Electronic Supplementary Information

The ANCIENT CHARM project at FRM II: three-dimensional elemental mapping by Prompt Gamma Activation Imaging and Neutron Tomography

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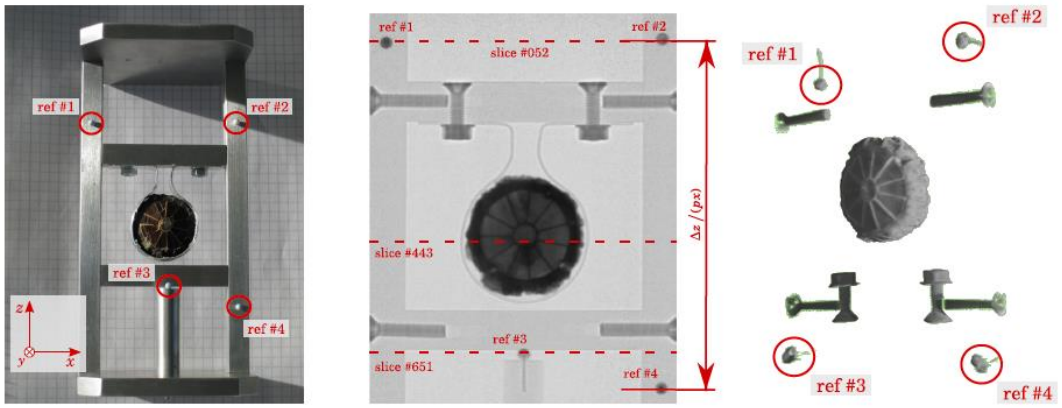
S.1. Sample positioning and registration

S.1.1. Introduction

A critical task of analysis of the samples is the correct positioning during the measurements and the correct registration, i.e. alignment, of the different datasets from NT, PGAI, etc., afterwards. In general, the facilities where different datasets are acquired may have different geometries, i.e. different sample sample stages, other detector positions, etc., hence they do not share a common (x, y, z, ω)-coordinate system. To address these problems, it was decided to establish a common reference coordinate framework, by attaching reference markers to the sample support. These reference markers should be small enough to define the location with the desired precision (≈ 1 mm accuracy was seen as adequate, with respect to the expected measurement resolutions), while still being large enough to be easily detectable by the different neutron methods. At least four reference markers, that must not lie in one plane, are needed for the unique alignment of a sample in three-dimensional space. Another important request is that the markers must never cover the analyzed sample during a rotation, because this will, due to the high absorption properties of the markers, disturb the NT reconstruction process. Needle heads of about 3 mm diameter that were painted with gadolinium blended nail-polish, proved to be suitable (see figure S.1). They are clearly visible in the radiographies and the final reconstructions (fig. S.1(b) and (c)) and are clearly recognizable in the PGAI spectra. For a determination of their position with an accuracy of the desired 1 mm one should refer all coordinates to the center of the markers, which can be determined quite accurately.

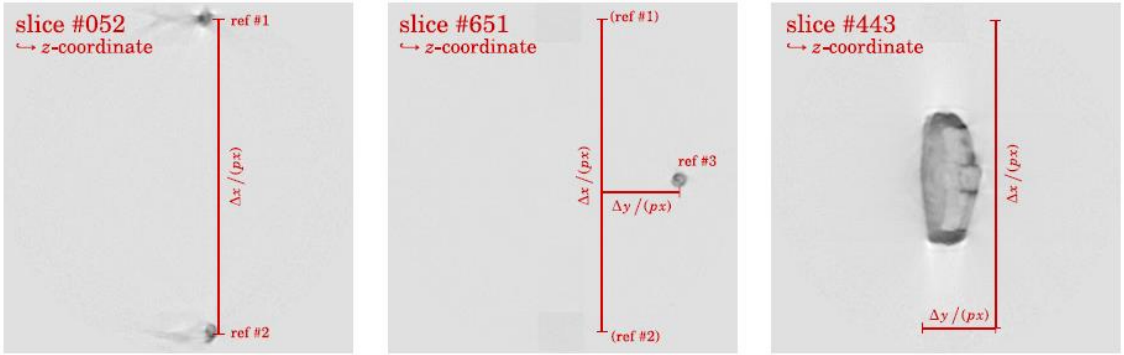


(a) Reference markers at four distinct positions on the sample holder for a fibula object

(b) Normalized radiography of the fibula. The reference markers are clearly visible and can be used for (pixel-mm)-conversion

(c) Reference marker positions in the final NT reconstruction

Figure S.1.: Reference markers from needle heads painted with Gd blended nail polish used for positioning and data-set registration



(a) *x*-coordinate pixel-mm conversion

(b) *y*-coordinate pixel-mm conversion

(c) Determination of a measurement position inside the object

Figure S.2.: Determination of measurement positions with the help of the reference markers

