Development of a novel rapid screening technique for radionuclides using test-stick technology

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2023 Triskem User Group Meeting, 22nd February

Contents

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Introduction – the challenge

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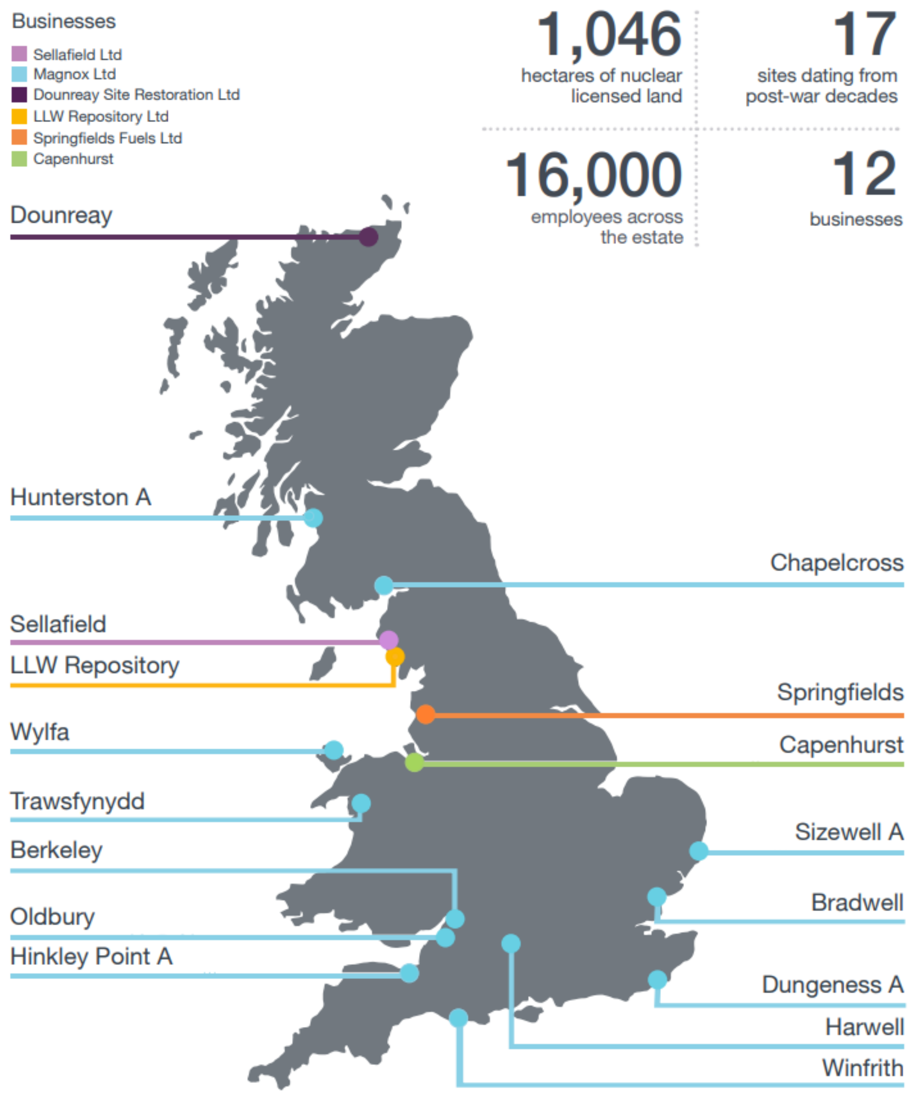
- UK undergoing large scale decommissioning process led by the Nuclear Decommissioning Authority (NDA)
- Site Licensed Companies (SLCs) have been contracted to tackle decommissioning in 17 different sites
- Legacy waste and site complexity challenges
- Radionuclides such as ^{90}Sr, ^{99}Tc and ^{137}Cs are some of the most commonly found contaminants



NDA Business Plan 2018 to 2021 11

Introduction – NDA strategy

- NDA has identified and promoted the development of rapid screening techniques to streamline the decommissioning process at VLLW (Very Low Level Waste) and below.

Real time measurement with ease of data manipulation is preferred[1]

Promoting timely characterisation and segregation of waste[2]

Portable versions of existing characterisation techniques and non-destructive evaluation technologies[2]

[1]Technical Memorandum, National Nuclear Laboratory, 2017
[2]NDA Technical Baseline, Nuclear Decommissioning Authority, 2016

Test-stick technology – a possible solution?

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- Test-stick technology looks to offer a method of rapidly screening a selected major contaminant radionuclide

- Test-sticks could be used to potentially determine whether:
  a) A specific radionuclide is present
  b) The specific radionuclide is present in activities that are above or below legislative thresholds (EPR 2016, for example) – support waste sentencing

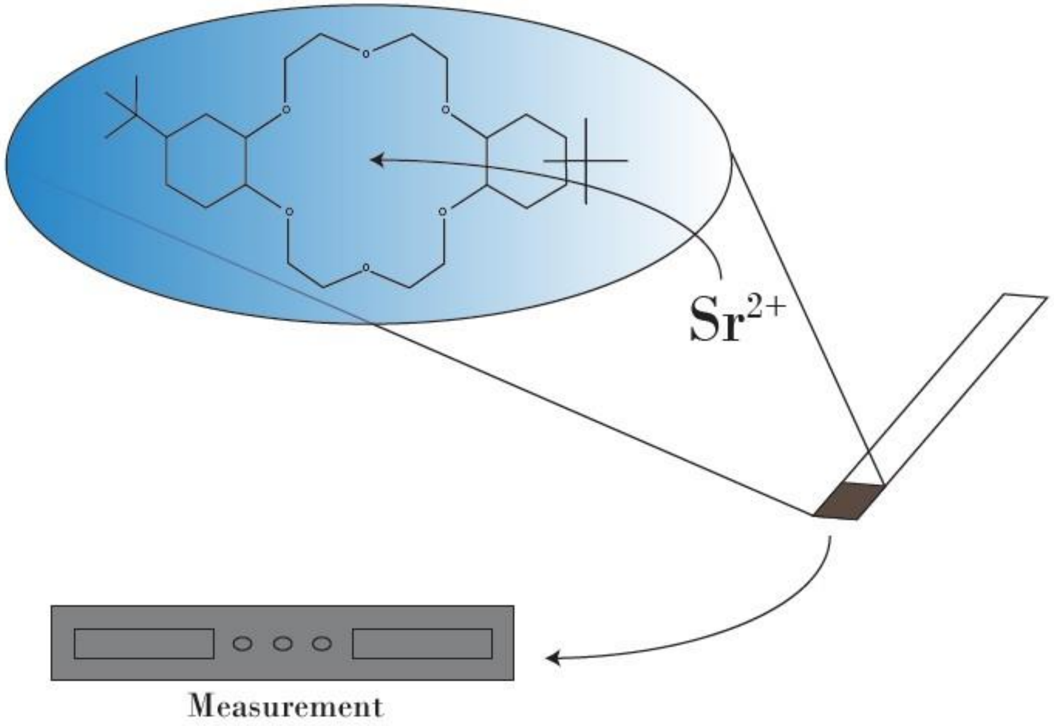
- Test-stick technology would be a readily deployable on-site technique and act as a first-response screening technique

Test-stick technology - overview

- A rapid screening technique to be utilised on-site to rapidly determine the presence of specific radionuclides of nuclear decommissioning importance.

