Optimization of the efficiency of diamond based alpha-sensors for spectrometry in aqueous solutions

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Optimization of the Efficiency of Diamond Based Alpha Spectrometers in Solutions

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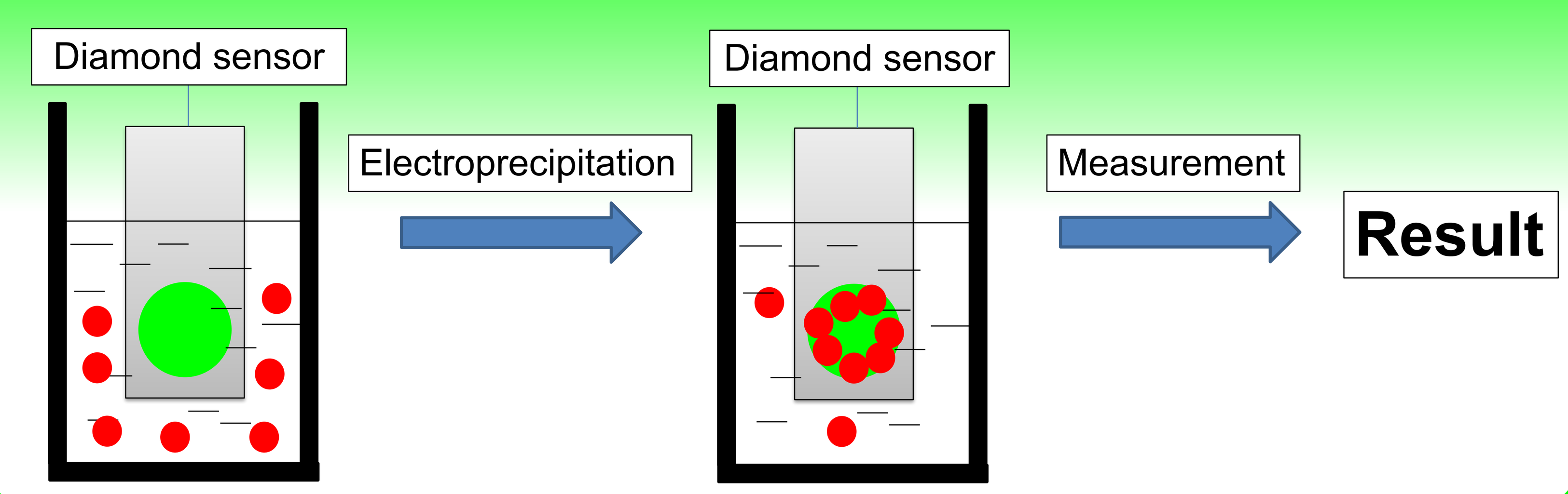
Introduction

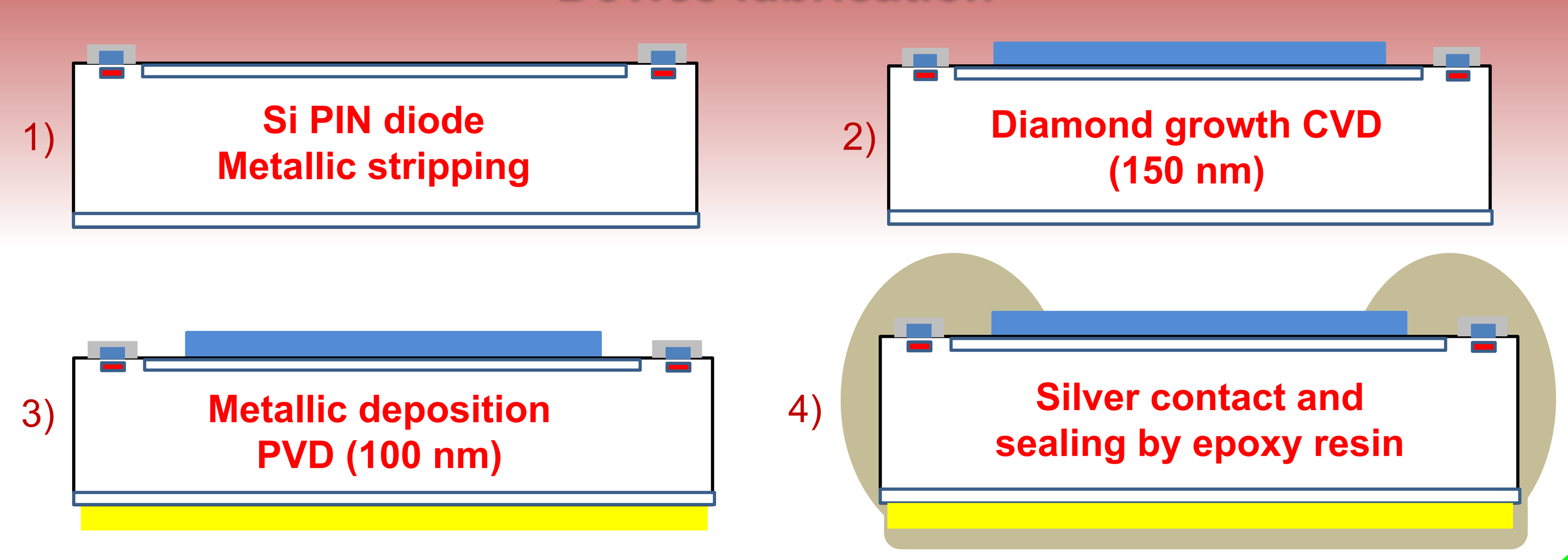
- Nuclear industry or nuclear accident (e.g. Tchernobyl 1986 and Fukushima 2011) generate non negligible amount of radioactive sources in the environment. Among of radionuclides, actinides are very hazardous due to their radiation emission and chemical toxicity, and more than their long half-life to hundreds thousands years.
- We propose herein an innovative device consisting of a diamond electrode and a conventional silicon alpha sensor, directly detecting alpha activity in solutions [1]. Diamond electrode is dedicated to collect alpha emitter onto sensor by electroprecipitation technique.
- This work aims at optimizing electroprecipitation step by fine tuning parameters of electrochemical process such as: pH of electrolyte, current density, electrolyte nature, geometry of electrochemical cell, sensor area and precipitation times.