S.1.2. Positioning

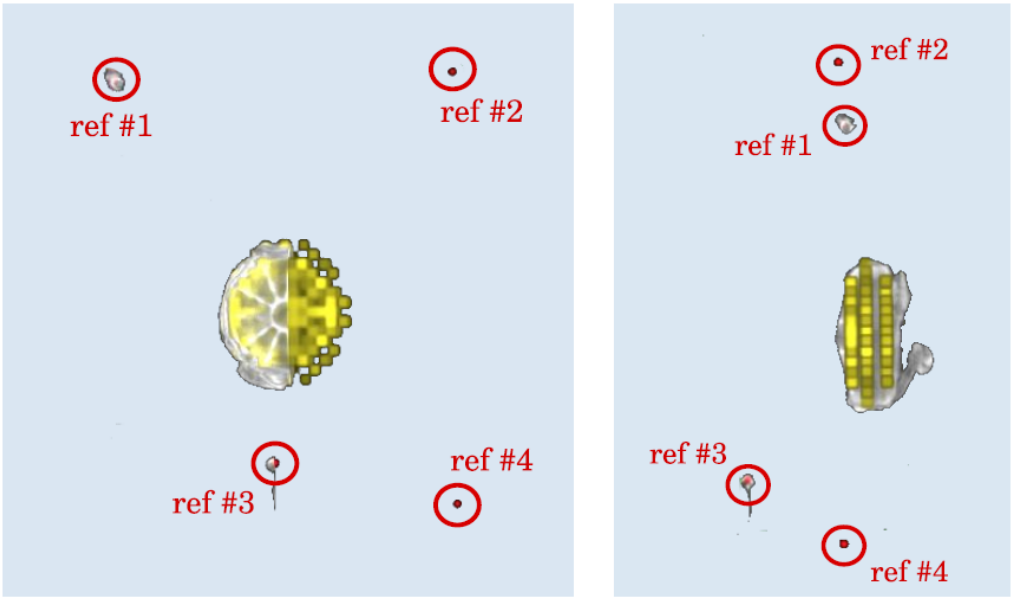
The reconstructions of a NT measurement deliver an ideal tool to identify interesting positions for e.g. 3D-PGAI measurements. They deliver the locations of interest in units of reconstruction pixels and slices, which have to be converted to the coordinate system of the measurement setup. In praxis this is normally given through (x, y, z, ω)- steps of the stepper motor driven sample manipulator. This conversion can easily be done with the help of the reference markers. Their positions in reference to a known position of the sample table, i.e.

the center of the supporting base plate, can easily be measured to high accuracy, with e.g. a caliper. This, together with their pixel-positions in the NT gives the desired (pixel-mm)-conversion (figure S.2). Together with the known isocenter position the final measurement positions in motor steps are readily available.

It may be advisable to rotate the sample during the measurements, e.g. to minimize self-absorption and -shielding effects. In case of such measurements the data have to be transformed with a rotation matrix with the (x, y)-coordinates given relative to the center-of-rotation of the sample table.

S.1.3. Registration

Because all measurement data-sets were finally available with positions relative to the reference markers they can easily be aligned for detailed combined analysis and interpretation or mutual correction. In figure S.3 the combination of a NT reconstruction and a single full 3D-PGAI dataset is shown. Both 3D datasets have been imported into a standard visualization software. For the purposes of presentation and interpretation an alignment "by eye" utilizing the reference marker positions is sufficient. For correctional calculations the real relative coordinates of measurement positions and reference markers have to be used.