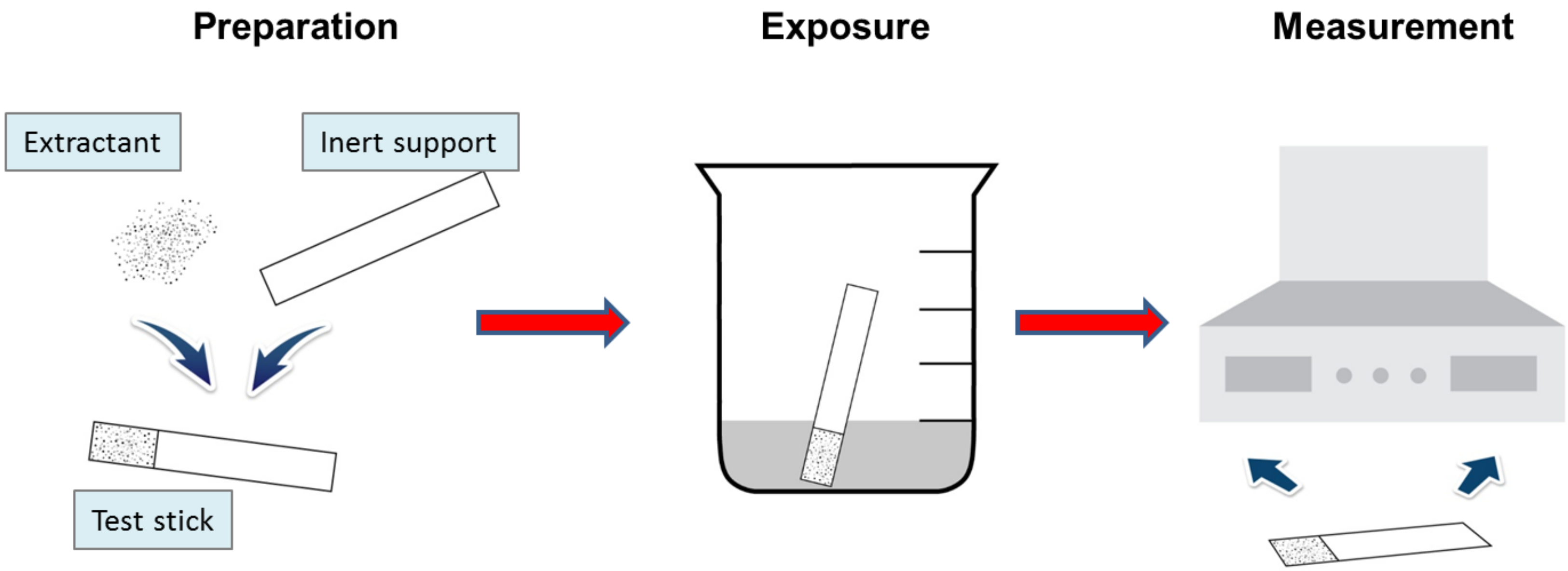
- The chosen radionuclide to conduct this study was ^{90}Sr due to its:
  - High fission yield (~5.7%)
  - Pure β-emitter
  - $t_{1/2}$ = ~29 years
  - Bone seeker properties

- TK100 is an ideal extractant to utilise in this study due to its Sr selectivity and operability in neutral pH conditions (compared to other Sr extractants such as Sr-resins).



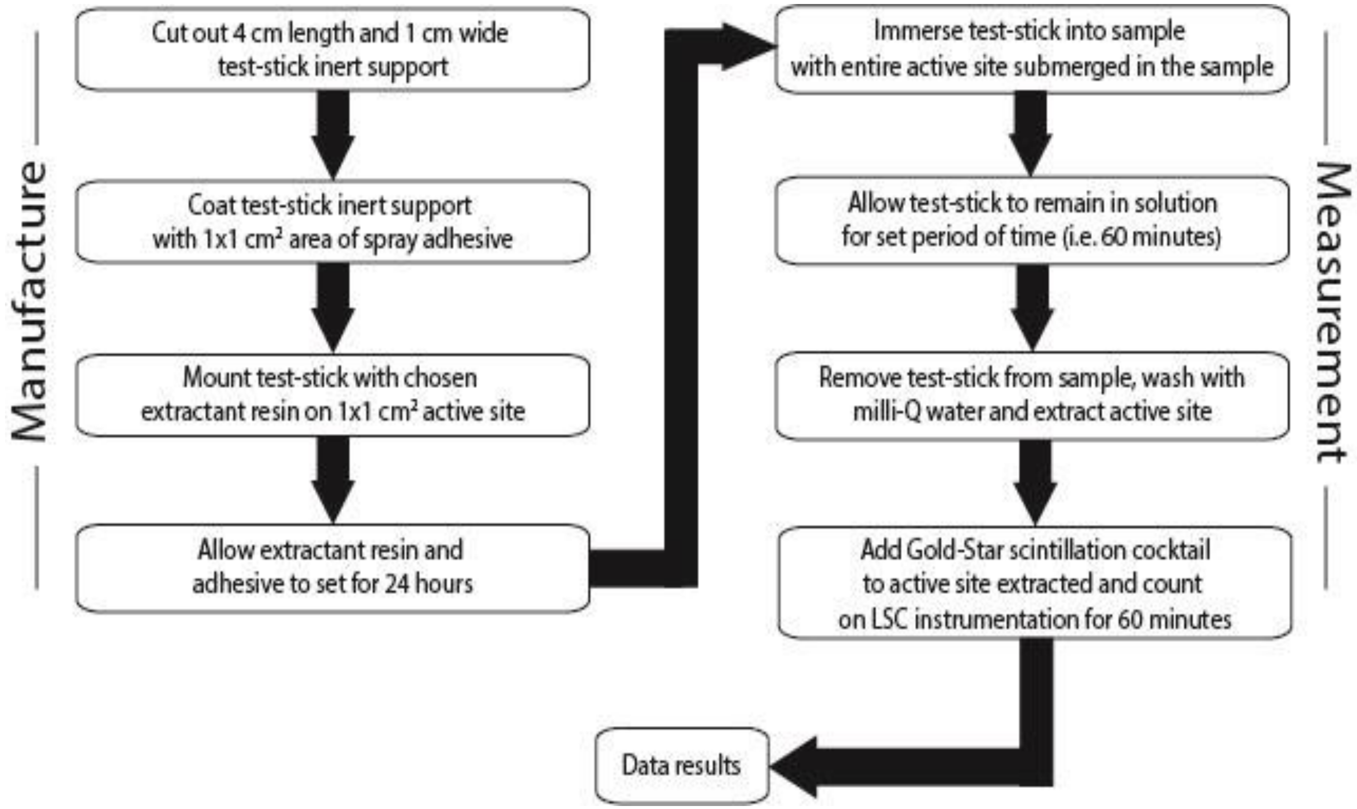
Test-stick technology: how-to guide

- Test-stick technology can be organised into 3 phases – preparation, sample exposure and measurement.



