Development of Nanoparticles for Radionuclide Generator Systems of Alpha Emitting Radionuclide Pairs

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INTRODUCTION

- Traditional radionuclide generators use organic resins to bind the parent radionuclide and then elute the daughter after it grows in
- Resin-based generators begin to fail with high activities of alpha-emitting pairs such as $^{225}Ac/^{213}Bi$ and $^{230}U/^{226}Th$.
  - High linear energy transfer (LET) of alpha particles damage the resin
- Super-paramagnetic iron oxide nanoparticles (SPIONs) can trap radionuclides and be rapidly precipitated with strong magnets to separate the SPIONs from the daughter radionuclides in solution.
  - ~100 keV of kinetic energy is imparted to the daughter radionuclide, ejecting it from the nanoparticle
  - Metal framework may prove more radio-resistant than organic-based resins.
  - Gold coating may prevent leaching of parent radionuclides
- Monte Carlo simulations with SRIM were performed to show the release of the daughter radionuclides from the nanoparticles and to determine the optimal number of nanoparticles per volume to minimizes re-implantation.

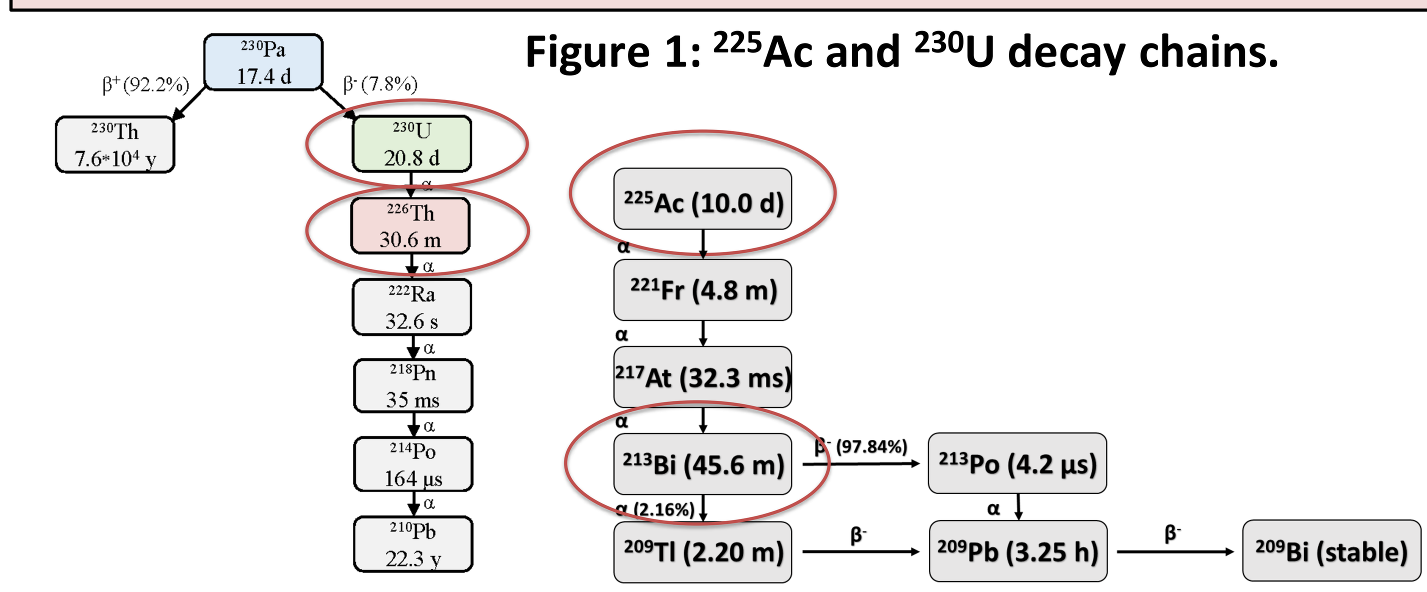

Figure 1: ^{225}Ac and ^{230}U decay chains.