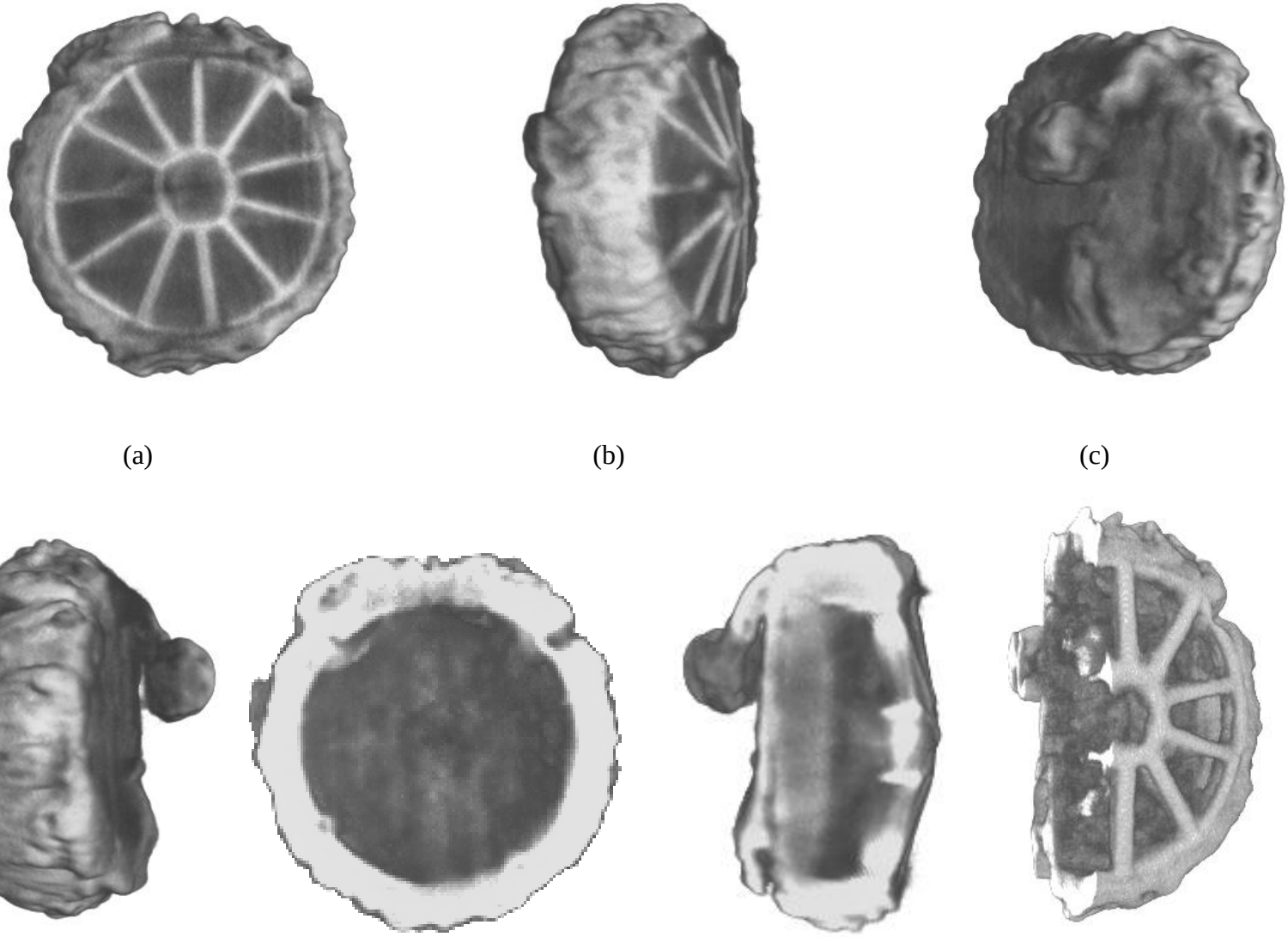


(a) Front view: The NT visualization has been cutted in the middle of the fibula.

(b) Side view showing the three measurement layers

Figure S.3.: Combined data-sets of a NT reconstruction (gray) and the 3D-PGAI map (yellow) in the fibula sample

Figure S.4.: Neutron tomography images and virtual cuts



(a) (b) (c)

(d) (e) (f) (g)

