USING A CHARGE-COUPLED DEVICE (CCD) TO SIMULTANEOUSLY GATHER X-RAY FLUORESCENCE (XRF) AND X-RAY DIFFRACTION (XRD) INFORMATION

S. Cornaby[a], T. D. Grow[a] A. Reyes-Mena[a], P. W. Moody[a], A. Stradling[a], T. Hughes[a], and L V Knight[b]

[a]MOXTEK, Inc., 452 West 1260 North, Orem, UT 84057, USA
[b]Department of Physics and Astronomy, Brigham Young University, Provo, UT 84602, USA

ABSTRACT

A breadboard setup constructed at MOXTEK, Inc., is capable of capturing both x-ray diffraction (XRD) and x-ray fluorescence (XRF) information simultaneously using a charge-coupled device (CCD) as the x-ray detector. This preliminary setup will lead to a prototype simultaneous XRD/XRF instrument. NASA is funding the project because it could be used for future Mars missions for analysis of rocks. The instrument uses a CCD to capture both the energy and the position of an incoming x-ray. This is possible because each pixel acts as a spatially addressable energy-dispersive detector. A powdered sample of material is placed in front of the CCD, which in turn is bombarded by a collimated x-ray beam. The instrument's critical features, the x-ray source, collimation optics and x-ray transparent windows need to be optimized in the size and power to allow the instrument to be portable. In this paper the instrument's design parameters as well as the properties of both the CCD as x-ray detector and the low-power consumption tube are investigated.

Keywords: x-ray diffraction (XRD), x-ray fluorescence (XRF), charged-coupled device (CCD), compact instrument, powdered diffraction, Mars, back-side illuminated CCD, front-side illuminated CCD, energy-dispersive detector

1. INTRODUCTION

The first x-ray diffraction (XRD)/x-ray fluorescent (XRF) system was developed at Ames National Laboratory and is currently in use at Los Alamos National Laboratory [1,2]. The intended applications for this instrument are for planetary exploration and as a portable instrument for terrestrial use. NASA is funding this project because of its possible use in future Mars missions. These missions have previously only used methods that give elemental composition, which are subject to sizable uncertainties for mineral identification. XRF gives the elemental composition and suggests certain minerals, but the traditional method of definitive mineral identification is by XRD [3]. With a small, portable XRD/XRF instrument, both the diffraction and the fluorescence information can be gathered on Mars as well as on Earth.

The purpose of the breadboard setup constructed at MOXTEK is to incorporate and test the key components used in an XRD/XRF instrument design. **Figure 1** shows an instrument concept drawing while **Figure 2** shows a picture of the actual instrument. This design allows samples to be placed on the outside of the camera so that they can be easily retrieved and exchanged. The information gained from this breadboard will lead to a prototype instrument, which will be small and portable when the final design is completed. All the components and their functions will be described, and the advantages and limitations will be discussed.

One key component of the instrument is the CCD detector. It can use each pixel as an addressable individual energy dispersive detector, so the spatial position and the energy of an x-ray event can be simultaneously recorded. Once all the events are recorded, the diffraction information and the fluorescent information can be extracted with the use of sorting algorithms. We have investigated some of the properties of both a front-side illuminated (FSI) and a back-side illuminated (BSI) CCD, such as energy resolution and detection limits. In particular, the number of single x-ray events detected by the CCD was studied. This is a critical feature of using the CCD as an energy dispersive detector. The other key component of the instrument is the low-power x-ray source. X-ray diffraction is typically done using a high-power x-ray source such as a rotating-anode source. The reason that these high-powered sources are used is to get large amounts of flux. For a portable instrument we cannot supply

X-Ray and Gamma-Ray Instrumentation for Astronomy XII, Kathryn A. Flanagan, Oswald H. W. Siegmund, Editors, Proceedings of SPIE Vol. 4497 (2002)
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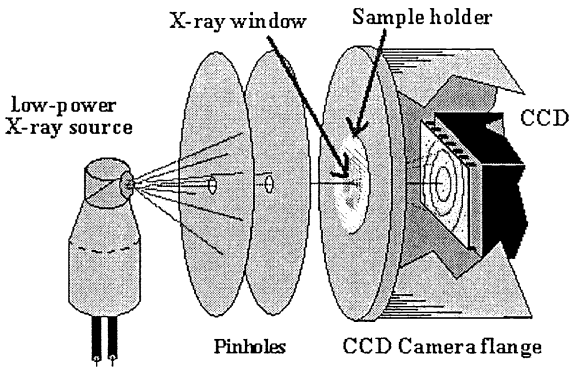


Figure 1. Schematic drawing of the XRF/XRD instrument concept

the power needed for need for a high-powered tube that runs at 900 Watts. We need sufficient flux on the sample with under 10 Watts of power.