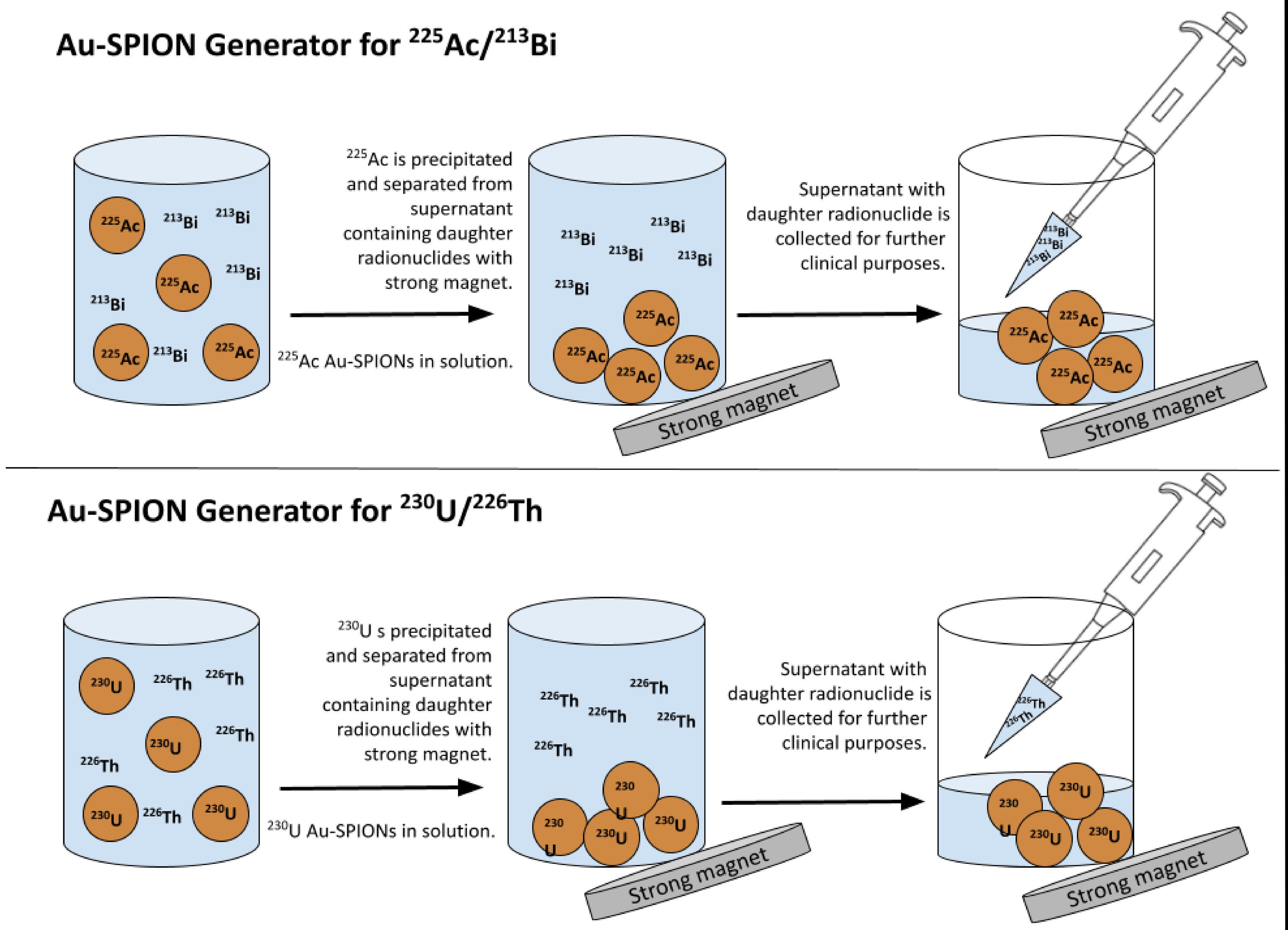

Figure 2: Schematic of the Au-SPION generator process for $^{225}Ac/^{213}Bi$ and $^{230}U/^{226}Th$.