Figure S.5.: Iron distribution

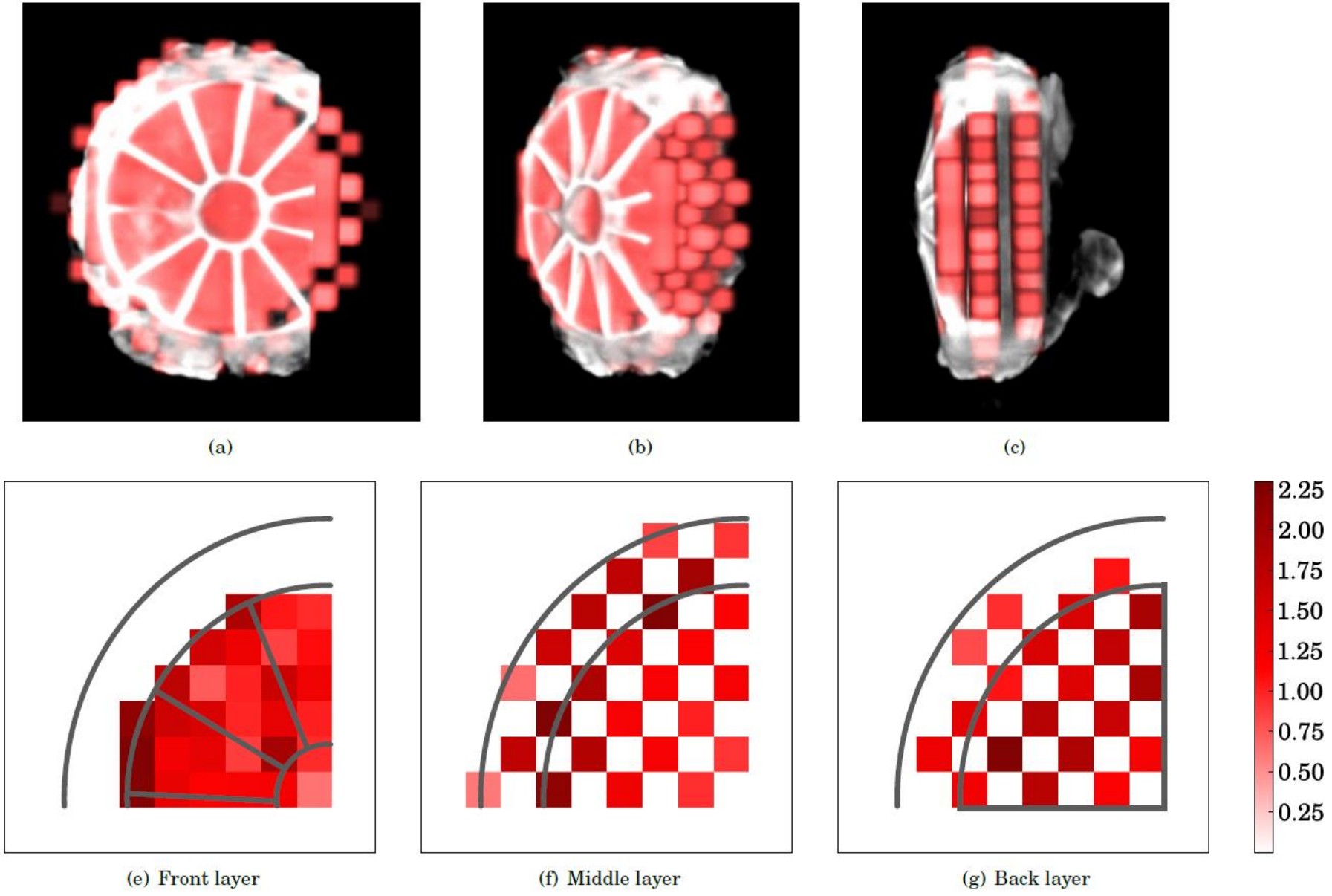


