New developments in Triskem: Preparation of extraction chromatographic separation materials based on new non-resin supports

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Abstract

Triskem develops extraction chromatographic resins specific for radionuclides and/or group of radionuclides that are used in different fields such as environmental monitoring, decommissioning, nuclear medicine, and more. Depending on the application, the resin format is not always the most efficient option. This is why in the last years Triskem has initiated the development of new formats such as selective discs/filtering membranes and impregnated Thin Layer Chromatography supports. TK100 Discs are used in DGT devices for the accurate quantification of trace-level ($\mu g.L^{-1}$) Sr and Pb concentrations and isotope ratios [$\delta_{SRM\ 987}(^{87}Sr/^{86}Sr)$ and $\delta_{SRM\ 981}(^{207}Pb/^{206}Pb)$] in labile, bioavailable element fractions in soils (1,2). TK201 Discs have been used for the preconcentration of Tc-99 from hospital waster waters before purification and quantification by ICP-MS (3). In the same format, TK-GA Discs have been developed to trap actinides from water samples during filtration and thus allowing subsequent alpha spectrometry analysis (4).

TLC supports functionalized via impregnation with selective extractants is another field of development to provide mostly nuclear medicine with fast quality control regarding labelling yields and radionuclidic impurity determination. DGA Sheets have been developed by Kozempel et al. at CTUP for the determination of radionuclidic impurities e.g. in Ra-223, Ac-225 or Pb-212 solution for use in labelling. They further have been used to quantify Ra-Macropa labelling yields. CU Sheets have been developed to facilitate the analysis of Cu radiolabelled peptides.

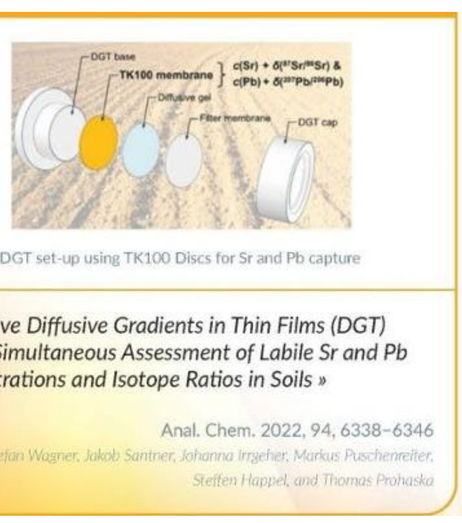
Further, the development of other selective Discs/filtering membranes and iTLC supports currently being developed are presented here.

Key words: impregnated discs, iTLC, TK100 discs, TK-GA discs, DGA Sheets, CU iSheets.

Extractive/Impregnated discs: Transposition of different resins extractive system onto discs to simplify preconcentration/purification/detection-counting

- Filtration or passive sampling on 25mm or 47mm diameter discs
- Already in-use/published: TK100, TK201 and TK-GA

TK100 Discs for passive uptake of Sr and Pb in soils [1][2]

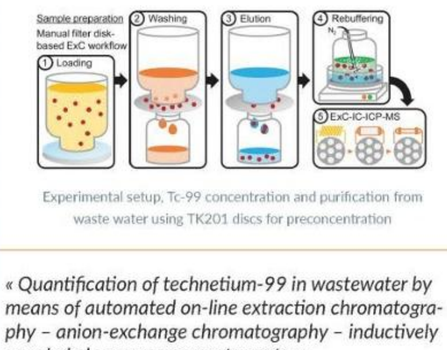


DGT set-up using TK100 Discs for Sr and Pb capture

« Selective Diffusive Gradients in Thin Films (DGT) for the Simultaneous Assessment of Labile Sr and Pb Concentrations and Isotope Ratios in Soils »

Anal. Chem. 2022, 94, 6338−6346
Stefan Wagner, Jakob Santner, Johanna Irrgeher, Markus Puschenreiter, Steffen Happel, and Thomas Prohaska

TK201 discs for Tc-99 preconcentration /determination in waste water [3]



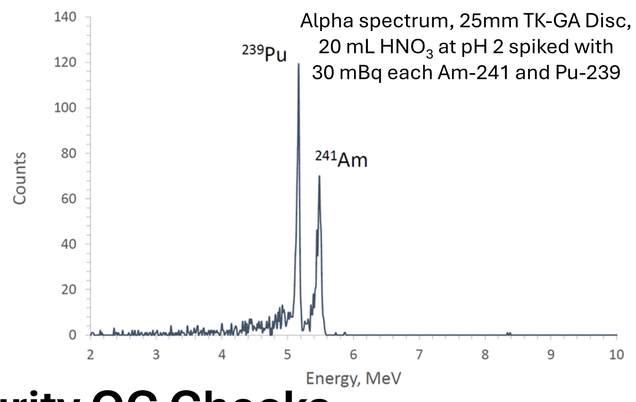
Experimental setup, Tc-99 concentration and purification from waste water using TK201 discs for preconcentration

