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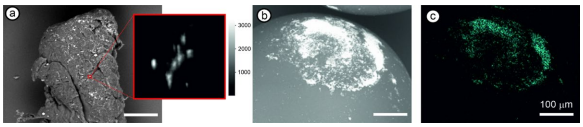

Spatially resolved transformation of ZnO nanomaterials adsorbed to microplastics in environmentally relevant solutions

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Understanding the chemical stability of zinc oxide (ZnO) engineered nanomaterials (ENMs) in realistic environmental scenarios is key for predicting their bioreactivity. Microplastics may act as transportation vectors of ENMs and can arise after UV-exposure and mechanical abrasion of the tonnes of plastic being disposed every year. While these microplastics may be cytotoxic to (micro)organisms, they can also sorb hazardous chemicals and materials, enhancing their potential toxicity.

Initial scanning electron microscopy-energy dispersive spectroscopy (SEM-EDX) analysis demonstrated the heterogeneous adsorption of ZnO ENMs (80-200 nm size) in both polyethylene (PE, 300 mm diameter) and polystyrene beads (PS, 900 mm) at the microscale. Complementary X-ray fluorescence (XRF) analysis at the hard X-ray nanoprobe (I14 beamline) [1] revealed nanometric aggregations as well as individual nanomaterials adsorbed on microplastic's surfaces (red-squared area, Fig. 1), illustrating the need for spatially resolved analysis.

We have previously developed a multi-energy XRF spectroscopy method at the I14 beamline to gain information about the intermediate Zn-species generated in environmentally relevant conditions, as a function of time, during *in situ* incubations. Using this technique, we demonstrated that ZnO ENMs largely transform to Zn-sulfide and Zn-phosphate in real wastewater solutions within hours [2], which present different toxicity and solubility. Therefore, the same methodology is applied to the study of the Zn speciation in microplastics' surfaces incubated with ZnO ENMs in synthetic and seawater environments, to determine if they may pose a threat to seawater microorganisms, and to reveal heterogeneities/ aggregations at the nanoscale. Fundamental awareness of how the chemistry of the individual particles change within complex aquatic systems will reveal the critical physicochemical properties determining environmental damage and deactivation of engineered nanomaterials and microplastics used in consumer products.

[1] Quinn *et al.* J. Synchrotron Rad. (2021) 1006-1013; 10.1107/S1600577521002502

[2] Gomez-Gonzalez *et al.* Adv. Sustainable Syst. (2021) 2100023; **10.1002/adsu.202100023**

Figure 1. SEM analysis of a) PE and b) PS microplastics after incubation with ZnO ENMs. c) EDX analysis confirmed the Zn adsorption on the microplastics (blue). The zoomed area analysed at I14 by XRF (red-square, 9x8 microns) revealed ZnO aggregation in the PE surface.