows a specific pretreatment before measurement. In the case of thiocyanate, the sample is filtered and concentrated by an anion exchange resin. As a consequence, the eluate contains a high NaClO_4 salt concentration [3].

Activity quantification of the salty solutions is performed by Liquid Scintillation (LS), the reference technique for beta emitter determination [4]. Measurement by LS requires mixture of the aqueous solution with an organic scintillation cocktail [5]. However, when samples with high salt concentration, such as those obtained in the thiocyanate radiotracer pretreatment, are mixed with organic scintillation solutions, problems with phase separation may occur and lead to errors in the detection efficiency calibration [6,7]. Additional characteristics of the LS technique are the production of mixed waste [8] and low selectivity [9].

Plastic Scintillation (PS) is an alternative technique to LS for beta emitter radionuclide determination. The main difference between them is that, in PS, the solvent is polymerised in the form of microspheres. The PS microspheres have the advantage of being reusable and they do not produce mixed waste. PS does, however, show a lower detection efficiency than LS for the low energy beta emitters [10,11]. An additional potential advantage of the PS technique is that it provides a solid surface for the immobilisation of selective extractants. Using this characteristic, it is possible to prepare a PS resin that is selective for a radioisotope that allows direct measurement of the sample (after it passes by the PS resin), avoiding the elution step. This approach saves time, reagents and manpower. It also improves selectivity and concentrates the samples [12].

The objectives of this study were to prepare a PS resin for the determination of ^{14}C in the form of thiocyanate and to establish and apply a procedure for the quantification of radiotracers in water samples from oil reservoirs.

2. Experimental

2.1. Reagents and solutions

All reagents used were of analytical grade. Deionised water was used to prepare the calibration and test solutions. An active stock solution of $133.1 \pm 1.8 \text{ dpm g}^{-1} \text{ S}^{14}\text{CN}^-$ was prepared from an active water solution (PerkinElmer) and standardised using the CIEMAT–NIST method [13]. This stock solution was used to prepare all of the active solutions.

The solid polystyrene (UPS-89) plastic scintillator microspheres were supplied by Detec–Rad. The diameter of the microspheres ranged from 250 to 350 μm . For the LS measurements, Optiphase Supermix cocktails from Wallac PerkinElmer were used. The measurement solutions were contained in 20 mL polyethylene vials (Packard Instruments Co.). The PS resins were contained in column vials specially designed on the basis of standard 6 mL polyethylene vials.

Production water samples from a North Sea oil reservoir were provided by Institute for Energy Technology (IFE) and were used in

the validation step. The activity determination procedure followed at the IFE consisted of passing 1 L of the water sample through an anion exchange resin (Bio–Rad AG1–X8) to concentrate the ^{14}C . The radiotracer that was retained on the resin was later eluted with 10 mL of a 2.8 M NaClO_4 solution. An aliquot of the eluted solution was mixed with the scintillation cocktail and measured [3].

For the production water samples from petroleum extraction, the salinity content was characterised using electrical conductivity and the residual dissolved oil content was quantified using total organic carbon (TOC).

2.2. Apparatus

A P–Selecta ultrasonic bath was used to apply ultrasonic waves to the column vials filled with PS resin.

A Quantulus liquid scintillation spectrometer (EG&G Wallac) with logarithmic amplification, a multichannel analyser (MCA) (4096 channels distributed in four segments of 1024), alpha/beta discrimination and background reduction by active guard were used for scintillation measurements.

Secondary electron images were obtained using Scanning Electron Microscopy (Stereoscan S–360).

Electrical conductivity was performed using a Crison EC–Meter Basic 30+. TOC determination was performed using an Analytic Jena TOC|MULTI N/C 3100.

2.3. Procedure

2.3.1. Immobilisation procedure

The PS resins were prepared by impregnating the PS microspheres with Aliquat–336 (Triethylmethylammonium chloride) that acts as an extractant of thiocyanate. A mass of 20 g of the PS microspheres was suspended in 35 mL of methanol and mixed with the selected quantity of extractant dissolved in 15 mL of methanol. The solution was slurried for 45 min. The methanol was then evaporated under vacuum at 40 °C and 23 mmHg. When there was just enough methanol to cover the microspheres, an additional quantity of methanol was added to re-suspend the resin, followed by slurrying for 5 min. This procedure was repeated three times using different volumes of methanol (15, 7.5 and 3.75 mL), and after the last addition, the solvent was evaporated from the resin over a period of 1 h, leaving a dry product. Finally, the PS resin was filtered in a Büchner funnel with 4 L of water in fractions of 100 mL and dried for 3 h at 40 °C in a drying oven [14]. Three proportions (1:3/4, 1:2/4 and 1:1/4) of PS:extractant were prepared. The reference proportion of 1:1 used in the TEVA columns from Eichrom was 20 g of support particles and 13.75 g of extractant [15].

2.3.2. Active solutions preparation

In all cases, active solutions were prepared by diluting a known amount of active stock solution and KSCN solution to obtain a concentration of 1.127 ppm in deionised water. A blank was prepared with the same composition but without the active stock solution.

For the preliminary approach, PS microspheres:extractant proportions and reproducibility testing experiments active standard solutions of 7.5 dpm mL^{-1} were prepared. In order to measure the breakthrough volume, a solution activity of 1.5 dpm mL^{-1} was used. To measure the detection limit, synthetic samples of 0.7, 0.2 and 0.06 dpm mL^{-1} were prepared. All solutions were prepared and tested in triplicate. The production water samples were also prepared in triplicate by adding KSCN solution to 100 mL of sample to obtain a concentration of 1.127 ppm.

All of the solutions were prepared by weight.

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