[1] de Sanoit et al. French Patent #105815, Oct. 7,2010

Operation mode (direct detection in solutions)



Device fabrication



Electroprecipitation process

SO_4^{2-} electrolyte [2], [3], [4]

$2H_2O + 2e^- \rightarrow H_2\uparrow + 2OH^-$

$Ac^{3+} + 3OH^- \rightarrow Ac(OH)_3\downarrow$

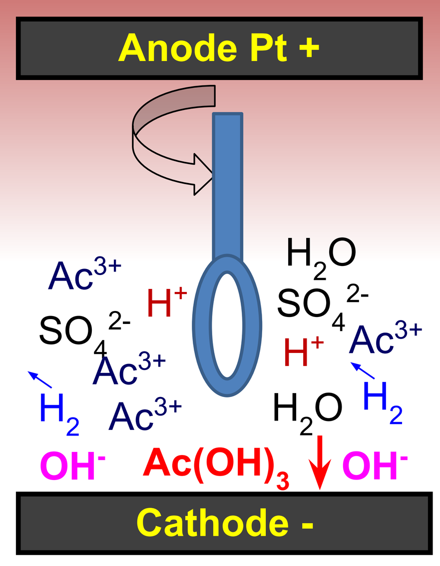
NO_3^- new electrolyte proposed

$NO_3^- + H_2O + 2e^- \rightarrow NO_2^- + 2OH^-$

$NO_3^- + 3H_2O + 5e^- \rightarrow 1/_2N_2 + 6OH^-$

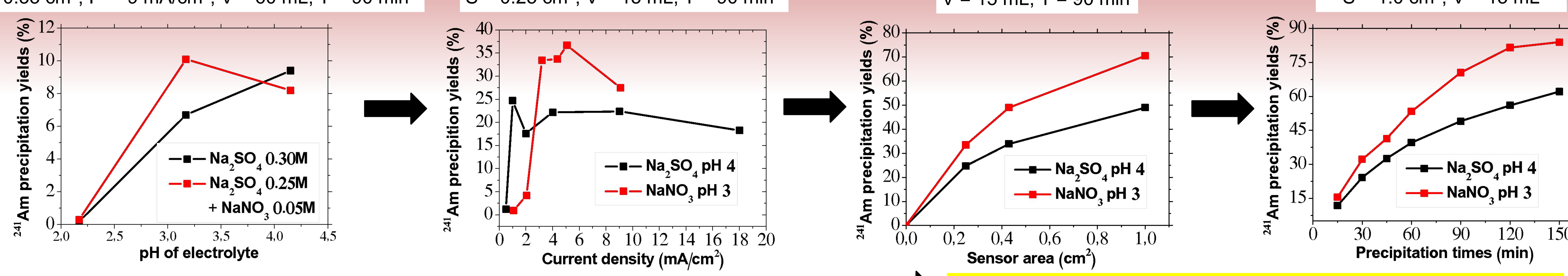
$NO_3^- + 7H_2O + 8e^- \rightarrow NH_4^+ + 10OH^-$