Figure S.6.: Sulfur distribution

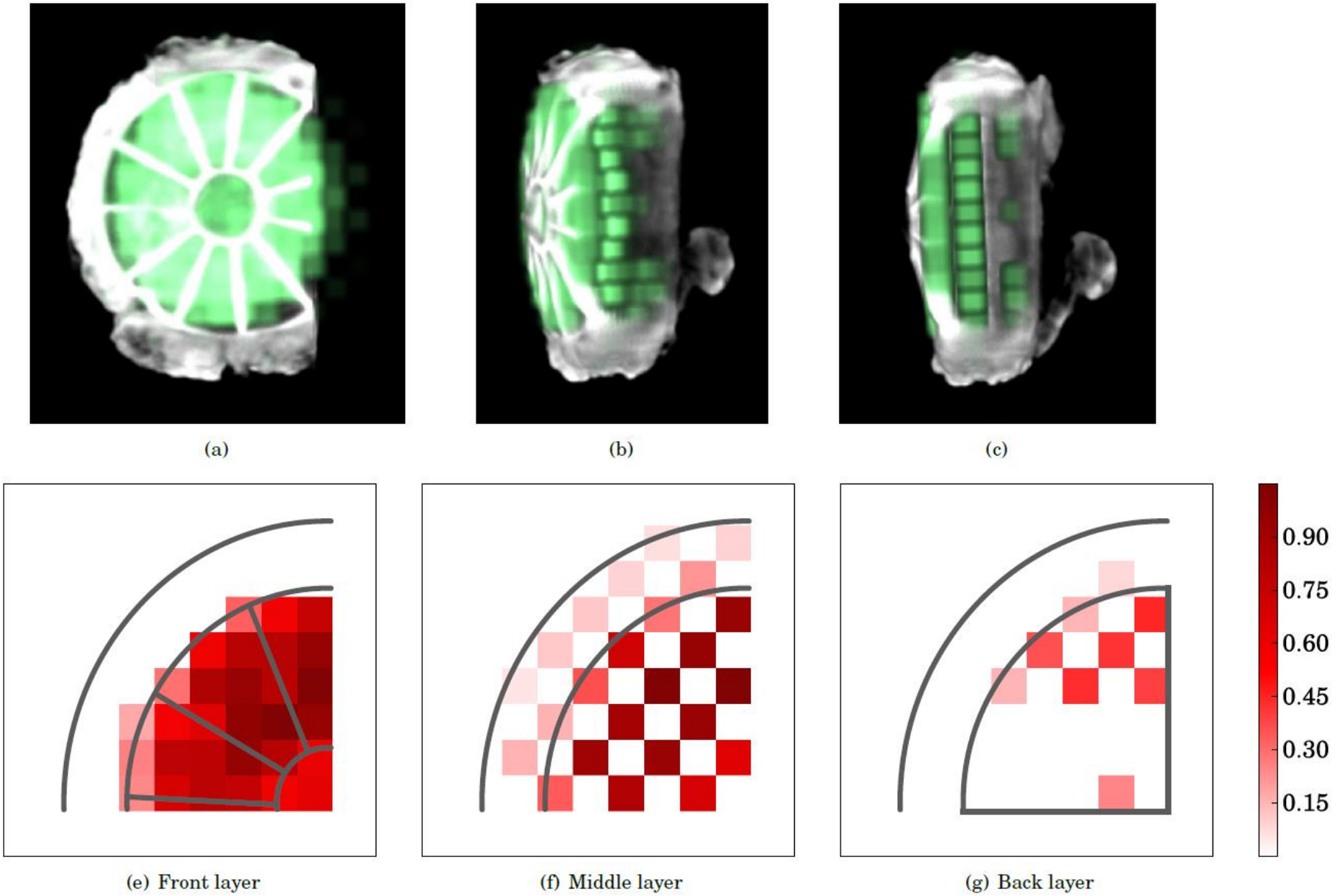


Figure S.7.: Gold distribution