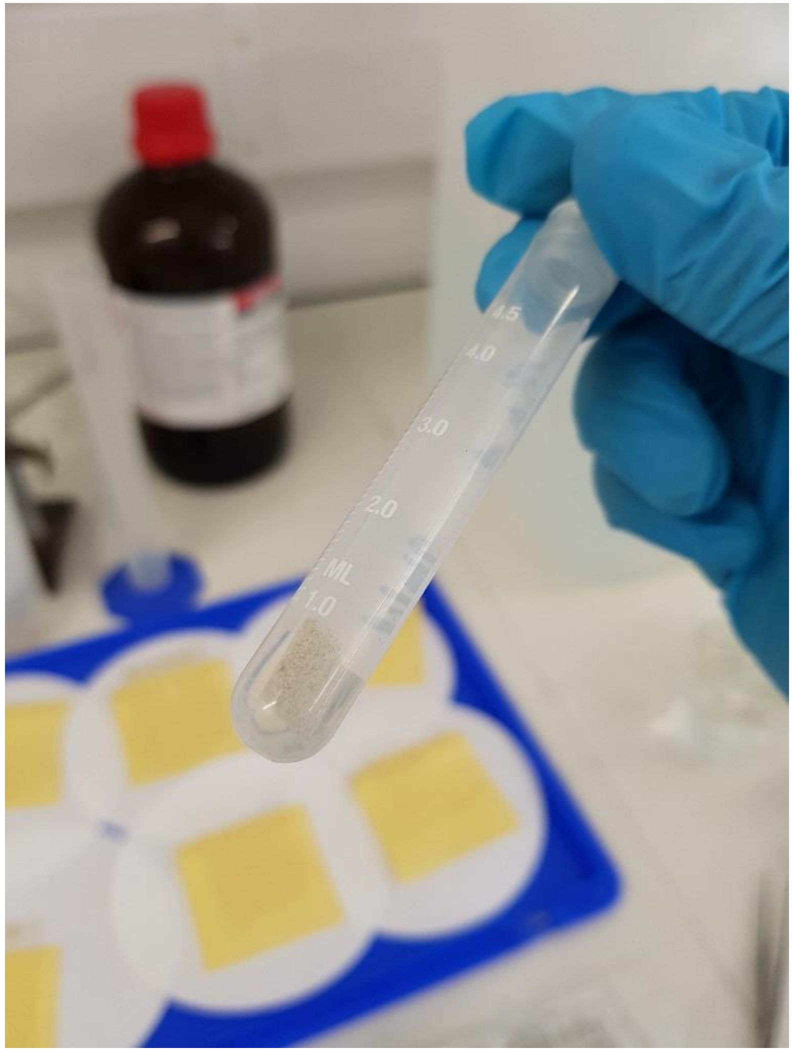
- The active site consisting of 1 cm^2 area at the tip of the test-stick is the focal point as this is where ^{90}Sr uptake will occur.

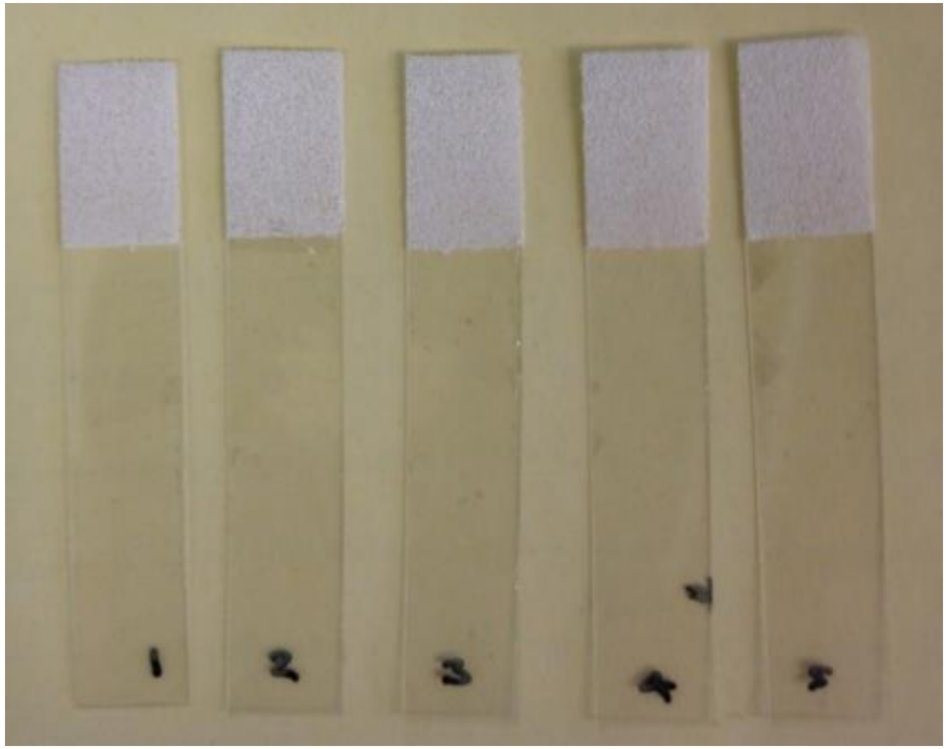
Test-stick technology - methodology



- Flow chart demonstrating the methodology of test-stick technology from manufacture to data acquisition.

Test-stick technology





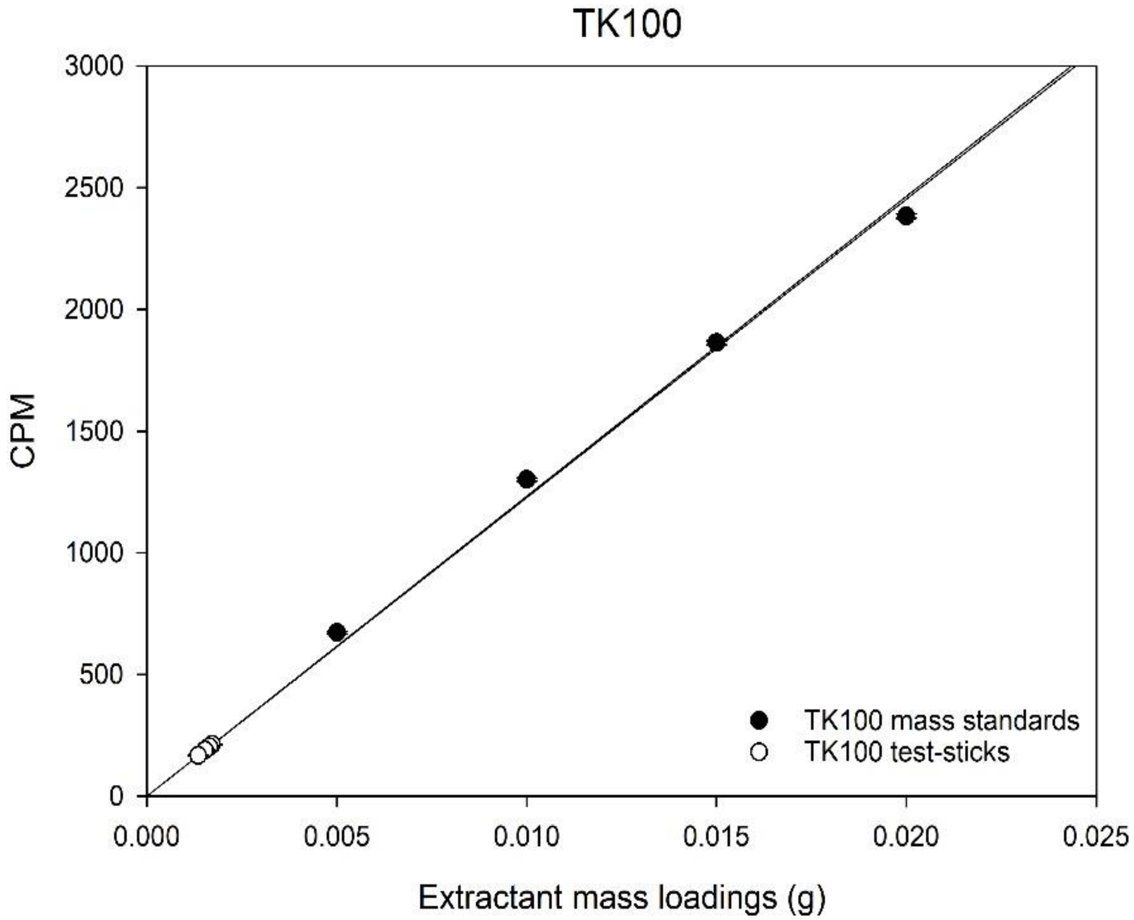
Test-stick technology: target parameters