- Interest of NO_3^-: generation of more hydroxyl limitation of hydrogen gas



[2] Talvitie 1972; [3] Hallstadius 1984; [4] Tsoupko-Sitnikov et al. 2000

Optimising the electrochemical process: ^{241}Am activity in solutions = 5.96 Bq



- $\frac{NaNO_3}{Na_2SO_4}$ = 1.4 times

Optimal conditions: 120 min, - 3 mA/cm², 15 ml of 0.3 M $NaNO_3$ pH 3

Actinides precipitation

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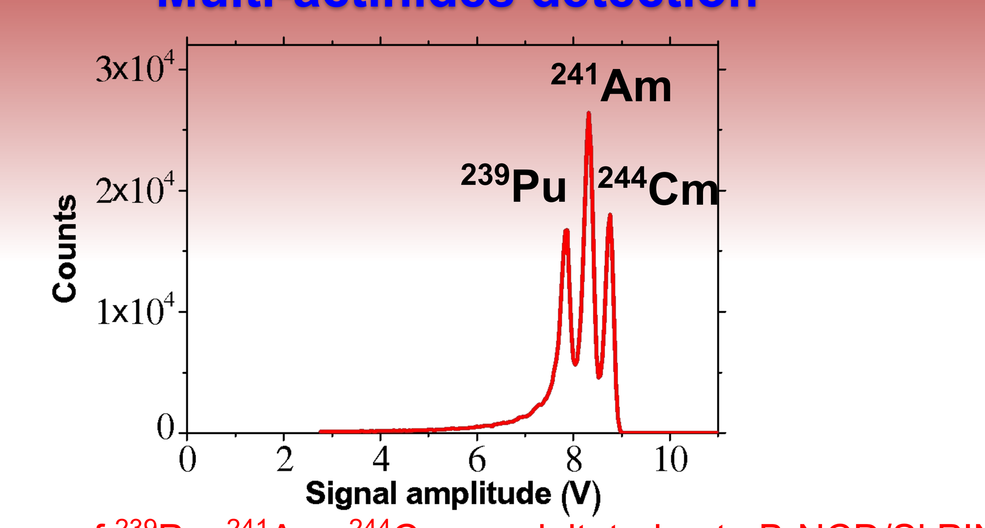
Actinides	Precipitation yields (%)	Sensor cleaning (%)
^{241}Am (5.96 Bq)	81.5	> 99.9
^{239}Pu (5.13 Bq)	49.9	> 99.9
^{244}Cm (4.40 Bq)	78.2	> 99.9

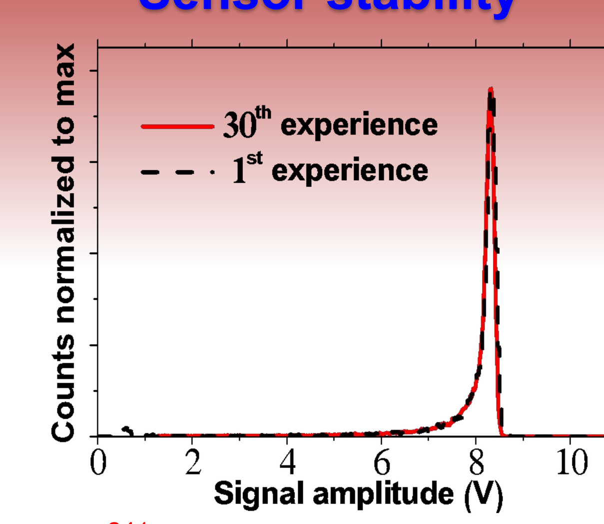
- Fast cleaning with high efficiency > 99.9 % by anodic courant of +2mA/cm² in acid solution (Na_2SO_4 0.3M pH 3)

Multi-actinides detection



Spectrum of ^{239}Pu, ^{241}Am, ^{244}Cm precipitated onto B-NCD/Si PIN

Sensor stability



Spectrum of ^{241}Am precipitated onto B-NCD/Si PIN

Limit of detection (LOD) = 0.24 Bq/L

- Calculation from alpha measurement background noise (7 counts for 2h)
- LOD sensor = 2.71 + 3.29$\sqrt{2\mu_B}$ (counts)
- LOD in solutions = LOD sensor * $\frac{1}{T}*\frac{1}{V}*\frac{1}{R}$ (Bq/L)

μ_B: counting number
R: precipitation yields (%)
T: times measurement (second)
V: electrolyte volume (L)

Conclusion

- Fabrication of an innovative detector based diamond/silicon for direct alpha detection in solution with limit of detection up 0.24 Bq/L
- Sensor could be reused after a fast cleaning by applying a weakly anodic current of +2 mA/cm²
- Sensor showed very high stability due to unique properties of diamond electrode
- Precipitation efficiency is improved up about 80 % for ^{241}Am and ^{244}Cm, and 50 % for ^{239}Pu
- Actinides electroprecipitation in $NaNO_3$ is 1.4 times more efficient than in Na_2SO_4 due to nitrate reduction generate the limitation of hydrogen gas than water reduction

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