METHODS

Synthesis of Nanoparticles

- Fe (II) and Fe (III) chloride precipitate into ~10 nm particles after the addition of ammonia
- SPIONs are then coated with trisodium citrate to reduce Au (III) onto the surface
- Solution turns from black to reddish-brown color indicating speckles and/or light coating of gold.
- Au-SPIONS are receptive to strong magnets for separation.

RESULTS

Synthesis and Gold Coating

- SPIONs ~10 nm in diameter were synthesized and the surface was covered in ~3 nm gold particles

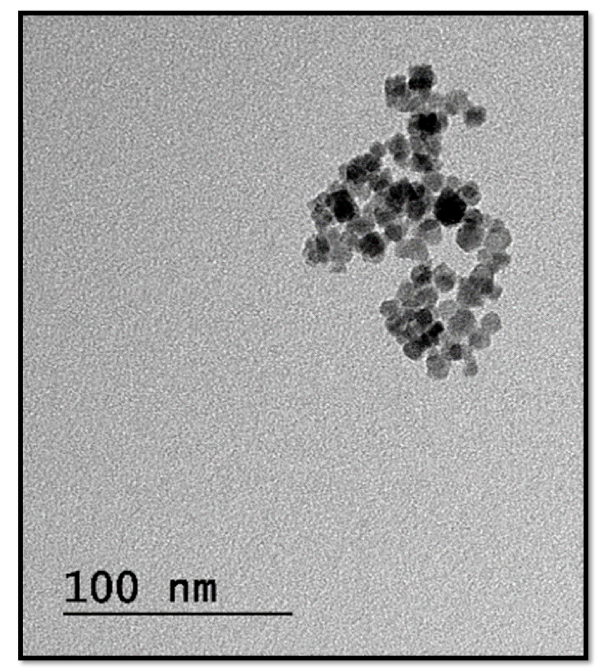

Figure 3: TEM image of ~10 nm SPIONs.

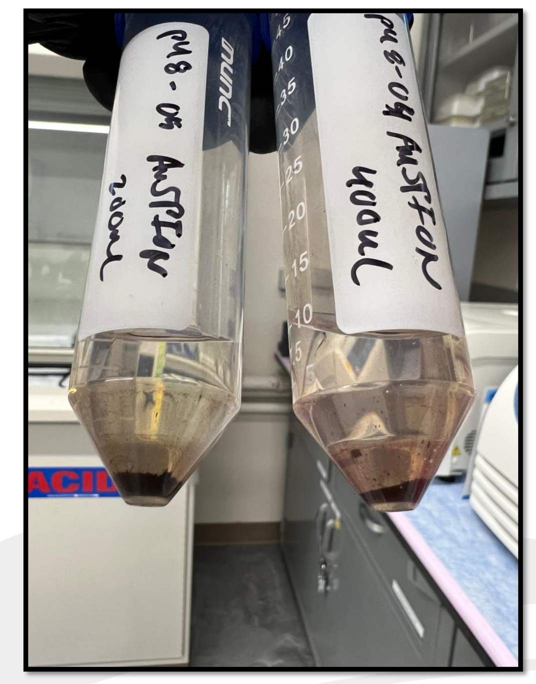

Figure 4: Au-SPIONs in solution.