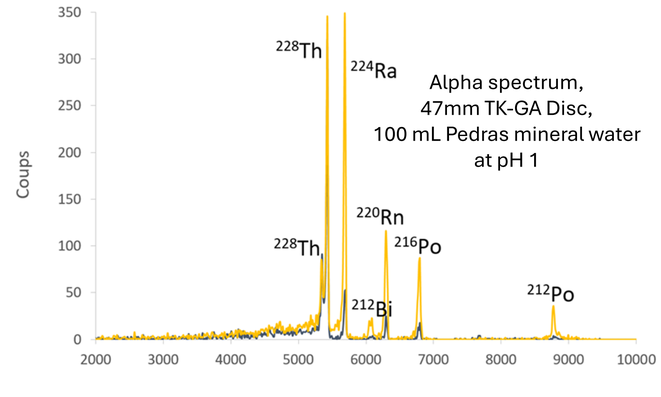
« Quantification of technetium-99 in wastewater by means of automated on-line extraction chromatography – anion-exchange chromatography – inductively coupled plasma-mass spectrometry »

J. Anal. At. Spectrom., 2024, 39, 2774-2782
Maximilian Horstmann, C. Derrick Quarles Jr, Steffen Happel, Michael Sperling, Andreas Faust, David Clases and Uwe Karst*

Typical applications of TK-GA Discs [4]:

- pH 1 – 2 (for some actinides up to 3M HNO_3)
- Disc preconditioning : 20% EtOH:water, water, then loading medium
- Sample Loading, flow rate: 1 - 10mL/min
  - Lower flow rates favors better resolution
  - 25 mm Discs => up to 100mL samples
  - 47 mm Discs => up to 1L (higher volumes possible depending on extractant system)
- Rinsing with water and 20% EtOH:water
- Drying and glueing onto steel disc => alpha spectrometry
- Optional: subsequent Alpha/Beta by LSC

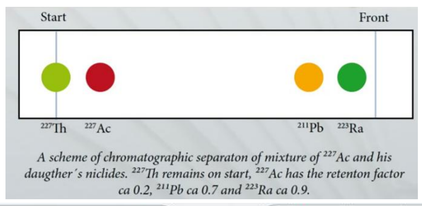




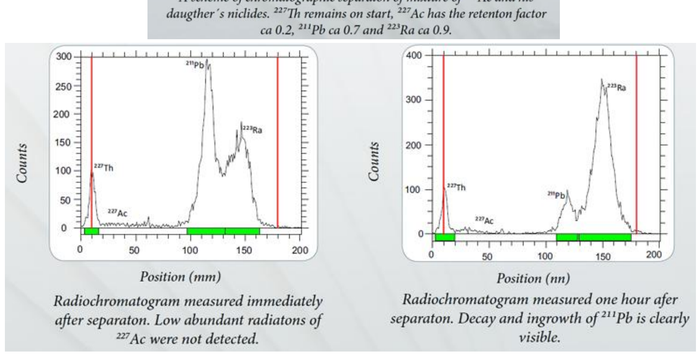
Impregnated TLC supports: for Fast Radionuclidic purity QC Checks

DGA Sheets by Kozempel et al. (CTUP) [5][6]

QC of radionuclides and generator eluents
(e.g.: Ra-223, Ac-225/Bi-213, Pb-212,...)



A scheme of chromatographic separaton of mixture of ^{227}Ac and his daugther´s niclides. ^{227}Th remains on start, ^{227}Ac has the retenton factor ca 0.2, ^{211}Pb ca 0.7 and ^{223}Ra ca 0.9.



Radiochromatogram measured immediately after separaton. Low abundant radiatons of ^{227}Ac were not detected.

Radiochromatogram measured one hour afer separaton. Decay and ingrowth of ^{211}Pb is clearly visible.