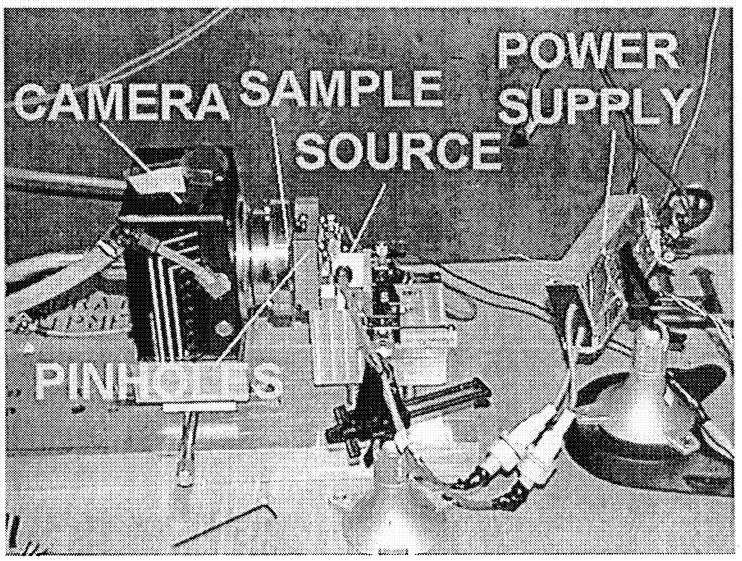


Figure 2. The actual breadboard layout

2. CCD CAPABILITIES AND RESULTS

Along with others [6,7] we have investigated the basic characteristics of both FSI and BSI CCDs in the X-ray region. The CCDs we used in comparison are FSI CCD06-02 made by EEV and BSI a Si502 made by SiTe. Within the CCD industry there is an effort to optimize the sensitivity, resolution, timing, and readout speed of a CCD. Finding the right CCD for the XRD/XRF instrument is the key to making a useful and fast instrument. The construction of FSI CCD is such that radiation must pass through the gate structure in order to get into the depletion region where the radiation is detected. The gate structures are in front of the depletion region because of the fabrication process, and for customary regions of detection the gate

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structures are largely transparent [7]. The BSI CCD was developed in an effort to increase the range and sensitivity of CCDs. This device has the supporting silicon wafer etched away until the depletion region is reached. The radiation is exposed to the backside where there is no interference from the gate structure. This increases the sensitivity and range of the CCD because the radiation does not have to pass through any other structures [7]. The next four sections give the properties of the two types of CCDs. In comparing the properties of the two types of CCDs, we have found that the FSI CCD is best suited for use in a simultaneous XRD/XRF instrument.

2.1. Energy Resolution

The energy resolution of both CCDs were explored experimentally and researched in the literature. The major factors that control the resolution of both of the CCDs are temperature and exposure time. Decreasing the temperature increases the energy resolution. Also, decreasing the exposure time increases the energy resolution. Table 1 contains the energy resolution for the FSI device at various energies at a temperature of -50° C for a 5- to 8-second exposure time. The present energy resolution is 185 eV at 5.89 keV. With the present resolution allows discrimination between the K_α and K_β peaks above 3.3 keV (potassium) in the energy spectra.

Energy (keV)	Resolution (FWHM) eV at -50° C
1.74 (Si K alpha)	200-220
2.62 (Cl K alpha)	220-240
3.69 (Ca K alpha)	220-240
6.40 (Fe K alpha)	280-300
8.01 (Cu K alpha)	300-320

Table 1. The energy resolution of the FSI CCD.

