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Error reduction in simultaneous proton induced X-ray emission tomography and on/off-axis scanning transmission ion microscopy-tomography

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Rapid micron-resolution quantitative elemental mapping is possible at the University of Surrey using a combination of proton induced X-ray emission tomography (PIXE-T) and simultaneous on/off-axis scanning transmission ion microscopy-tomography (STIM-T). A preliminary analysis of hair was performed. However, experimental uncertainties lead to large errors in tomograms and this work focuses on identifying and reducing the sources of error in both tomographic and 2D mapping. The STIM-T counts per pixel are used to normalise the PIXE-T data for charge. However, the geometry of the collimator and the scattering foil affects the detection rate since the loss of protons in the collimator increases as energy loss increases due to scattering. Errors in the PIXE geometric efficiency are greater in mapping when the detector is close to the sample. Moreover when a 'funny' filter was used for PIXE-T the uncertainty in the efficiency was found to increase because the sample-filter distance changes during the experiment.

Introduction

The University of Surrey Ion Beam Centre has a 2 MV Tandem accelerator and beamlines including a proton microprobe with potential sub-micron beam-spot size of sufficient current for PIXE analysis. Simultaneous proton induced X-ray emission tomography (PIXE-T), on/off-axis scanning transmission ion microscopy-tomography (STIM-T) and Rutherford backscattering spectrometry (RBS) has been performed.[1] STIM-T, by measuring the proton energy loss, gives the mass density while PIXE-T gives the elemental masses. Tomography benefits analysis by giving more than just depth-averaged measurements and, hence, is more accurate for inhomogeneous samples. Delicate samples can be analyzed without sectioning and exposure to contamination. Correlations of elements are also more meaningful as the volume of correlation is significantly reduced (unless very thin samples are used). A preliminary analysis of hair was performed.

There are several sources of errors arising during the tomographic analysis. The errors arising from tomographic reconstruction is determined by for example the reconstruction method, number of counts per pixel and projections. Beam damage results in uncertainty in the original, undamaged state of the hair – this was corrected for by pre-damaging the hair in order to limit morphological changes during the PIXE-T experiment. The change in matrix composition and mass loss was measured. This paper focuses on sources of experimental error arising from uncertainties in the charge measurement and X-ray detection efficiency.

To reduce the time required to perform PIXE-T, STIM-T is performed simultaneously by using the on/off-axis configuration. On-axis protons (taking a straight path through the sample) are scattered through a thin gold foil and detected by an offset photodiode detector. This also allows PIXE-T and STIM-T data sets to be mapped accurately, thus density correlates with elemental masses.

As well as using STIM to determine median energy loss per pixel, STIM counts per pixel (proton count maps) were used to normalise PIXE counts per pixel as only the charge per projection could be determined directly. The number of protons per pixel measured is proportional to the charge per pixel, taking into account the deadtime for the STIM and PIXE data are very similar.[1]

Figure 1 shows a schematic of the experimental setup at the University of Surrey. The Si(Li) detector is placed at 90° to the beam, therefore, able to be placed much closer to the sample, 11 mm as opposed to 25 mm, than in the standard 315° position that the detector is placed at during 2D analysis at the ion beam centre. However, this results in higher bremsstrahlung but with a much increased efficiency for trace elements such as Zn and Fe that have little or no background radiation.

These two methods, however, generate sources of error during the experiment. The number of protons detected by the photodiode varied as lateral scattering increased due to the collimator. The charge per pixel is increasingly underestimated (as measured by the Faraday cup) as the energy loss increases hence the concentration is overestimated in central parts of the tomogram.