- The targeted parameters that were tested for and presented here include:
  - Determining mass loadings of the extractant (TK100) on test-sticks
  - Test-stick response over ^{90}Sr activity range
  - Uptake kinetics that meet the needs for rapid screening scenarios
  - Impact of strontium concentration on test-stick performance
  - Ionic interferences
  - Theoretical LODs

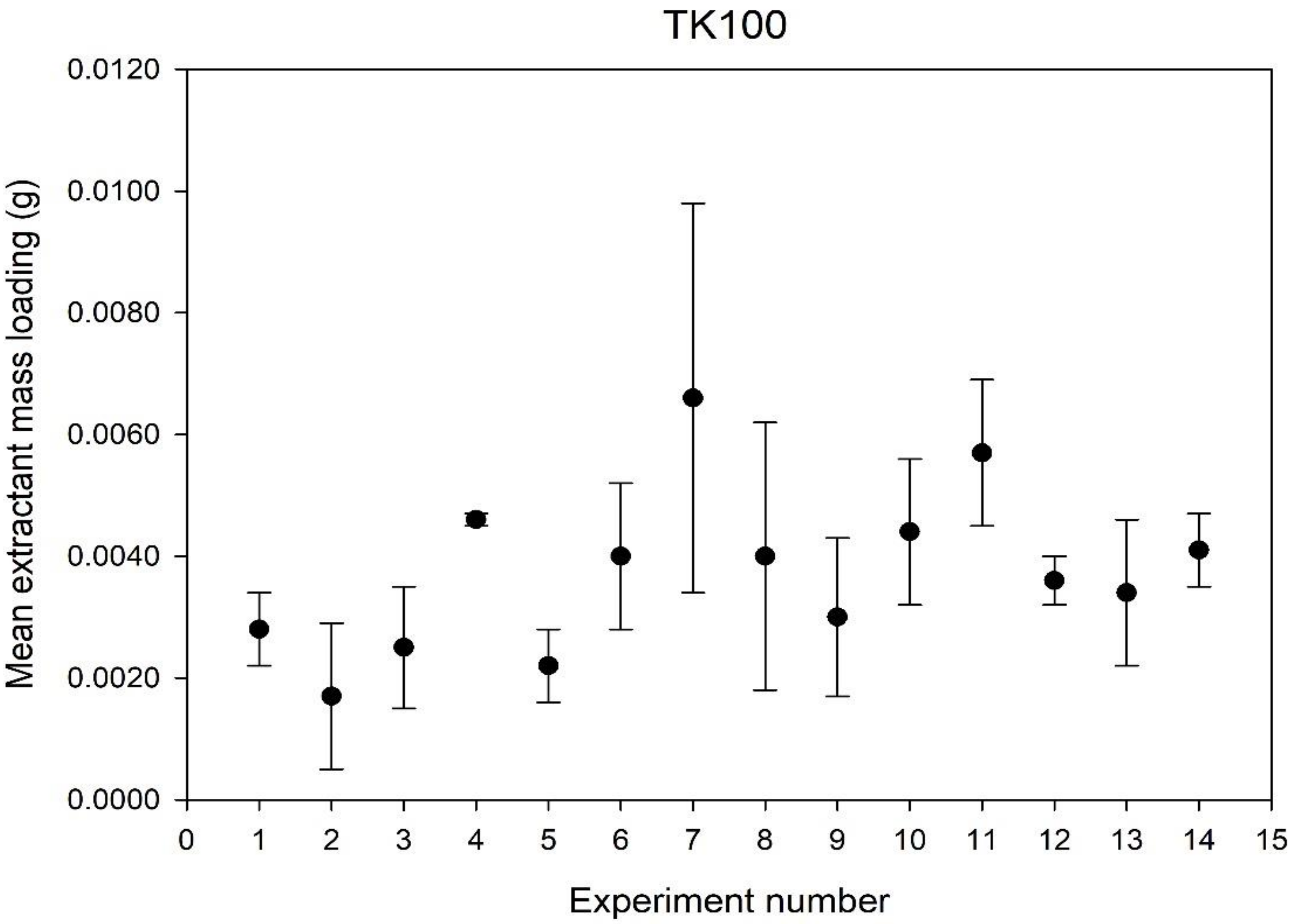
- Target matrix is groundwater as it presents a good medium to carry out test-stick sampling due to the presence and mobility of ^{90}Sr in groundwater. It also presents a good foundation for future test-stick applications.

Results – mass loadings

- Mass resin loading experiments using radiolabelled TK100 extractant showed that over four test-sticks the average TK100 test-stick mass loading was 1.6 ± 0.4mg (2σ, n = 4). This suggests that very low extractant mass is required for test-stick manufacture.

