



Separation of detector non-linearity issues and multiple ionization satellites in alpha-particle PIXE



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ABSTRACT

Multiple ionization satellites are prominent features in X-ray spectra induced by MeV energy alpha particles. It follows that the accuracy of PIXE analysis using alpha particles can be improved if these features are explicitly incorporated in the peak model description when fitting the spectra with GUPIX or other codes for least-squares fitting PIXE spectra and extracting element concentrations. A method for this incorporation is described and is tested using spectra recorded on Mars by the Curiosity rover's alpha particle X-ray spectrometer. These spectra are induced by both PIXE and X-ray fluorescence, resulting in a spectral energy range from ~1 to ~25 keV. This range is valuable in determining the energy-channel calibration, which departs from linearity at low X-ray energies. It makes it possible to separate the effects of the satellites from an instrumental non-linearity component. The quality of least-squares spectrum fits is significantly improved, raising the level of confidence in analytical results from alpha-induced PIXE.

1. Introduction

In a recent paper [1] in this journal we discussed in detail the impact of two issues on the spectra recorded by the alpha particle X-ray spectrometer (APXS) on board the Mars Science Laboratory's Curiosity rover. The APXS, whose purpose is elemental analysis of rocks and regolith material, uses an annular array of six ²⁴⁴Cm radionuclide sources to provide a combination of particle induced X-ray emission (PIXE) and X-ray fluorescence (XRF). A silicon drift detector (SDD), protected by a zirconium collimator, records the resulting X-ray spectra emitted by the sample. The SDD is located a few mm behind the centre of the source array and the sample is typically ~20 mm in front of the sources, although this distance must be increased for safety when sample surfaces depart from smoothness. The unique combination of PIXE and XRF provides a smoothly varying sensitivity across the elements sodium to yttrium [2].

We fitted spectra from both the flight model and its terrestrial calibration twin using our GUAPX code [3], which is a derivative of the widely used GUPIX code [4]. An example spectrum is in Fig. 1. Both codes combine non-linear least-squares fitting with a fundamental parameters approach to matrix effects. In our earlier spectrum fits, we observed residue excursions in certain energy regions, which suggested

non-linear behaviour of the detector or the pulse-processing chain. Further examination revealed apparent shifts in energy of the K X-rays of the lightest observed elements; the magnitude of these apparent shifts increased with decreasing X-ray energy. These were not surprising observations since in PIXE work with 3 MeV protons [5] we had already observed and quantified small non-linear effects in the energy-channel calibration at low X-ray energies (< 5 keV) in silicon drift detector systems.

A second contributor to the observed energy shifts in alpha particle PIXE is the presence of multiple ionization satellites (MIS). These satellites make almost negligible contribution in proton PIXE but a review of the literature [1] shows that in the alpha particle case some of the MIS intensities are similar to or greater than those of diagram lines. In our previous work [1] we were not equipped to explicitly separate the MIS issue from the detector system non-linearity. Nevertheless, we were able to effect an empirical removal of the MIS effect from the overall energy shifts. Despite its approximate nature, this exercise left us with a dependence of energy shift upon true energy that was close to linear but not exactly so.

The effects which we discussed in Ref. [1] in the context of the MSL APXS are pertinent also to the accuracy of accelerator-based PIXE using alpha particle beams. We have therefore developed a means of

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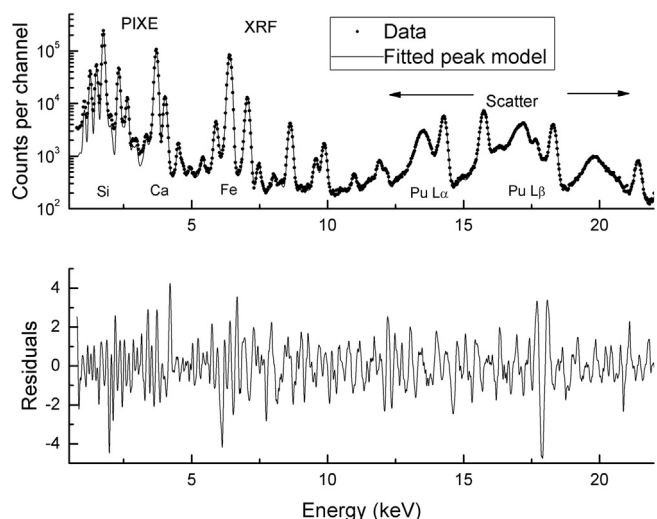


Fig. 1. Martian sample spectrum recorded by the Curiosity rover APXS and fitted with the GUAPX code.

incorporating alpha-induced MIS into GUPIX and GUAPX, and this is described below. To test our methodology it is desirable, as we have demonstrated in Ref. [1], to have X-ray spectra with an extensive energy region above ~ 5 keV, well-populated by diagram lines of sufficient intensity, so that the parameters of the linear portion of the channel-energy relation can be accurately determined as a foundation for examining any non-linear region at lower X-ray energies. With conventional terrestrial PIXE the rapid decrease of ionization cross-section with target atomic number makes this difficult to accomplish. In contrast, the APXS makes it easy to accomplish since the PIXE excitation is accompanied by an XRF component as described above. The Curiosity rover APXS spectra are well populated from 1 to over 20 keV. Moreover, they have excellent energy resolution due to the low night-time temperatures on Mars. By employing these spectra we are able to test both our methodology for incorporating MIS into alpha-PIXE and the quality of the existing MIS database.