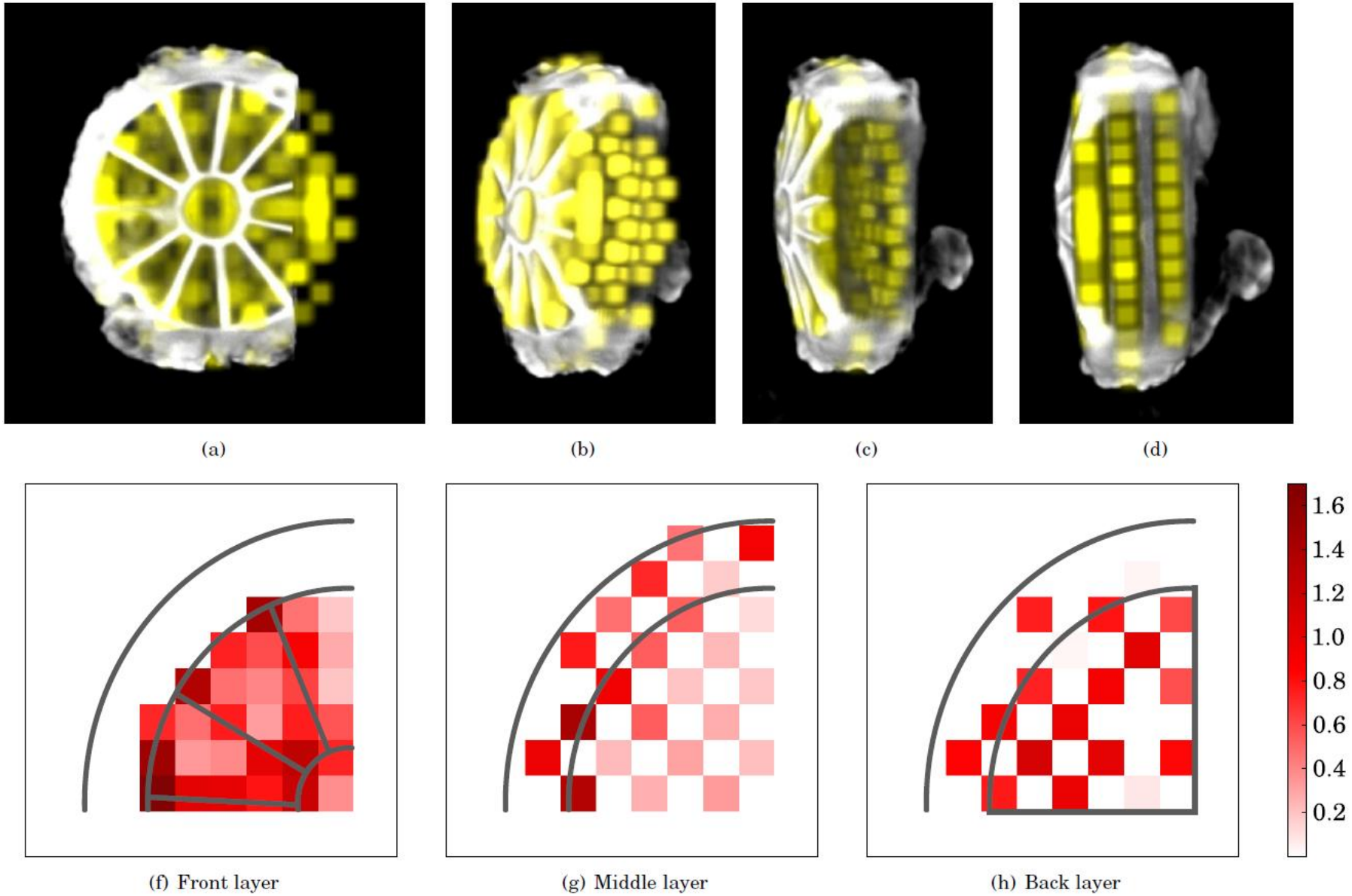


Figure S.8.: Copper distribution