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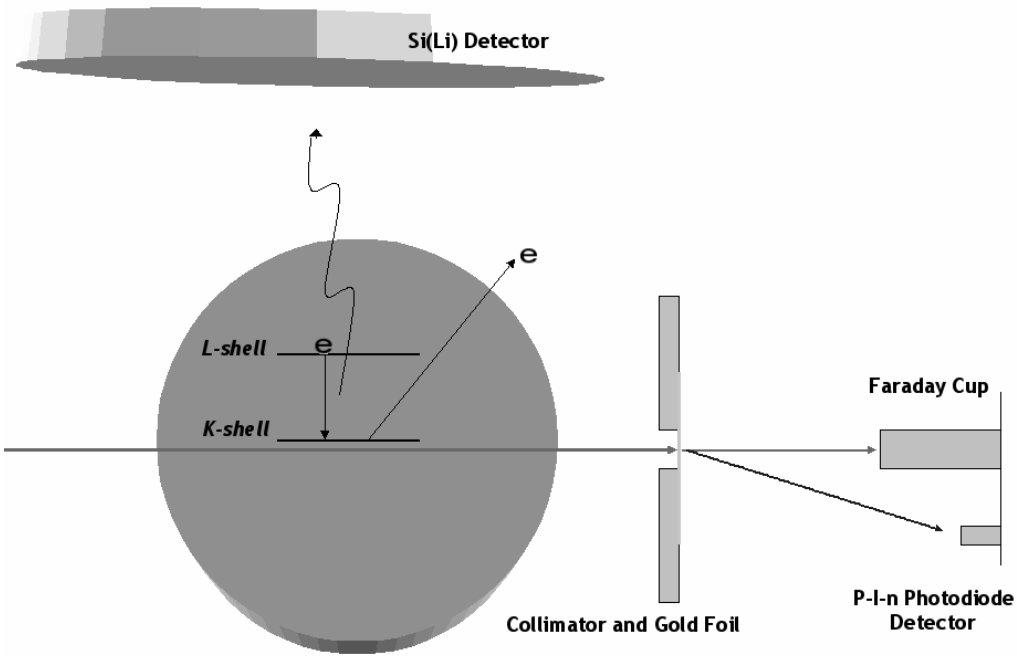

Fig. 1. Schematic illustration of the experimental setup at the University of Surrey

By placing the Si(Li) detector close to the sample the width of the scanning area is a greater proportion of the width of the sample-detector distance. Hence, the geometric efficiency differs from one side of the sample to the other. During the experiment a 'funny' filter was used. The filter contains holes on a 1 mm thick polyethylene disc. This is a similar problem to the collimation of the protons – the geometry changes during the experiment as the position of the holes and 'view' of the holes changes during the experiment. The problem of funny filters has been highlighted before[2] and unless the filter is placed fully on the detector, the sample-filter and filter-crystal distances and geometry are not fully known.

Experimental

A strand of untreated hair 51.6 μm long and approximately 100 μm across at its thickest point was analysed with a 3 MeV proton beam with a 4.3 μm by 4.3 μm beam spot size. This gave 12 reconstructed cross-sectional slices of hair. DISRA (Discrete Image Space Reconstruction Algorithm),[3] an iterative software package, was used to reconstruct the data.

Proton collimation corrections

The gold leaf on the far-side of the collimator is approximately 150 nm thick. This was measured using RBS and STIM. It was found to contain approximately 2% Cu which was expected as this helps the process of making gold leaf.

The length of the collimator was approximately 1 mm and the width was 1 mm. The foil-end of the collimator was placed approximately 11 mm from the sample (sample–foil distance). This gives a half-acceptance angle of 2.9°. This ensures that the protons that pass through the collimator have passed through the sample ideal path and not scattered and passing through surrounding voxels. However, the number of protons being lost through the collimator varies as a function of energy loss and scattering properties of the sample. Added to this, as the sample did not rotate around the centre of mass, the sample-collimator distance varied. In order to normalize the PIXE charge, corrections and uncertainty in STIM counts has to be determined.

The simulation code SRIM[4] was used to model the collimation, varying the sample-collimator distance and energy, and to correct the proton count maps. Figure 2 shows the loss of protons through the collimator.

The charge maps were normalized and the total charge is also increased due to the loss of protons. This is used to normalise the PIXE maps. The energy loss can be seen to increase the collimation. The maximum energy loss of the real experiment is 1700 keV, the decrease in counts is approximately 20%. The counts were corrected and reconstruction performed. Assuming an error of 0.5 mm, the error is 2% for the maximum energy loss.

