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Report:

The purpose of this experiment was to investigate the possibilities of performing truely three-dimensional compositional/structural analysis of microscopic particles of environmental origin with the ultimate aim to be able to measure alterations that occur either in the composition, in the structure or in both of specific parts of airborne and other particulates as they interact with their environment.
Previously, a number of experimental sessions during 1995 and 1996 had revealed the possibility to obtain correlated compositional/structural information from microscopic fly-ash particles of different origin through combined μ-XRF/μ-XRD 2-dimensional mapping. For these experiments, capillary formed X-ray beams of 1 or 2 μm cross-section were used. Although the elemental and crystallographic distributions obtained in this way have very high lateral resolution (among the best reported in the μ-XRF literature) and revealed considerable details of the internal structure of the particulates and on the compositional contrast between the surface layers and the interior parts of large fly-ash particles, the 2-dimensional scanning strategy employed in these experiments yielded only a-dimensional projection images of the 3-dimensional structure of the particles. In the resulting images, therefore ambiguities exist on, e.g. the exact location of certain enriched areas, on the shape and orientations of the channels linking the inner parts of the particulates with the outer shell etc.

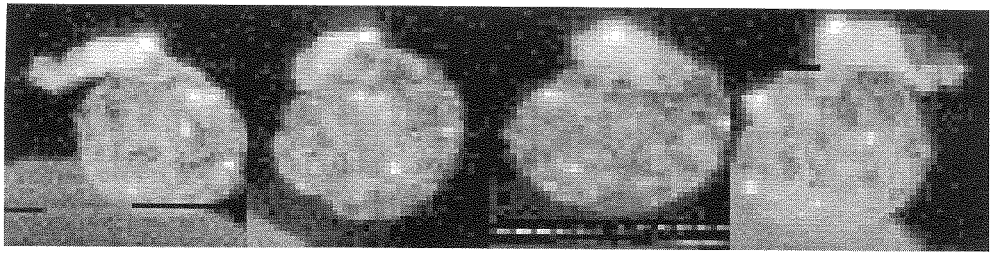

Figure 1. Fe maps of a fly-ash particle under different orientations (0,45,90,180 degrees)

In order to extend these investigations towards a truely 3-dimensional analysis free of interpretation errors, the previously employed conventional XY scanning strategy was replaced by a tomographic approach, involving a rotation/translation scanning scheme. To this effect, individual fly-ash particles of size 30-50 μm were placed on the tip of a thin glass capillary and mounted onto a goniometer head which was aligned to the axes of a rotation/translation stage.
In this way, a number of cross-sections at different heights through large fly-ash particles were obtained with a resolution of ca. 2-4 μm. Combination of the various sections provides a fairly detailed picture of the inner structure and elemental distribution inside the particulates. In order to verify the validity of the results, under a number of observation angles, conventional XY projection images of the same particles were collected. In the obtained sections, the position, size, shape, crystallographic nature and composition of various areas inside the particulates showing high concentrations of heavy metals such as V, Fe, Ni etc. could be located in an objective way. As an example, the Fe sinogram and reconstructed Fe-section of the particle shown in Fig. 1 are shown in Fig. 2.

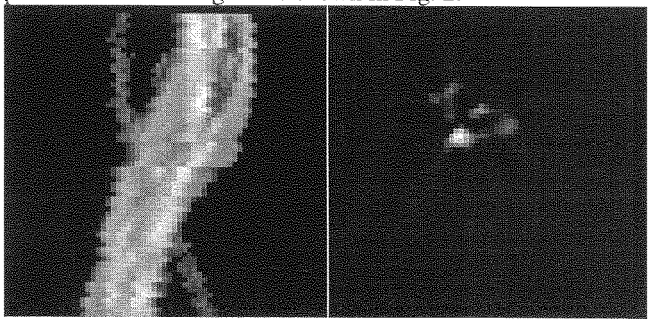

Figure 2a Sinogram (60 x 60, 2 μm x 3 degrees), b:reconstructed Fe tomogram (60 x 60, 2 μm x 2 μm)

On the other hand, also a number of problems were identified during the experiment: one practical problem concerns erratic movements of the rotation axis relative to the beam position in the plane of rotation with an amplitude of ca. 2-3 μm. This wobbling motion, probably caused by random variations in the grease thickness on the ball-bearings of the rotation stage, introduces low frequency noise in the sinograms, resulting in diffuse reconstructed tomograms and a reduction of the lateral resolution. In the present case, where fairly large objects (- 50 pm) are investigated, this does not present a problem in practice, but this effect does put a limit on the ultimate resolution of obtained tomograms in case smaller beams (<= 1 μm) are used.

A second, more fundamental problem is the self-absorption of the fluorescent radiation, even in the low-density material investigated here. This effect can be very clearly observed in the sinograms of the elements Ca, V and Fe.(Fig. 2a) Correction of these effects is in principle possible, but only if the absorption of the transmitted beam is measured together with the fluorescent signals. This was not done during this experiment. Validation of the procedure employed for correcting of the self absorption in these heterogeneous samples will require measurements on heterogeneous samples of simplified structure and composition in the near future.