Energy resolution decreases at higher temperatures in the CCD due to thermal noise. This noise is measured as the number of electron-hole pairs created per second or counts per second (cps). For each decrease of 6° to 7° C in the temperature of the CCD, the thermal noise is halved. The counts per second bottom out at about -100° C, at which the counts on individual pixels are about one electron-count per hour.

Within the present limits of the breadboard instrument, a longer exposure time will increase the dark current noise on the CCD and cause a decrease in energy resolution. The limit of cooling that we can now achieve is -50° C. Decreasing the exposure time to increase resolution is not the best solution. There are other problems to be considered when decreasing the exposure time. In order to collect the data we need to make 300 to 500 exposures at about 5 to 8 seconds per exposure [4,5]. Also, it takes 2 to 3 seconds to read out the CCD between exposures. If we were to shorten the exposure time and increase the number of exposures, we would increase the data-capturing time. We have stayed with 5 to 8 second exposures to keep the x-ray counts high on each individual exposure. The exposure time needs to be set to optimize the data collection rate [4,5]. The best way to increase the energy resolution is to cool down the CCD to a lower temperature.

We have compared the resolution of the FSI and the BSI CCDs around -50° C; the resolution is roughly equal. In order to get this energy resolution a background exposure is subtracted from a foreground exposure pixel by pixel. We have been able to increase the energy resolution of the detector by taking five to ten background exposures and averaging the background for each pixel in the CCD. When we used the average of five backgrounds we were able to increase the resolution on the CCD increase the resolution at Fe55 emission at 5.89 eV from 225 eV to 185 eV on the FSI CCD ran at -50° C.

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2.2. X-Ray Detection Range

The FSI CCD has an x-ray energy detection range from 1.7 keV to 12 keV. **Figure 3** shows a spectrum collected from a KCl sample on the XRD/XRF instrument with the FSI CCD. The detection range on the FSI ranges from the silicon K_α peak at 1.7 keV to the lead L_α line at 10.55 keV. The FSI CCD has a dead layer, consisting of the gate structure, on the surface that renders poor quantum efficiency at lower energies. In general CCDs are not able to detect energies too far above 11 keV because the active volume of the CCD is not thick enough to absorb all of the harder x rays' energy. Most of the samples taken are excited by 8.01 keV (Cu K_α) and 8.91 keV (Cu K_β) x rays and, consequently, the energies detected by the CCD are below these energies.

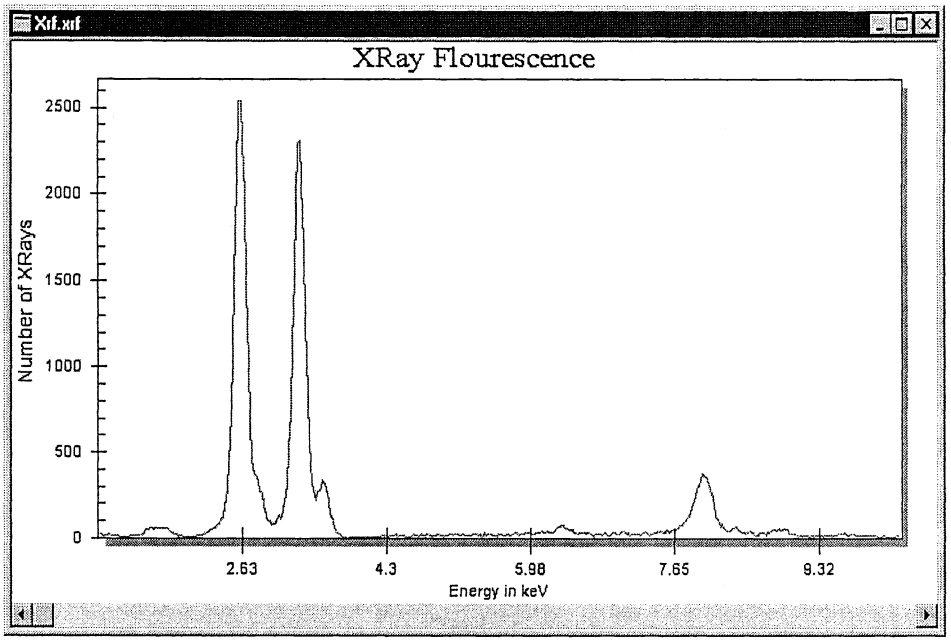


Figure 3. A spectrum collected from a KCl sample on the XRD/XRF instrument with the FSI CCD.