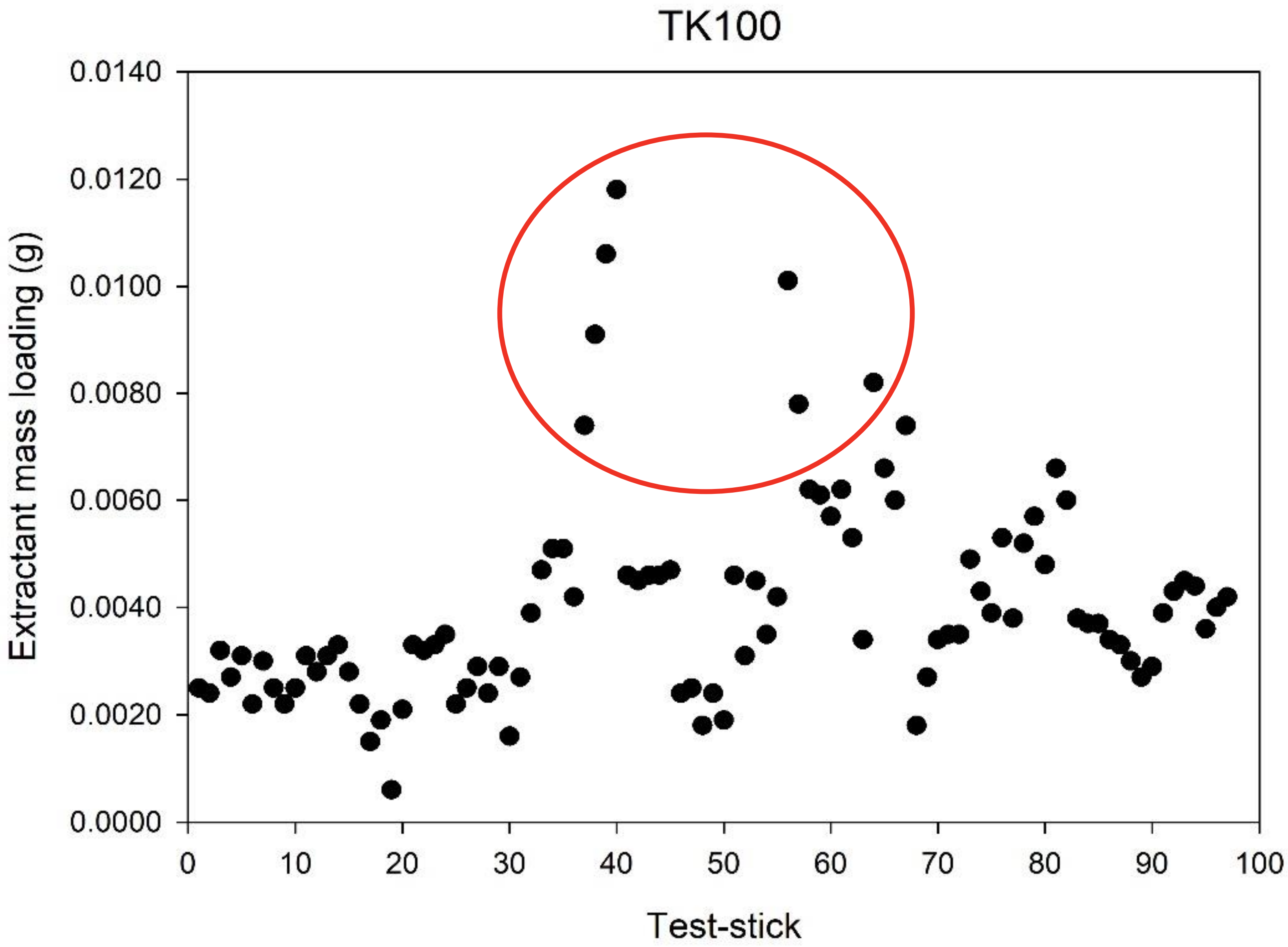


Results – mass loadings



- Alternative method involving individually weighing test-sticks both prior to (inert support only) and post-manufacture (inert support + adhesive + extractant). Gravimetric analysis revealed larger variance in extractant loadings: 3.8 ± 1.1 mg (2σ, n = 14)

Results – mass loadings



N = 96

Results – proportionality mechanism

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- Effectiveness of test-stick technology is dependent on the proportionality of uptake.

$$A_{stick} \propto A_{aqueous}$$

- This means that with increasing activity of ^{90}Sr in the aqueous phase (groundwater), the uptake of ^{90}Sr onto the test-stick will also increase…

…proportionally, hopefully.

$$A_{stick} = b_t \cdot [A_{aq}]_0$$

Results – proportionality mechanism

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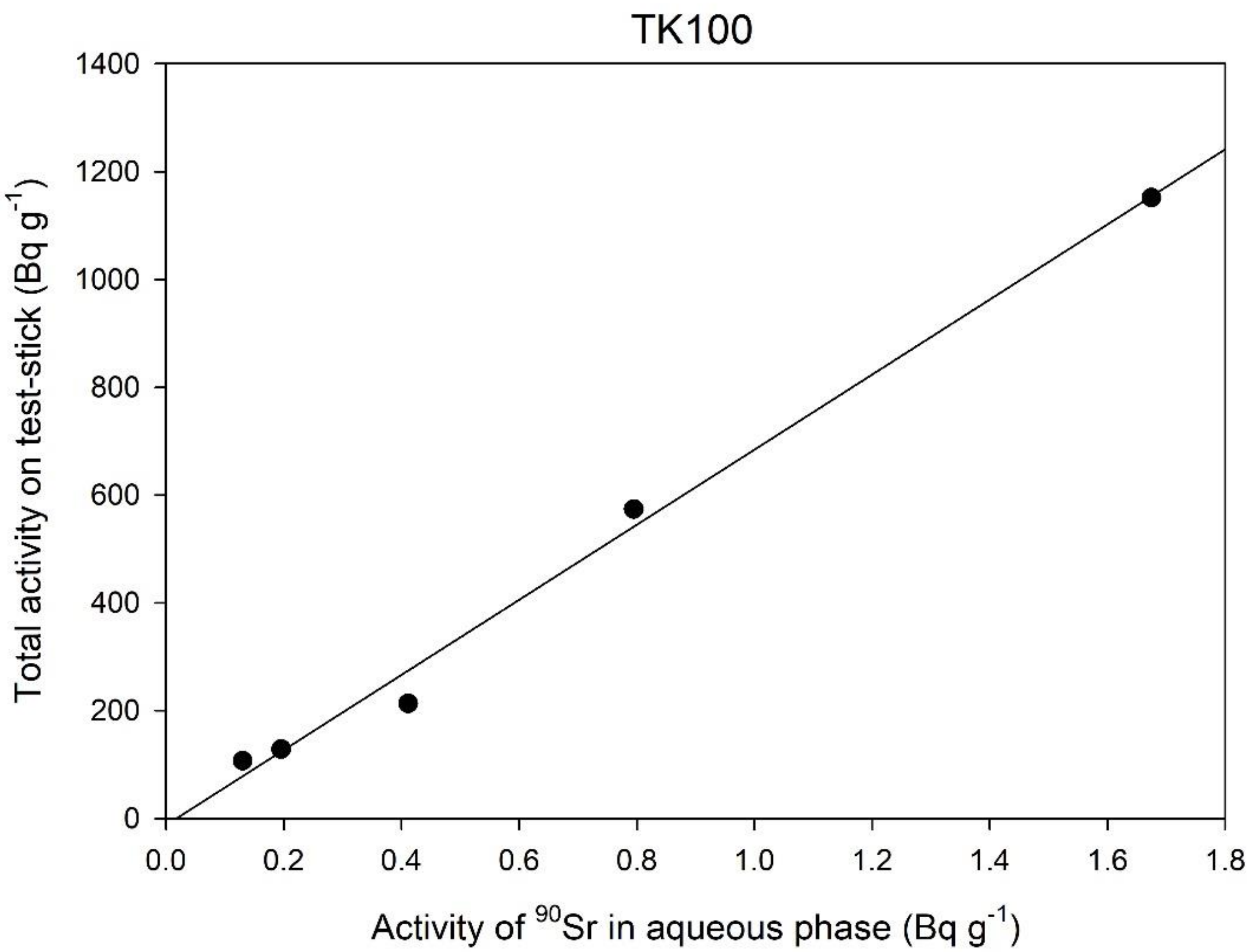
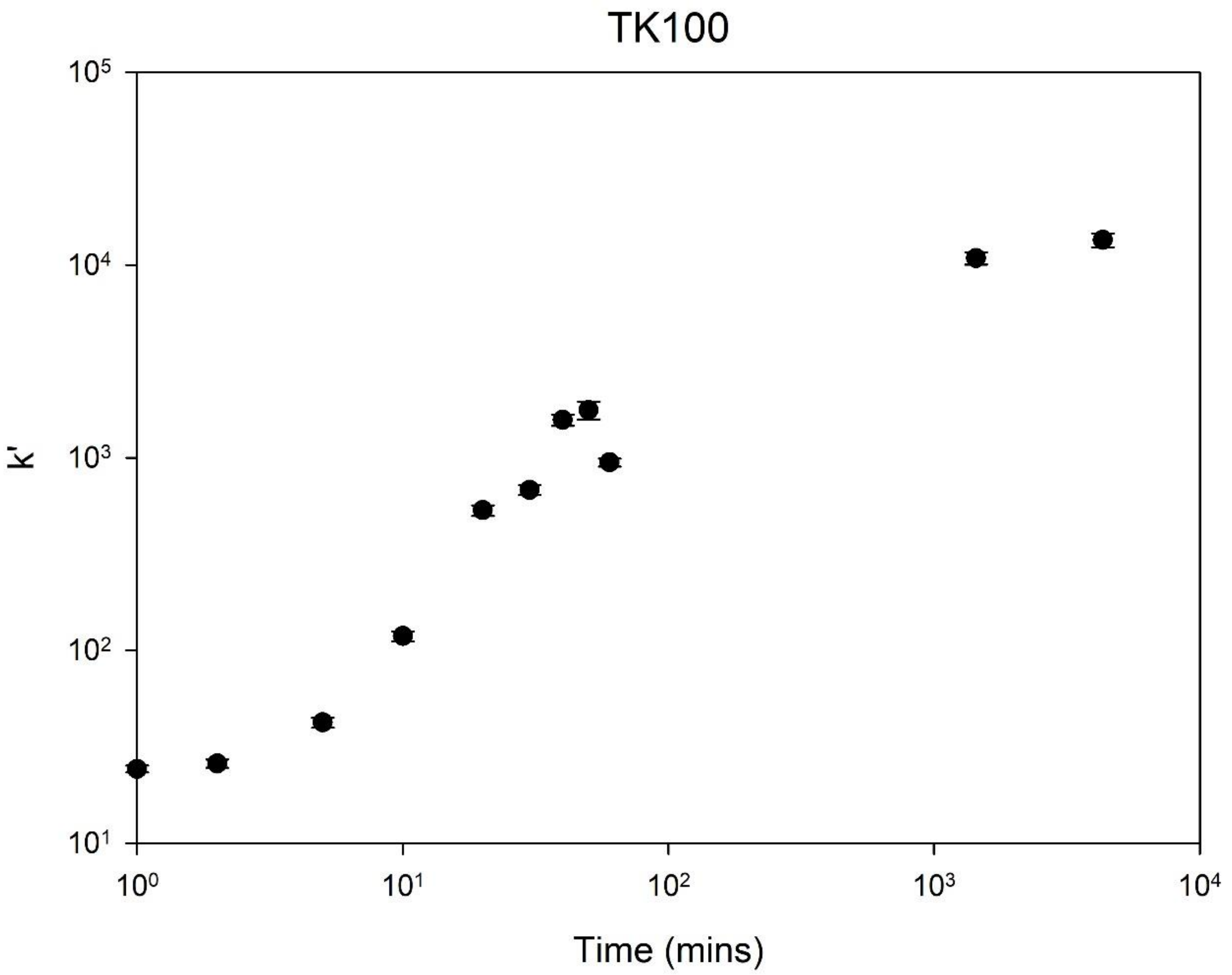