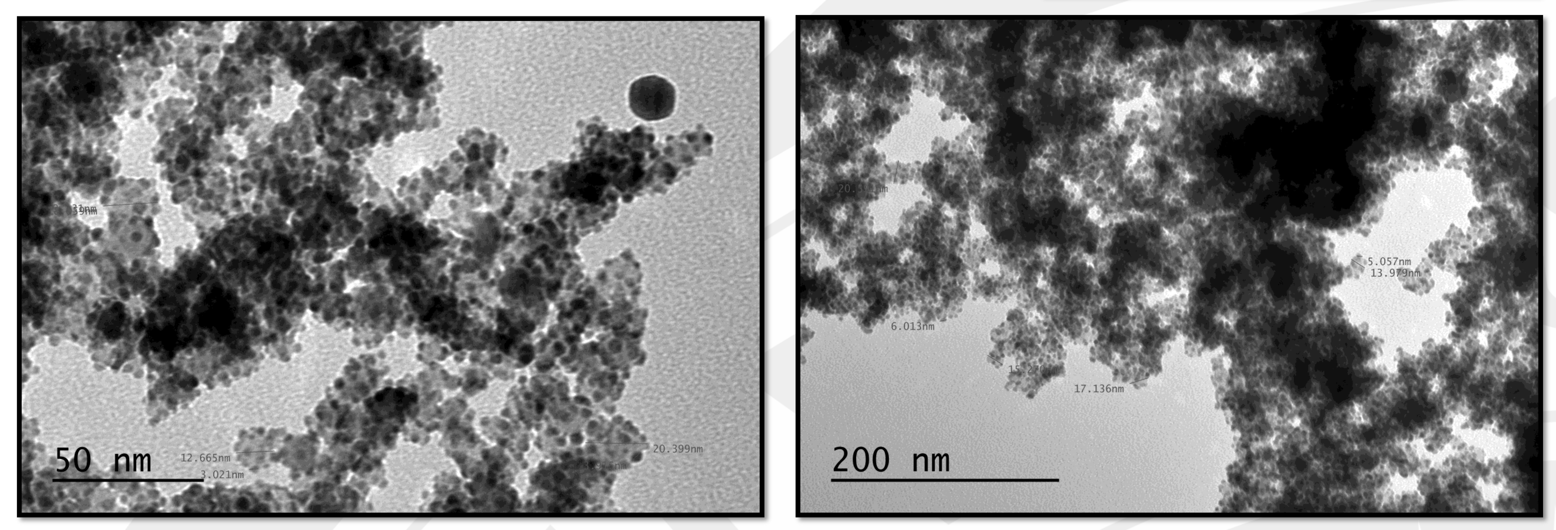

Figure 5: TEM images of Au-SPIONs show ~10 nm SPIONs spotted with ~3 nm gold particles.

Stopping and Range of Ions in Matter (SRIM) Simulations

- SRIM simulations were done to show the release of daughter radionuclides from the nanoparticles
- Ejected daughter radionuclides from one nanoparticle have sufficient energy to re-implant into another nearby nanoparticle if the solution is too concentrated, or if the nanoparticles aggregate
- SRIM was used to estimate the distance ^{225}Ac and ^{230}U daughter radionuclides travel in solution
- This was used to calculate the maximum concentration of particles before re-implantation will occur

