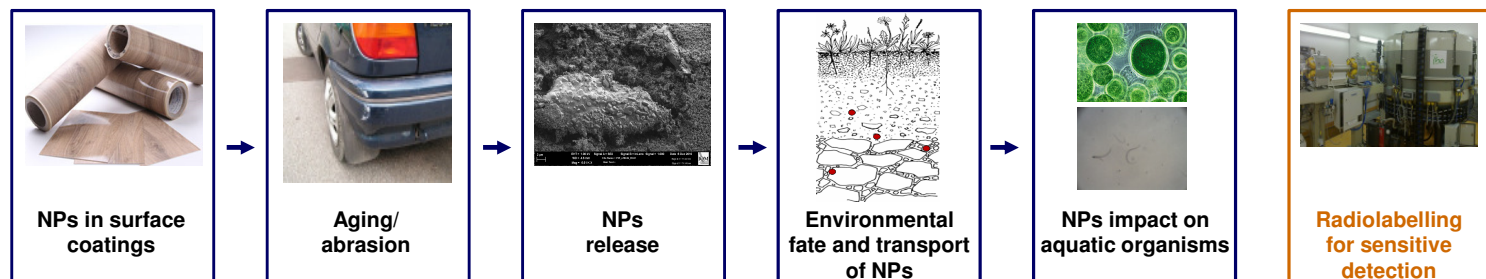


Motivation



→ Radiolabelling for detection of NPs in complex media – even at low concentrations ←

Radiolabelling of engineered nanoparticles

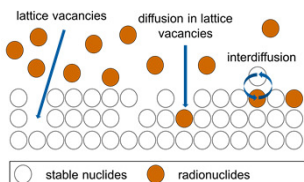
Isotopic radiolabelling of commercial TiO₂ and Ag⁰ nanopowder

Labelling method:

→ Diffusion of radionuclides into NPs via low temperature annealing

Materials:

→ Ag⁰ (Sigma-Aldrich, d_p < 100 nm)
→ TiO₂ (AEROXIDE® P25, Evonik Degussa, d_p = 21 nm)



Commercial radionuclides:

→ ⁴⁴Ti (T_{1/2} = 63 a)
→ ^{110m}Ag (T_{1/2} = 249.9 d)

Production with a cyclotron:

→ ⁴⁵Ti (T_{1/2} = 185 min)
→ ¹⁰⁵Ag (T_{1/2} = 41 d)



Solid state target system
Nirta Solid Compact



Target disk:
Body: Al Ø 24 mm
Target: Sc Ø 12 mm

Radiolabelling yield for Ag⁰: > 99 %

→ (0.2 – 1000) kBq ^{110m}Ag⁺/mg Ag⁰ NPs
→ Low temperature annealing
T = 50°C – 105°C, t = 3 h

Radiolabelling yield for TiO₂: > 99 %

→ (0.2 – 10) kBq ⁴⁴Ti⁺/mg TiO₂ NPs
→ Low temperature annealing
T = 180°C, t = 2 h

→ Up to 140 MBq ⁴⁵Ti⁺/mg TiO₂ NPs
→ Low temperature annealing
T = 180°C, t = 2 h

Specific activities and limits of detection

[^{110m}Ag]Ag⁰ 1 MBq/mg → 125 ng/L
[⁴⁴Ti]TiO₂ 0.01 MBq/mg → 7.500 ng/L (costly nuclide!)
[⁴⁵Ti]TiO₂ 140 MBq/mg → 0.5 ng/L

Proton activation of TiO₂ and Ag⁰ nanopowder using a cyclotron

Ag⁰-NP

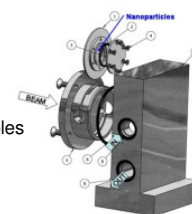
→ ¹⁰⁷Ag(p,3n)¹⁰⁵Cd → ¹⁰⁵Ag
→ ¹⁰⁷Ag(p,2n)¹⁰⁵Ag
¹⁰⁵Ag: t_{1/2} = 41.29 d / β⁺ 100 %



TiO₂-NP

→ ⁴⁸Ti(p,n)⁴⁸V
⁴⁸V: t_{1/2} = 15.97 d / β⁺ 100 %

Target system for irradiation of nanopowder samples



Specific activities and limits of detection

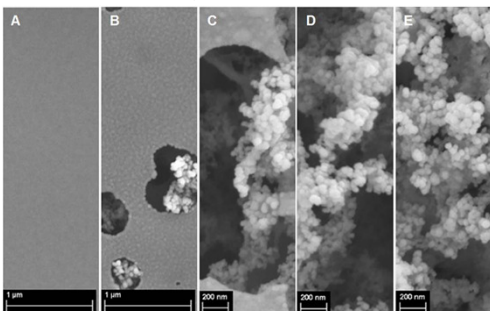
[¹⁰⁵Ag]Ag⁰ 69 kBq/mg → 1.8 µg/L

Scanditronix MC40, JRC

[⁴⁸V]TiO₂ 36 MBq/mg → 2 ng/L

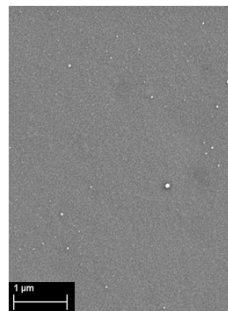
Application of radiolabelled NPs for release studies

labile surface coating → NP release is visible



Time-dependent degradation of a polyacrylate TiO₂ nanocomposite by UV-A-irradiation (intensity ~ 15 mW/cm²) ;(A) original, (B) after 2 days (50.000x); (C) 4 days, (D) 8 days, (E) 16 days (75.000x); SEM micrographs: Leibniz Institute for Surface Modification.

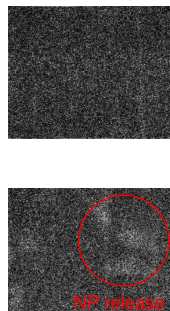
stable surface coating



Degradation of a **stable** TiO₂ nanocomposite by UV-A-irradiation for 10 days (25.000x).

No visible NP release, but ~ 4 % of the applied TiO₂ is released and was only detected by measuring radioactivity!

autoradiography



References

Isotopic radiolabelling of Ag⁰-NPs

Hildebrand & Franke *J Nanopart Res* 14 (2012)

Proton activation of TiO₂-NPs

Abbas K et al. *J Nanopart Res* 12 (2010)

Gibson N et al. *Arch Toxicol* 85 (2011)

Holzwarth U et al. *J Nanopart Res* 14 (2012)

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