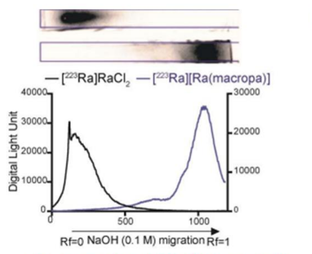


Fig. 1 Formation of [^{223}Ra][Ra(macropa)] under migration of [^{223}Ra]$RaCl_2$ (top) and [^{223}Ra][Ra(macropa)] (bottom) on DGA-coated chromatographic strips. [5]

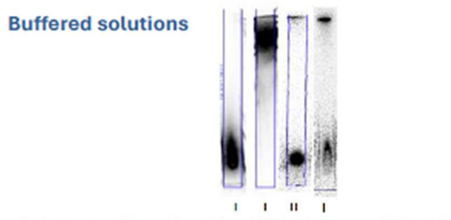


Fig. 2 TLC autoradiographic readings of [^{223}Ra][Ra(macropa)] (10 mM), DOTA (10 mM) and EDTA (5mM) migrated on DGA-coated TLC using a NaOH (0.1 M) mobile phase.
I. [^{223}Ra]$RaCl_2$ migration (Rf≈0)
II. [^{223}Ra][Ra(macropa)] (Rf ≈ 1)
III. ^{223}Ra tentatively chelated with DOTA showing unsuccessful radiolabeling (85% remained unchelated)
IV. ^{223}Ra partially chelated with EDTA (65% remained unchelated). [5]

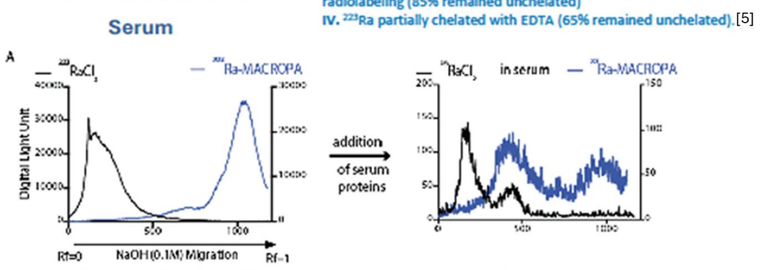


Fig. 3 TLC profiles of pure [$^{223/224}$Ra]$RaCl_2$ and labeled macropa in PBS (left) and in human serum (right). [^{223}Ra]$RaCl_2$ (black trace) and [^{223}Ra][Ra(macropa)] (blue trace) mixed in serum were poorly separated, questioning the suitability of the DGA sheets to detect Ra decomplexation in serum. Weak ionic interactions of [^{223}Ra]Ra^{2+} or [^{223}Ra][Ra(macropa)] with proteins may occur and result in the inability of TLC to clearly separate each species. [5]

CU Sheets by Svedjehed et al. [7]:

QC of Cu radiolabeled peptides (labeled vs free Cu): Both iTLC paper (SG and SG-CU) developed in less than 10 min. while Whatman paper took 25 – 30 min.

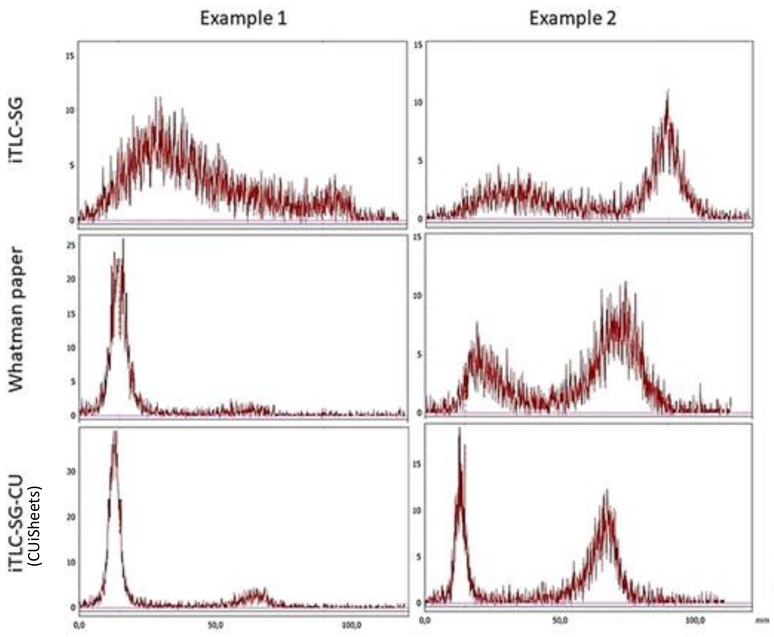


Figure 1. TLC scans of [^{61}Cu]Cu-NOTA-octreotide spotted on: top, iTLC-SG; middle, Whatman paper; bottom extractant-impregnated iTLC-SG. Example 1 notes elevated levels of unlabelled ^{61}Cu, while example 2 notes comparable levels of labelled to unlabelled ^{61}Cu.

On-going studies

- **Extractive membranes :** TK221 and TK200 for Actinides rapid screening // new application using TK200 for Po in Ac-225 // TK101 for Ra and TK102 for Sr // TK227 for Ra, indirect measurement of Sr-90 via Y-90 // TK201 for Tc

Methods developments and optimisation

- **iTLC sheets :** Zr, TK201, TK213,...

2D TLC for radionuclide screening

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