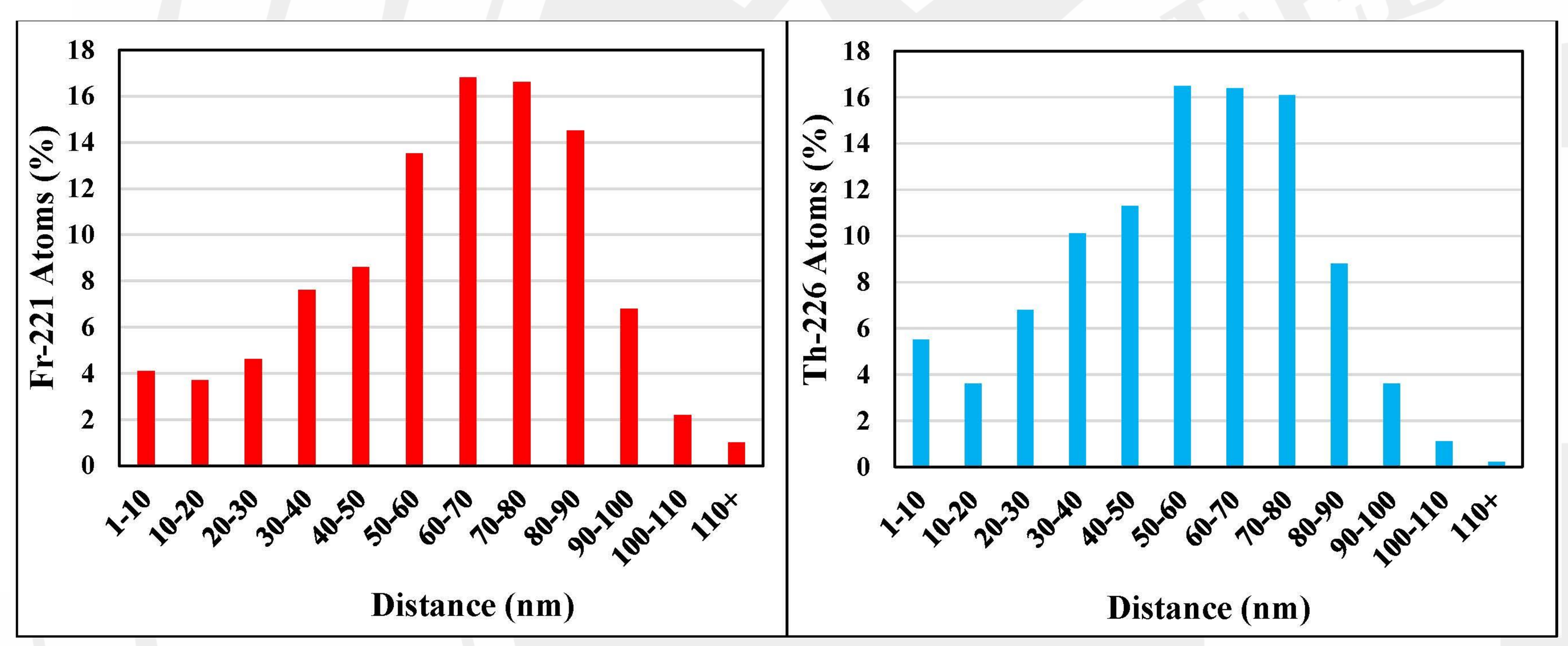

Figure 6: SRIM simulations showing the distribution of distances traveled for ^{221}Fr and ^{226}Th through 5 nm of iron oxide, 2 nm of gold, and then into water.

$$\text{SPIONs} = \frac{\text{Volume } (\text{nm}^3)}{(\text{Max distance daughter radionuclide travels (nm)} + \text{NP diameter (nm)})^3}$$

- $\sim 5.8 * 10^{14}$ *SPIONs/ml for each radionuclide pair*

Addition of PEG to Prevent Aggregation

- Even when dilute, nanoparticles may still aggregate in solution due to surface properties
- Coating the surface with polyethylene glycol (PEG) chains can prevent aggregation
- A barium-iodide assay was used to develop a standard curve so the amount of PEG added per nanoparticle may be determined