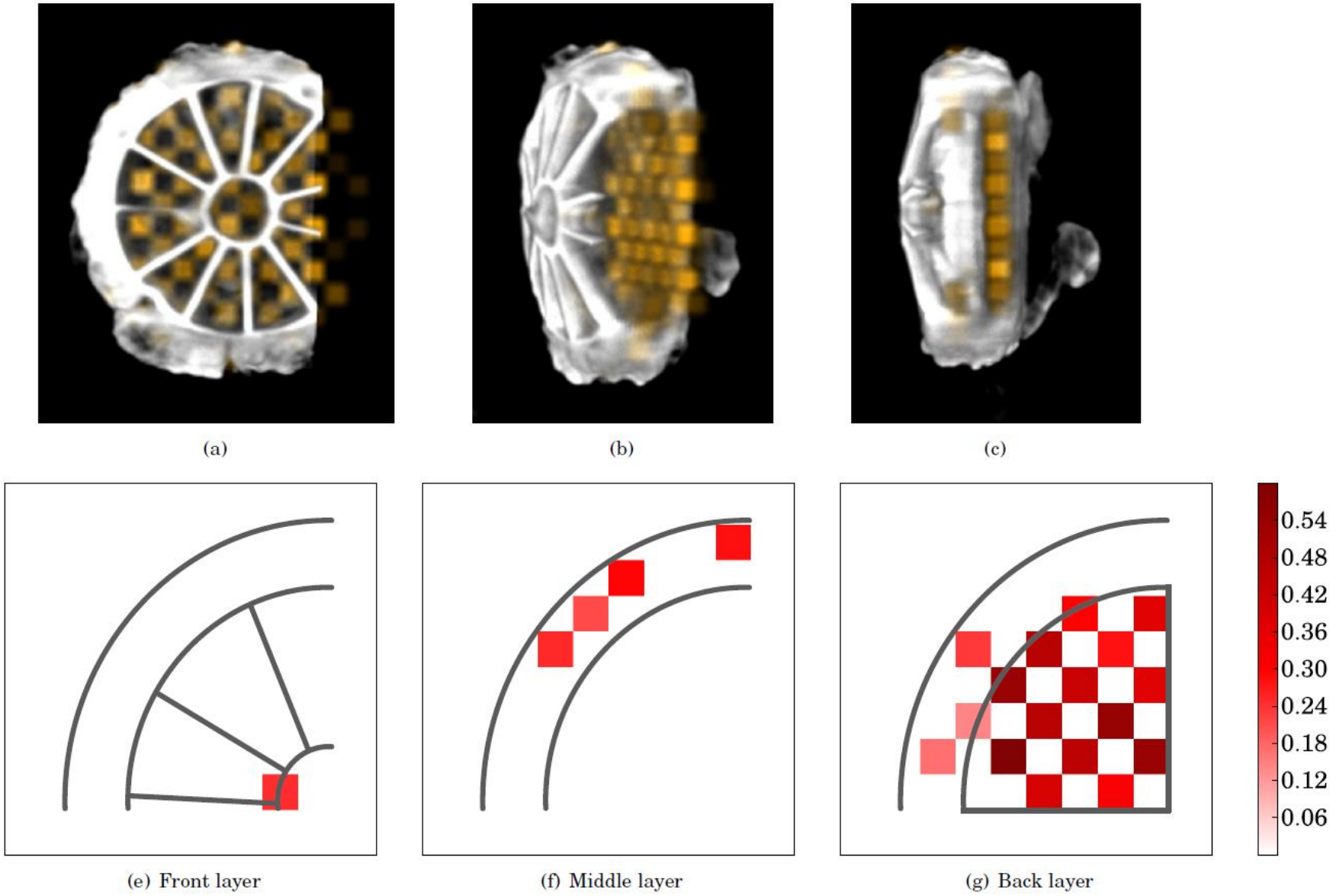
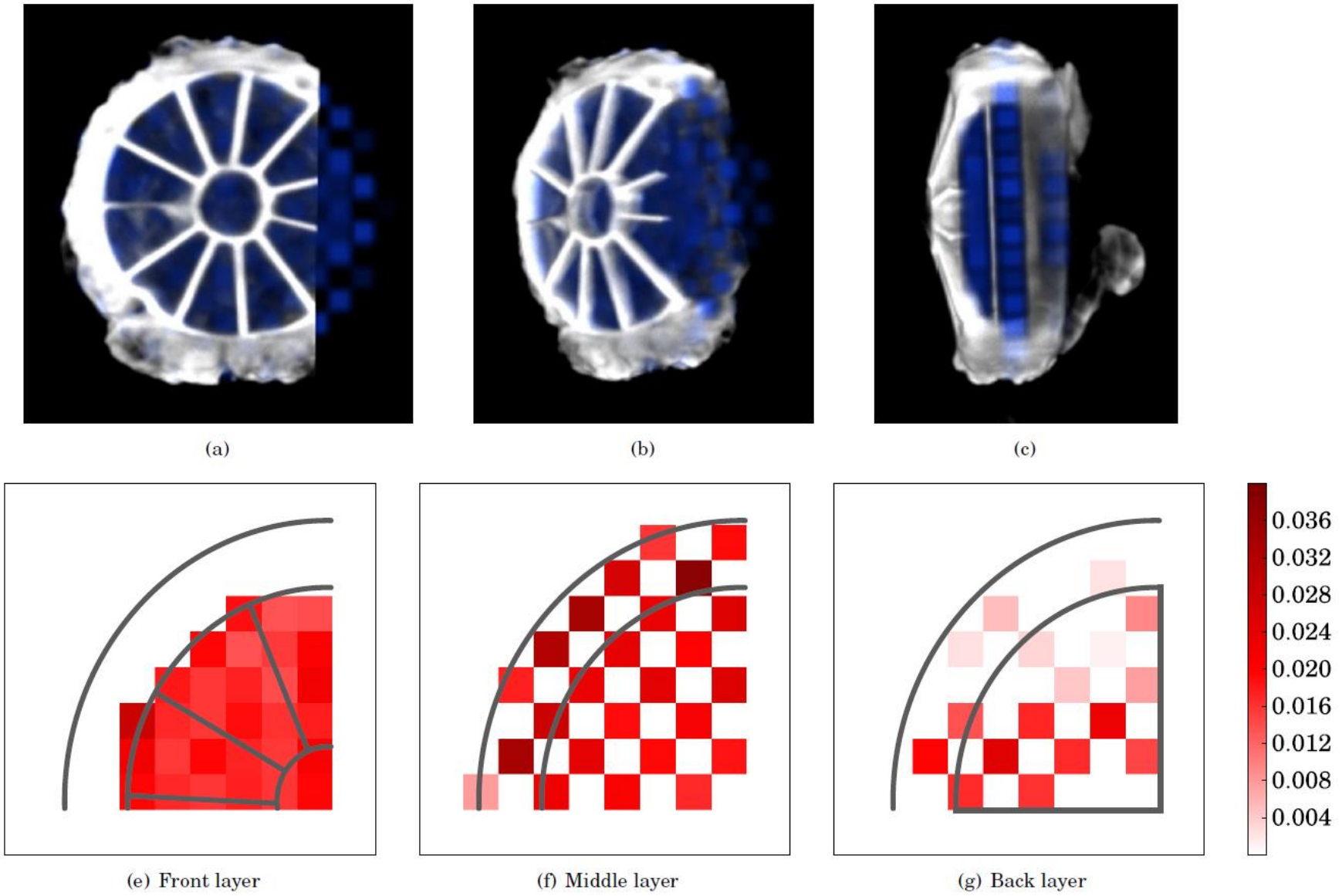