- Proportionality mechanism is found to function well over ^{90}Sr activity spike ranges ranges from 0.1 to ~9.0 Bq g^{-1}.
- Agitation was utilised to improve linearity of activity response (R^2 = 0.998) and subsequently the proportionality mechanism exhibited by the test-sticks.
- Activity range demonstrates TK100 test-sticks' potential application in waste segregation to distinguish whether ^{90}Sr is above or below VLLW limits (1.0 Bq ^{90}Sr, EPR 2016)

Results – uptake kinetics

- Kinetic data showed k' values increasing substantially from 1-4 mins all the way to 50 mins.
- Test-stick kinetics did not differentiate much from 50 mins (10^3) to 24-48 hours (~10^4) for on-site deployment.
- Ideal deployment time is shown to be between 40 to 60 minutes for TK100™ test-sticks.



Results – strontium loading capacity

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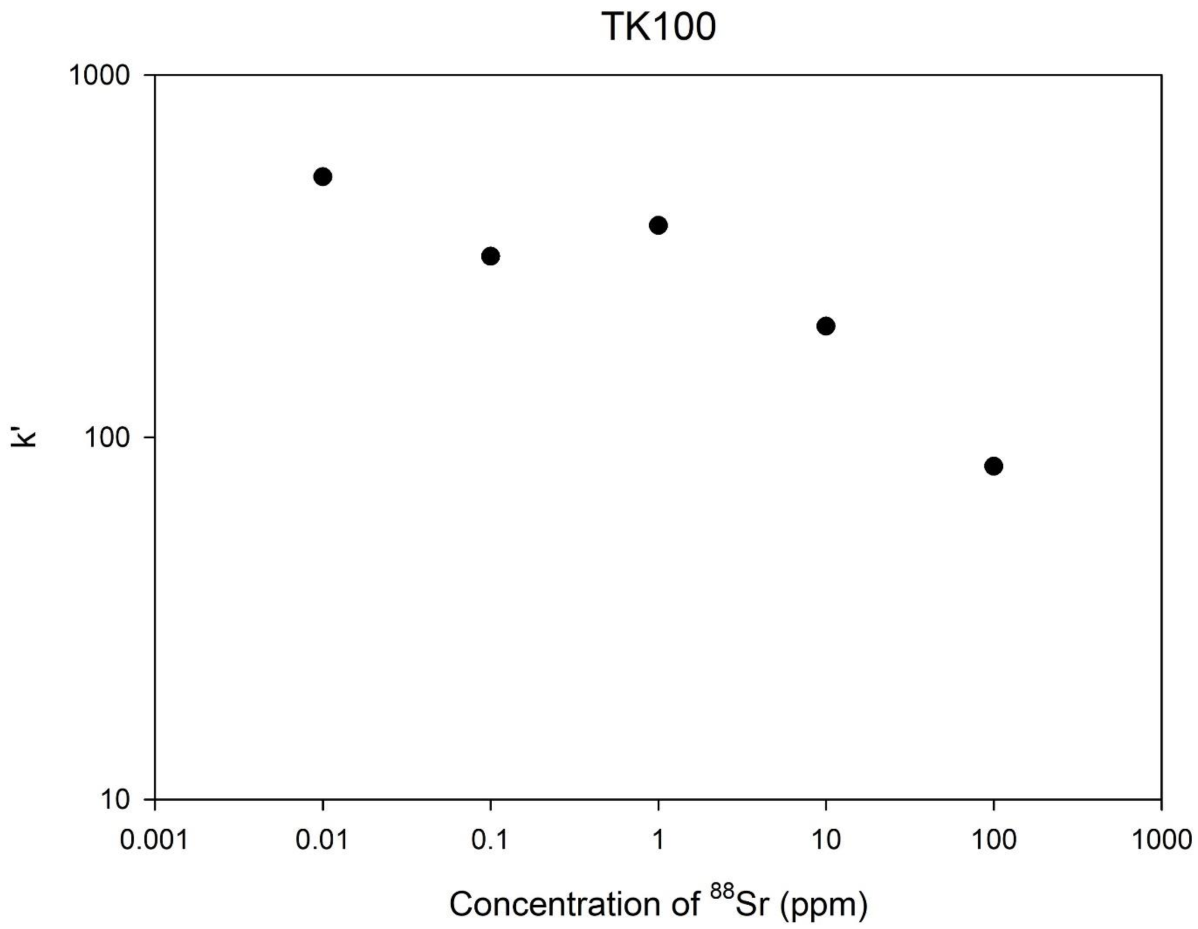
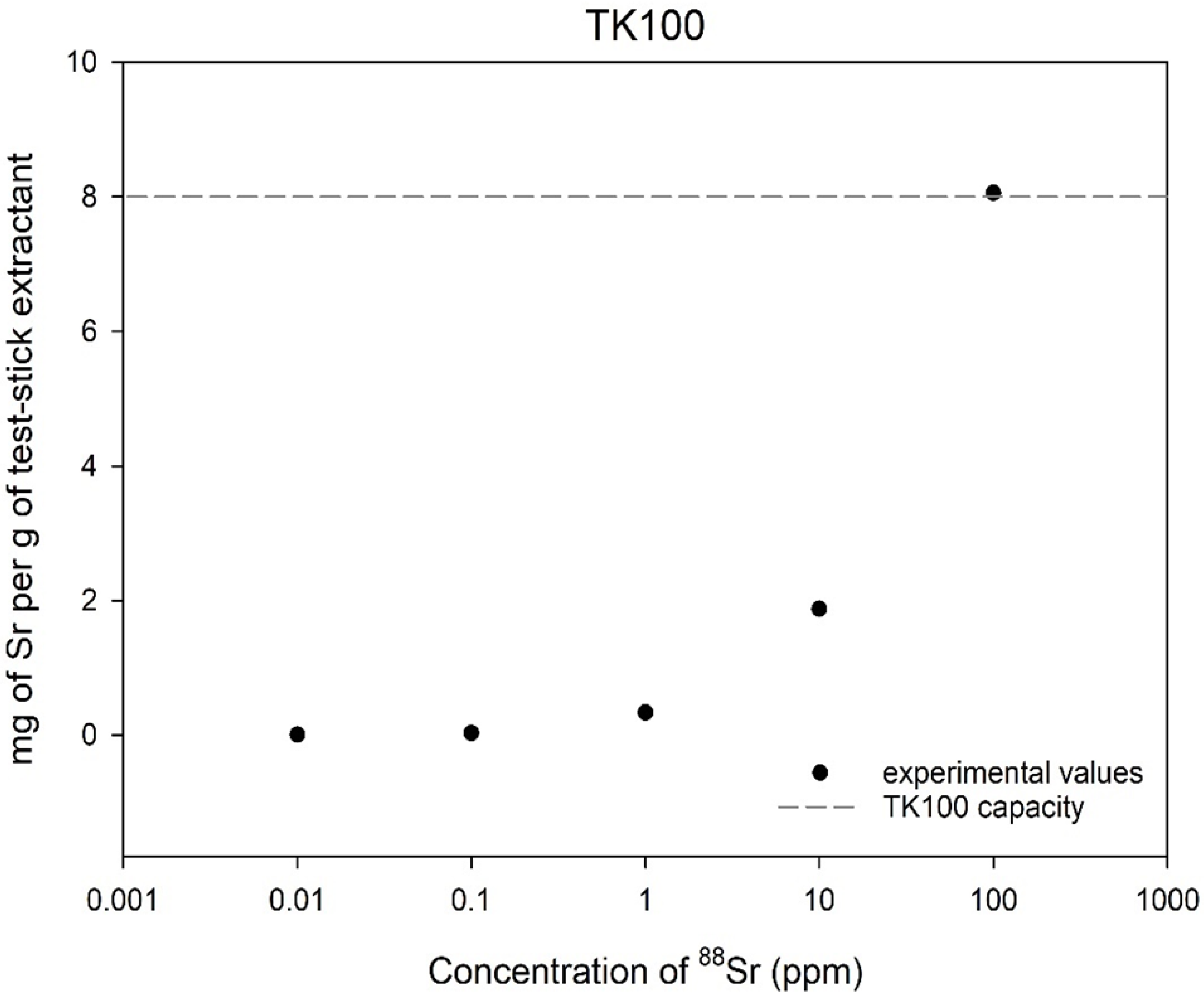