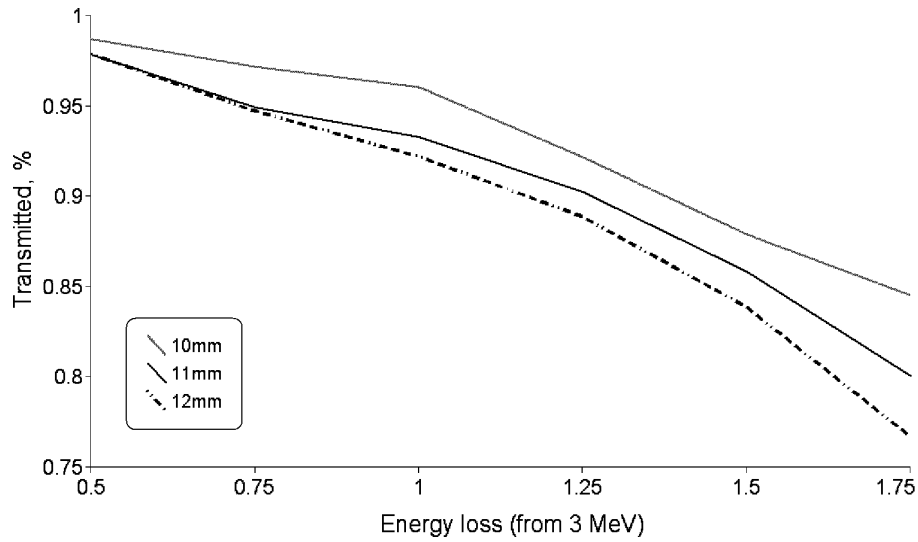

Fig. 2. Percent of protons transmitted through the collimator with varying 'sample–scattering foil' distances

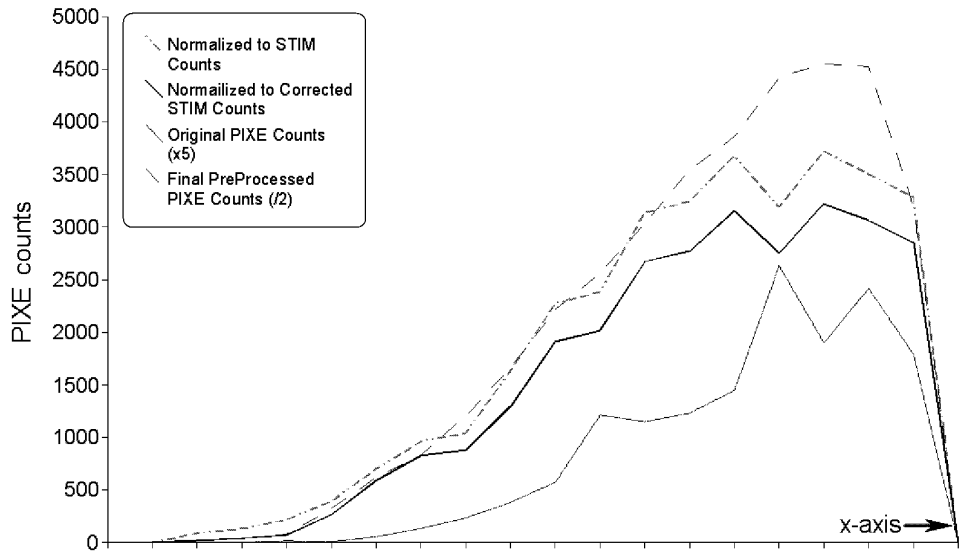

Fig. 3. Normalization of PIXE counts in a projection of hair to the number of STIM counts. Width of projection shown ~80 μm

The uncorrected profile of STIM counts clearly demonstrates the loss of protons – more protons are detected on either side of the hair. Figure 3 shows a profile of total PIXE counts across the first projection normalised to STIM counts and normalised to charge (corrected STIM counts). Spikes are still visible, however, further processing is performed. The final processed profile has been smoothed as a result of missing data – holes in the data acquisition. This is discussed later. The concentration of elements detected using PIXE is reduced in the centre of the sample as the estimated charge is increased in thicker parts of the sample.