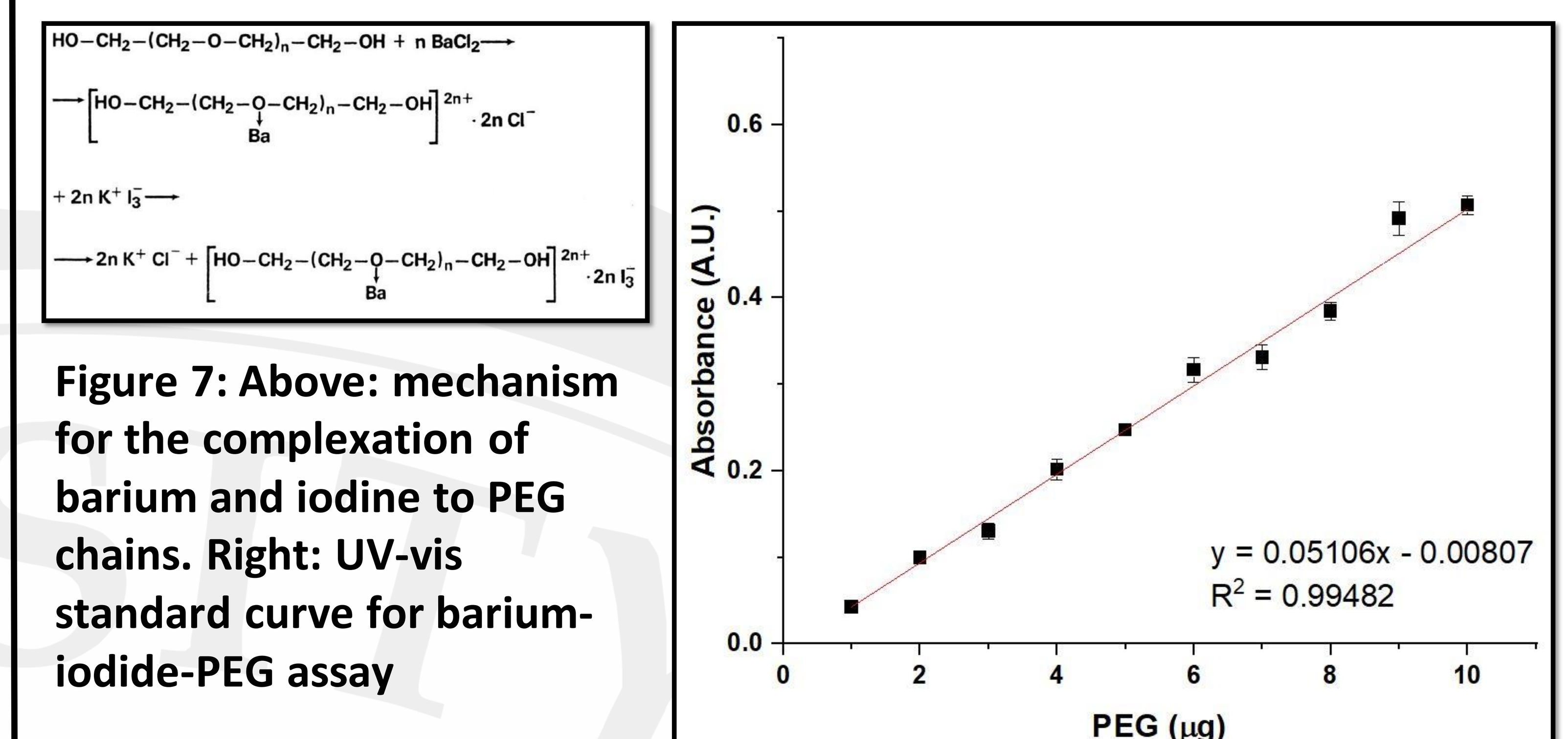

Figure 7: Above: mechanism for the complexation of barium and iodine to PEG chains. Right: UV-vis standard curve for barium-iodide-PEG assay

CONCLUSIONS

- SPIONs were successfully synthesized and covered with small gold particles
- SRIM simulations were done to optimize the number of nanoparticles in solution to prevent re-implantation of recoiling daughter radionuclides
- Standard curve for PEG was developed to quantify PEG addition to nanoparticles in the future

FUTURE WORK

- Achieve a uniform gold coating
- Addition of PEG molecules to the surface of the Au-SPIONs to prevent aggregation in solution
- Investigate the recovery of ^{213}Bi and ^{226}Th
- Monitor for any degradation of the nanoparticles and/or loss of ^{225}Ac and ^{230}U
- Scale up generator to larger activities

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