

ABSTRACT

- The IAEA-SGAS¹, in connection with the NWAL² measures individual sub- and micrometer U-bearing particles on swipe samples taken by IAEA safeguards inspectors during in-field verification activities.
- The elemental and isotopic composition of these particles are used to verify the absence of undeclared activities.
- To ensure the quality control of the analytical results from particle analysis, microparticles with well-defined properties as reference materials are required.
- In order to find applicability as a reference material, a certain stability (shelf-life) of the particles must be guaranteed.
- The stability of the particles in various atmospheres and in various solvents has been launched to optimize storage conditions.

BACKGROUND / INTRODUCTION

IAEA REQUIREMENTS FOR REFERENCE MICROPARTICLES

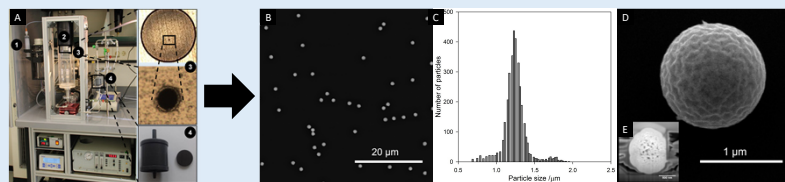
Isolated monodisperse homogenous dense spherical U-bearing microparticles are required with defined:

- interparticular distance on substrate,
- particle size (0.5 μm to 2 μm),
- morphology (amount of uranium per particle),
- elemental and isotopic composition, and
- Shelf-life conditions.



In 1997, the synthesis of U-particles started at Harwell Lab, UK. In 2019/21, the first two available batches of reference microparticles produced at FZJ³, which fulfil IAEA's requirements, were certified.

PARTICLE PRODUCTION AND CHARACTERIZATION AT FZJ



Photograph of VOAG set-up for U-microparticle production (A) and SEM image (B), particle size distribution (C) as well as SEM image (D), and cross section view (E) of an individual microparticle.

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Systematic Shelf-Life Investigation on Uranium Oxide Reference Microparticles to Identify Optimal Storage Conditions

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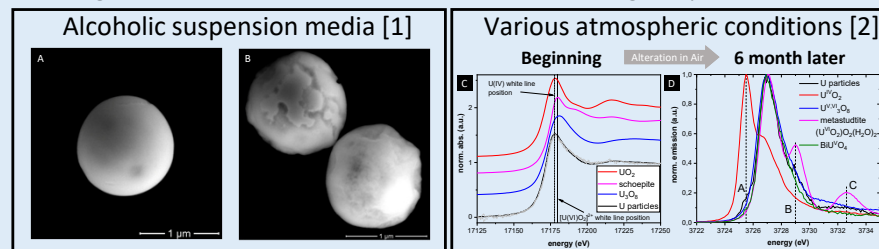
MAIN CONTENT

PRODUCED PARTICLES FULFIL IAEA'S REQUIREMENTS REGARDING interparticular distance on substrate, particle size, elemental and isotopic composition, and morphology.

SHELF-LIFE

Motivation for the systematic shelf-life investigations

- Observable alteration of the microparticles stored in ethanol for four years due to increasing surface roughness
- Change in the oxidation state of uranium after storage of particles in air



SEM image of U-particles after 0 days (A) and four years (B) of storage in ethanol and U-L_{III} edge (C) and U-M_{IV} edge (D) XANES spectra of the particles stored in air.

Investigation plan

Shelf-life	Storage conditions	Plan	Aim
Atmospheric conditions	<ul style="list-style-type: none"> • In air and in air at 90 °C • In argon gas atmosphere • In humid atmosphere 	Investigation over 2 years with various characterization methods	<ul style="list-style-type: none"> • Identify ideal storage conditions • Alternative suspension media to ethanol
Suspension media	Alcohols (Ethanol, 2-Propanol, n-Butanol, tert-Butanol)		

OUTCOME / RESULTS

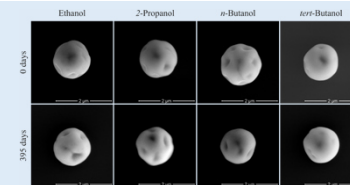
SHELF-LIFE IN

Alcoholic suspension media

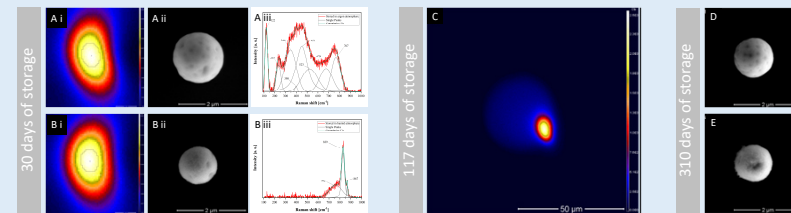
Selection criteria for the solvents:

- Moderate boiling temperature
- Moderate detachment efficiency
- No dissolution behavior of particles

Various atmospheric conditions



Overview of time-dependent SEM study of particles stored in alcoholic solvents.



²³⁸U ion image (i), SEM image (ii), and μ -Raman spectra (iii) of a particle stored for 30 days (A, B), ²³⁸U ion image of particles stored for 117 days (C), and SEM images of particles stored for 310 days (D, E) in argon (A, D) and humid atmosphere (B, C, E).

CONCLUSION

- Moderate to high detachment efficiency of selected solvents
- Shelf-life stability of U-microparticles demonstrated in various alcoholic media over 395 days
- Alteration of U-microparticles in humid atmosphere, U migration onto the surrounding substrate surface
- No observable alteration of particles with other conditions for 310 days

REFERENCES/ ACKNOWLEDGEMENTS

- [1] S. K. Potts et al. MRS Adv. 2022, 7, 134-139.
[2] P. Kegler et al. MRS Adv. 2021, 6, 125-130.

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