Figure S.9.: Hydrogen distribution



(a) (b) (c)

(e) Front layer (f) Middle layer (g) Back layer

Figure S.10.: Silver distribution

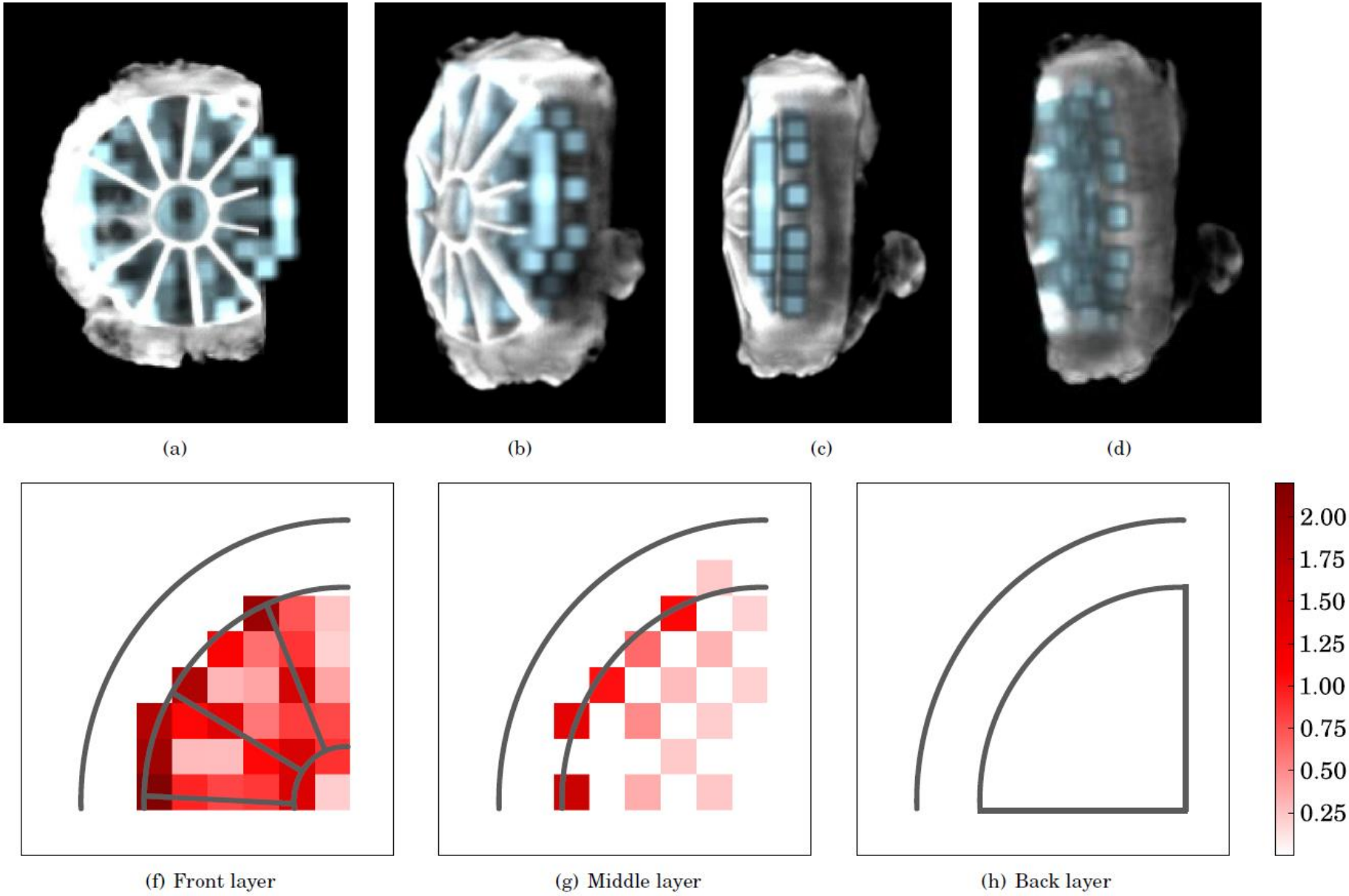
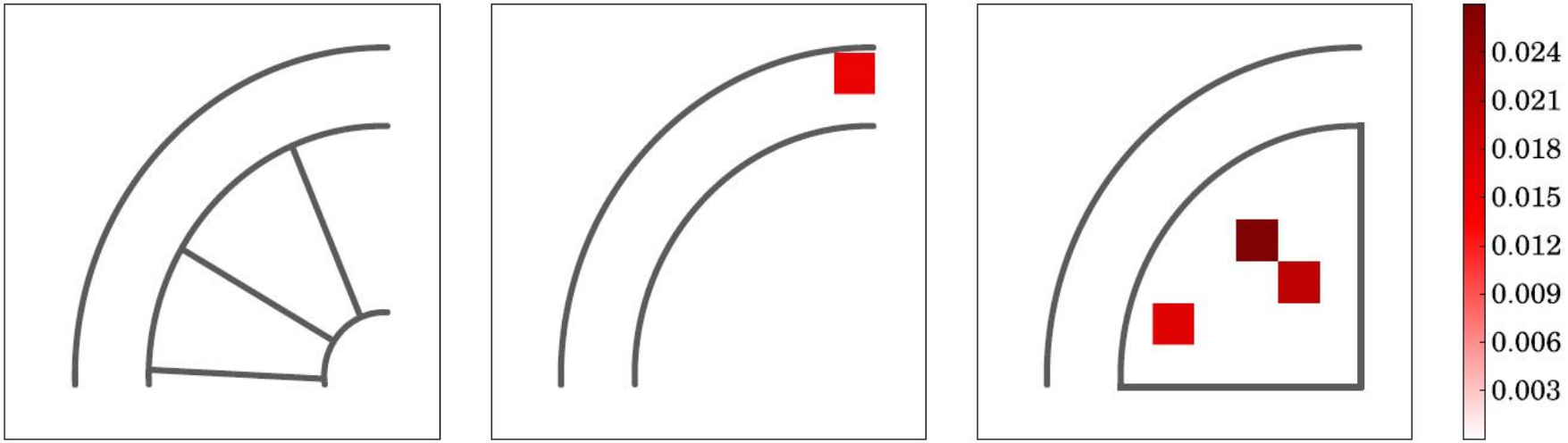
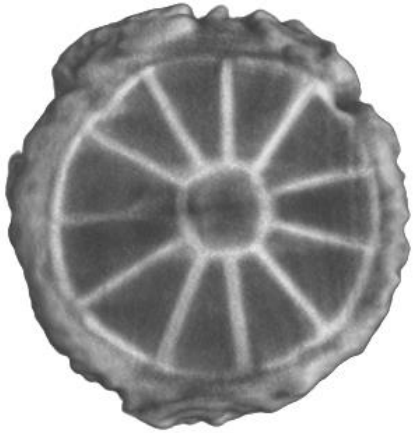


Figure S.11.: Chlorine distribution



(e) Front layer (f) Middle layer (g) Back layer

Animation S.1: 3D visualization of the fibula based on the NT reconstruction



Animation S.2: 3D visualization of the metallic frame of the fibula based on the segmentation of the NT reconstruction. The filling material, the pearl and the almandine inlays were made transparent.