- TK100 test-sticks were shown to operate similarly to the product sheet specification for bulk-phase TK100.
- This is despite the fact that uptake geometry has shifted from 3D (bulk-phase) to 2D (test-stick) uptake.
- TK100 test-sticks were shown to operate at Sr concentrations of up to 100 ppm before reaching capacity.

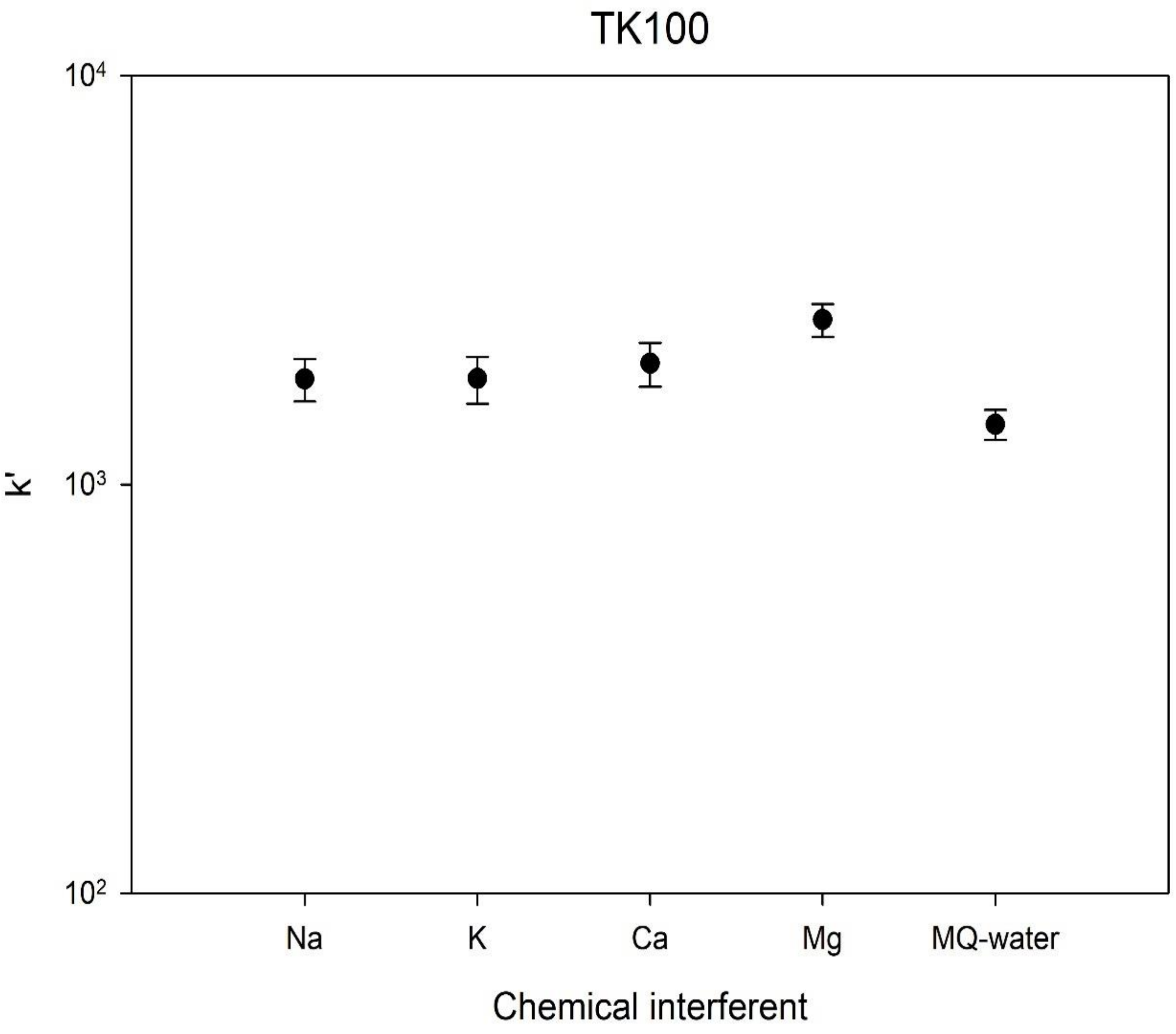
Results – strontium loading capacity



Results – ionic interferences

- Partition coefficients, k’, values for TK100 test-sticks revealed minimal differences between the ionic species.