PIXE efficiency errors

During the experiment the sample rotated around an area approximately 250 μm across. Therefore, the sample–Si(Li) detector distance varied from 11.1 to 10.9 mm. The X-ray efficiency, therefore, varied from 4.48% to 4.63%.

A funny filter was used. Unfortunately, a detailed measurement of the holes was not taken. By taking measurements with and without the filter is the easiest method however as the distance varies the change in efficiency would be measurable. During the experiment the effective hole size changed. However, this was not performed and only an approximation of efficiency was

given by the makers. The ratio of L and M gold peaks was used to determine a ratio of the efficiencies.

It is, therefore, recommended to not use a funny filter for the errors it introduces. It was planned to use two separate detectors, one on either side, and create a composite sinogram, each corrected for the different efficiencies, and for the simulation to take into account the two positions of the detectors. This would allow both high and low energies to be detected by using different filters and increase the geometric efficiency considerably.

Other experimental sources of error

RBS efficiency is also affected by the change in solid angle, however, less so due to being 55 mm from the sample.

The system dead-time was very high during the experiment, approximately 13%. Moreover, this was not evenly spread. Spots in the maps are visible and appear to be related to the computer being occupied either updating graphics or saving list-mode file data. When the mouse was moved no data was collected at all. Hence, for accurate mapping, 2D or 3D mapping, OMDAQ cannot be recommended. Detector dead-time was reduced by increasing the pulse-width at the expense of resolution, however, from previous analyse the elements that were reconstructed did not have any significant overlapping problems. S, for example, was extremely large relative to any neighbouring peaks.

Scattered protons hit the collimator and gold foil and the resultant-rays were detected by the Si(Li) detector, hence, Al, Au and Cu could not be reconstructed. This was known from previous testing and the collimator holder was adjusted in the hope to move it back to be in the shadow of the detector but it was not sufficient. As has been suggested, the Si(Li) detector could have been collimated. However, some samples were knocked out of place by the cramped space in the chamber.

Discussion

The sample–collimator distance plays a significant role in the error in charge measurement. Greater than 11 mm and the loss of protons increases and the uncertainty grows accordingly. Assuming a distance of 11 mm with an error of 0.5 mm the error is approximately 2% at the maximum energy loss.

The new correction method has also resulted in better resolution images as spikes are observed and not removed sufficiently when the collimation is not corrected for.

The correction of the charge normalization taking into account the collimation leads to a small decrease of the elemental masses from the centre to the outer part of the tomogram as expected – the energy loss, therefore, charge normalization, is greater when passing through the centre of the hair, hence, the concentration. The concentration is slightly lower, especially in the central parts, as the real charge is higher than the measured charge.

The normalization brings errors when the charge in a pixel is very low, therefore, leading to massive uncertainty in the PIXE counts which can lead to very high PIXE counts. Smoothing and filtering for spikes is performed after charge normalization to identify and remove blank and very high values.

MICHELET et al.[5] suggest that SRIM underestimates the scattering. Maybe the interface between the sample and vacuum creates further scattering. Collimation can be a reliable method for measuring lateral straggling by varying the sample–collimator distance and measuring the change in count rate.

It was planned to use two PIXE detectors to measure both high and low energy X-rays, one with a thin filter, the other with a thicker filter but major modifications to the microscope port would have been required.

The sample needs to rotate around the centre for several reasons – to reduce artefacts from tomographic reconstruction; to minimize analysis time by reducing the scanning area; to minimize errors in the efficiency of X-ray detection and of STIM counts. The sample was adjusted prior to placing in the chamber but was knocked when placing in the chamber. This happened several times but as there was little time, it was decided to perform PIXE on the hair even though it did not rotate around the centre of the shaft of hair.

Conclusions

Great care is required when performing simultaneous PIXE-T and STIM-T. The geometry of the apparatus needs to be known exactly and the sample should rotate around a small area as possible. This does, however, ensure the 3D mapping is possible in a feasible timed-scale. For example, the ion beam could not run overnight, therefore, the experiment had to be completed during one day. For certain inhomogeneous samples, this is an invaluable method for time-saving yet accurate analysis.

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