2. Choice of spectra

Our previous work was based on spectra recorded with two APXS units. The first of these is the so-called proto-flight model (PFM) and the six spectra used were recorded on Mars, where low temperatures at night-time provide excellent energy resolution (typically ~ 140 – 145 eV FWHM at 5.9 keV). An example spectrum is shown above in Fig. 1. The second was the laboratory twin or flight-equivalent unit, which had somewhat poorer resolution. For the present work we use only PFM spectra, expanding the number of spectra studied to fourteen.

Our earlier work [1] on PFM spectra suggested that the linear region starts at 5–6 keV, and we chose to terminate it at 19 keV. The linear region always contains prominently the characteristic K X-ray peaks of iron (from the sample) and zirconium (fluoresced in the detector collimator), and the Rayleigh-scattered $L\alpha$ and $L\beta$ X-ray peaks of plutonium (scattered from the sample). The iron $K\beta$ energy is subject to some uncertainty from the intense $K\beta'$ satellite [6], hence we do not use it. This leaves us with energies 6.405 keV (Fe KL_3), 14.279 (Pu L_3M_5), 15.775 (Zr KL_3) and 18.294 (Pu L_2M_4). The gap between 6.4 and 14.3 keV is significant, and a further X-ray line around 10 keV is desirable to stabilize the energy-channel calibration.

Fortunately, five APXS spectra recorded by the Curiosity rover between sol 900 and sol 1000 contain germanium (KL_3 line at 9.886 keV) at concentrations of a few hundred ppm wt. These spectra also have zinc (KL_3 at 8.57 keV) lines at useful intensities due to concentrations averaging 1500 ppm wt. A second subset of two spectra had negligible germanium counts but had significant trace zinc concentrations

(4000–6000 ppm wt) All seven spectra were of long duration but one of the germanium cases was recorded at an unusually large stand-off for reasons of instrument safety. Its integrated silicon $K\alpha_1$ intensity was only one third of the average of the other six spectra, and so it was not used. Counting statistics in this reduced group of six, referred to below as Group 1, were excellent. For these cases, sensor head temperatures during data accumulation were between -38 and -48 °C, with a mean of -43 °C.

A second group of three spectra (Group 2) was selected where germanium content was negligible and zinc concentrations were ~ 500 ppm wt. The mean temperature of the sensor head was -44 °C. The objective was to determine if the lack of germanium peaks coupled with much less intense zinc peaks caused any changes in results.

A third set of four spectra (Group 3) was recorded at much lower temperatures, in the range -65 to -73 °C in order to investigate any temperature-dependence in the results. They contained zinc at a mean concentration of 400 ppm wt. and negligible germanium.

3. Determination of apparent energy shifts due to non-linearity

Our in-house code GUFIT is the precursor of GUPIX and is oriented towards accurate non-linear least-squares fitting of energy-dispersed X-ray spectra rather than towards the extraction of concentration information. It was therefore used to determine the apparent energy shifts. We had observed in the prior work [1] that non-linearity occurred below 5 keV in our spectra. GUFIT was therefore instructed to utilize only those X-ray lines above 5 keV in energy (E) for determination of the two parameters of the usually adopted linear energy calibration

$$C = A_1 + A_2 E \quad (1)$$

where C is channel number. The centroids of lines having lower energy were not restricted by Eq. (1). Instead, GUFIT was directed to vary the centroid values to achieve the best fit. The resulting upwards shifts of the centroids relative to the positions expected from the linear calibration do not imply actual changes of photon energies. What is implied is non-linear behaviour, giving rise to “apparent energy shifts”.

In Ref. [1] this variable energy fitting was done using only a single Gaussian function to approximate the full line description (average of all $K\alpha_1$, $K\alpha_2$, radiative Auger and KL^1 and KL^2 MIS lines). This, for example, would produce an initial placement of the Si K X-ray peak at 1.746 keV instead of its usual $K\alpha$ position of 1.740 keV which permitted a crude estimate of the observed energy shift after MIS incorporation. However, the absence of a complete line description in this approach limited the quality achievable in the final iteration of the fit.

To improve upon our previous work, all known diagram and satellite lines had to be included as individual components within each elemental peak description. The basis was the original GUPIX library description of the $K\alpha$ and $K\beta$ groups, including radiative Auger lines, to which we added first (KL_1) and second (KL_2) order satellites. The energy shift and intensity of the MIS relative to the diagram $K\alpha$ and $K\beta$ lines were derived from existing literature in Ref. [1] for the atomic number range $11 \leq Z \leq 32$. In principle, the first and second order satellites could be computed for each of the $K\alpha_1$, $K\alpha_2$, $K\beta_1$, $K\beta_3$ and KLV and KMM radiative Auger lines, but this would produce a large and unwieldy description. A simplified procedure was therefore adopted whereby KL_1 and KL_2 groups were calculated using a single $K\alpha$ and single $K\beta$ value. The energies of these were calculated using an intensity-weighted average of the individual diagram line energies (eg. for $K\alpha$, we used the weighted average of $K\alpha_1$, $K\alpha_2$ and KLL and KLM radiative Auger lines). Similarly, the total intensities of the $K\alpha$ and $K\beta$ groups were derived by summing the intensities of their individual lines.

In this manner, only a total of four MIS lines needed to be added to the base GUPIX line description for each element. The MIS relative intensities (KL^1/KL^0 and KL^2/KL^0) used on the $K\beta$ lines were the same

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