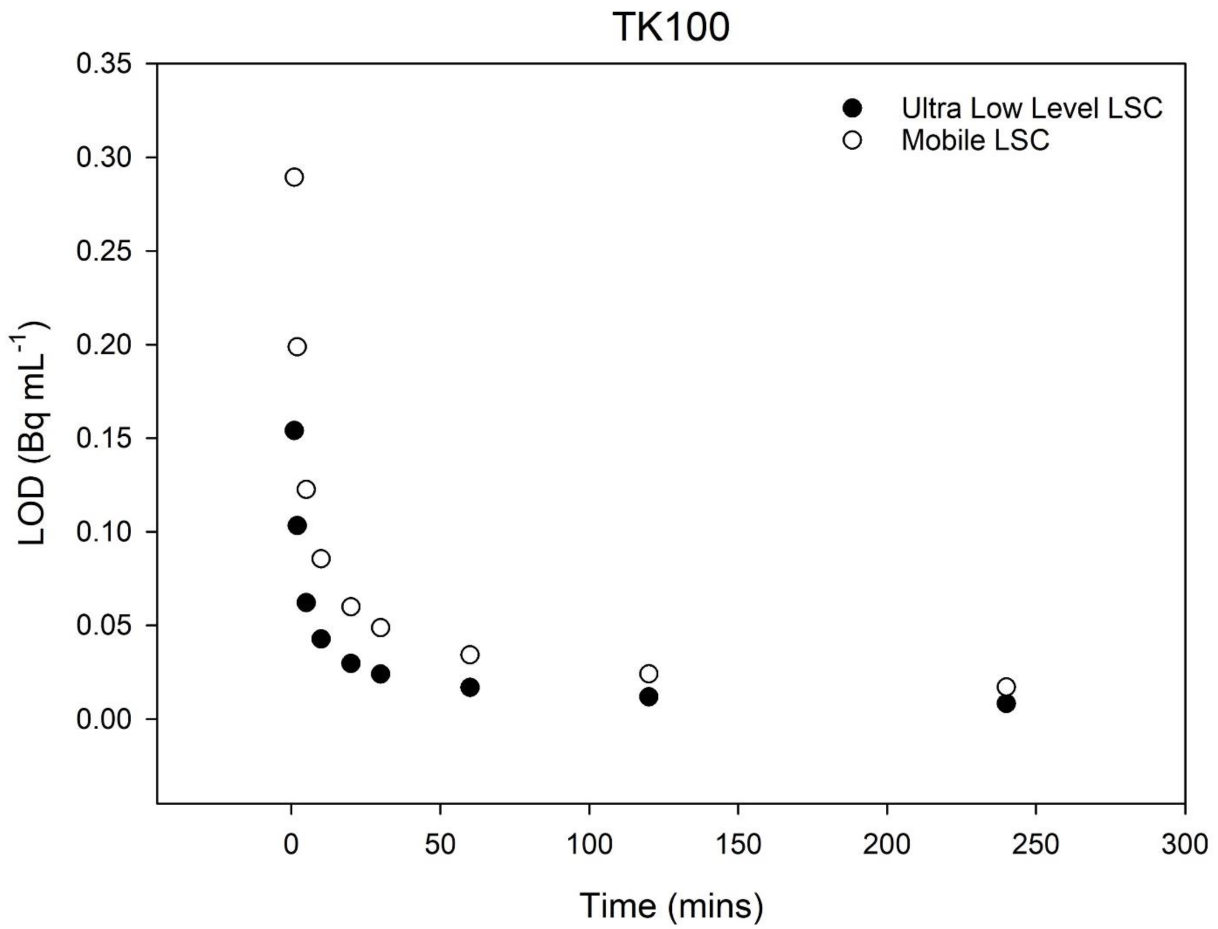
- Results revealed milli-Q water with the lowest obtained k’ value of 1402 whilst Na, K, Ca and Mg had k’ values of >1800 suggesting these species to exhibit minimal effect on test-stick uptake efficiency



Spiked activity of ^{90}Sr was ~10.1 Bq. Concentration of species were approximately: 94 ppm Na, 11 ppm K, 250 ppm Ca and 50 ppm Mg in Milli-Q water corresponding to NNL literature Sellafield groundwater composition

Results – limit of detection and count times

- Using Currie equation to calculate theoretical LODs for ‘measurement’ aspect of test-stick technology.
- $L_D = 2.71 + 4.65\sqrt{\mu_B}$
- LODs were found to reach target 0.1 Bq mL^{-1} within 5 minutes.
- This suggests that count times can be potentially reduced to 5-10 minutes and still achieve measurable counts.



Future work

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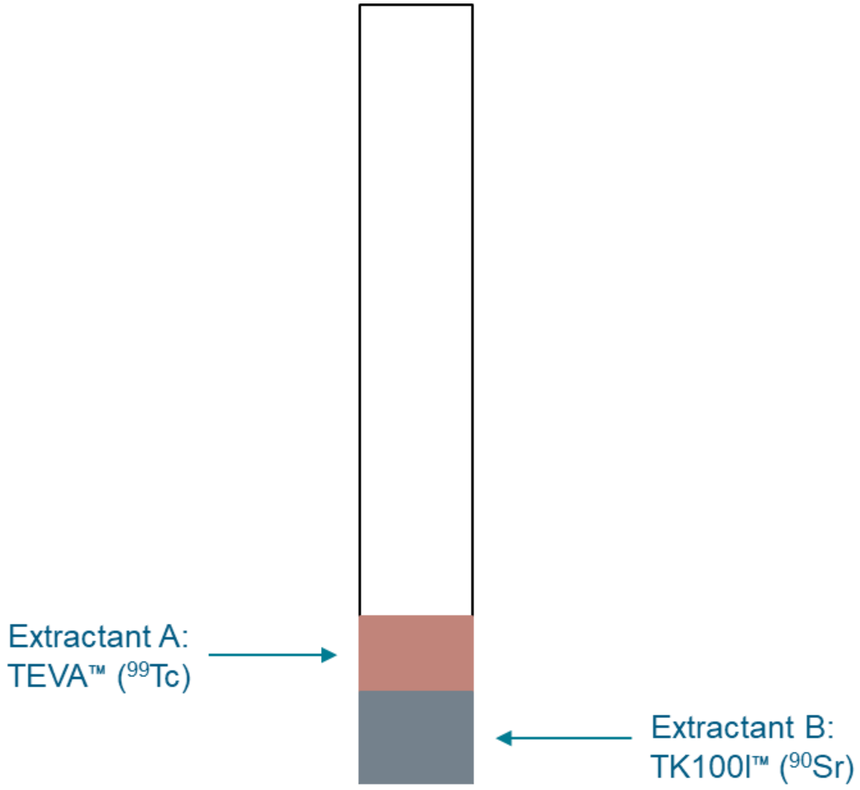
- Prospects for future work include improvements in test-stick manufacture including introducing automation to minimise test-stick extractant mass loading variations.
- Further streamlining of test-stick technology includes utilising potential scintillant embedded extractants which would remove the need to add scintillant cocktail prior to analysis.
- Exploring alternative extractants to selectively uptake other radionuclides of decommissioning interest.

Future work



"Kill two birds with one test-stick"

- Eventually look at a different radionuclide of nuclear decommissioning importance.
- Using the work carried out on both ^{90}Sr and other radionuclides to develop an MES test-stick.
- MES (Multi Extractant System) test-stick would aim to rapidly screen more than one radionuclide simultaneously.

Conclusion

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- Test-stick technology addresses the need for rapid screening techniques for efficient waste characterisation whilst also promoting simplicity and ease of use.
- Low mass loadings required for test-sticks to function.
- Initial results indicate near-maximum uptake efficiency in 40 to 60 mins and stirring the test-stick yields more efficient and proportional uptake over a range of activity.
- Demonstrated the potential to lower count times to 5 to 10 minutes which would result in total analysis being carried out within the hour.
- There is a large number of possibilities in future work regarding test-stick technology including MES and scintillant-embedded extractants.

Acknowledgements

- NDA PhD bursary scheme 2017 for sponsoring this project work as well as the support provided by NNL
- PhD supervisors: Prof Phillip Warwick and Prof Ian Croudace
- Industrial supervisor: Steve Walters
- GAU-Radioanalytical

Thank you for listening

FUNDED BY BEIS

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