Different types of CCDs have different detection limits. BSI CCDs are able to detect softer x rays. The increased detection limit is the main advantage that BSI CCDs have over FSI CCDs. The BSI has been able to detect fluorescence from less then 500 eV for previous applications in soft x-ray detection in a vacuum inverment.

2.3. Event Types

The major point of comparing FSI and BSI CCDs for applications to a simultaneous XRD/XRF instrument was to find the frequency of single events. For the instrument to capture useful XRF information the energy from one x ray needs to be captured into one pixel. This type of event is the so-called "single event". As long as one x-ray event is captured in one pixel, then the CCD can be used as an energy-dispersive detector. For the CCD to be used in the single-count mode each of its pixels needs to capture either one event or no events per exposure. If the energy from a single photon is divided into two or more pixels we get a so-called "split event". Depending on the type of CCD, these events cannot be used, so they are discarded from the data set with the use of an algorithm. We can reconstruct the energy information of split events with the FSI CCD but we cannot reconstruct split events from the BSI data. Others authors [6] have extensively studied split events in FSI and BSI CCDs with similar results. The team that worked on the Chandra X-ray Telescope has done extensive research on pixel splitting in CCDs. They modeled the interaction of x rays with a FSI CCD, a BSI CCD, and an epitaxial FSI CCD using Monte Carlo simulations. Both experiments and models concluded that a FSI CCD has fewer split events than a BSI CCD.

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When event comparisons were made, a Fe55 source was detected directly. We took equal exposure times with each camera. When the images were viewed it was clear that the BSI CCD had far more split events than the FSI CCD. Two-thirds of the events recorded on the FSI CCD were desired single events while the BSI CCD had about one-third of the total events being single events. At our present collection rate using the BSI CCD would almost triple the time required to collect data from a sample.

The reason the BSI CCD has a higher rate of split events than the FSI CCD is based on their contrasting structures. The radiation on the FSI CCD is deposited on the same side of the depletion region as the gate structures are located. This causes the pixel boundaries to be well defined because the creation of electron hole pairs occurs close to the surface of the depletion region. For the BSI CCD the radiation is deposited on the other side of the depletion region where the pixel boundaries are not as well defined since the gate structure is on the other side of this region [6,7]. It is this structural difference that causes the BSI CCD to have a higher probability for split events.

When using the CCD to gather energy information, the CCD needs to detect only one event per pixel, or the energy information will be lost. To identify a valid single x-ray event, the eight adjacent pixels need to be void of any events. As the CCD is exposed to more x-rays the number of single events will eventually decrease. This is because the x-ray events will have a higher probability of occurring in adjacent pixel or in the same pixel, thus losing the energy information of the x ray. We have determent that the best collection rate for our FSI CCD to be 5000 total events per exposure [5]. We have acquired data from ~1000 to 30 events per exposure, so we have not had problems due to over exposing the CCD. **Figure 4** shows the data capture rate with the low-power tube. The data was collected over 600 exposures at 5 seconds per exposure.



. Figure 4. The data capture rate with a low-power tube. The data was collected over 600 exposures at 5 seconds per exposure

2.4. Diffraction and Diffraction Resolution

The diffraction resolution in the breadboard instrument is presently limited by the spot size of the incident beam. **Figures 4 and 5** show the diffraction image and the 2theta plot of a KCl sample. Our spot size when viewed on the CCD ranges from 50 to 220 μm in size based on the pinhole optics used. This gives an angular resolution range of approximately 0.4° to 1.5° in a 2θ diffraction plot depending on the optical setup. The resolution of the diffraction patterns are improved by using smaller pinholes, but smaller pinholes will reduce the flux of x rays impingent upon the sample. We have constructed a model of the

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pinhole setup to optimize the flux for a given resolution on the CCD. This model has agreed with the experimental data and will allow us to set the desired resolution with an optimized flux. Smaller pinholes can, at the most, increase the resolution to ~ ~0.25°, which is the limit of resolution based on the pixel size of the CCD. The present resolution of the CCD will be able to give the crystal d-spacings within ± 0.05 Å.

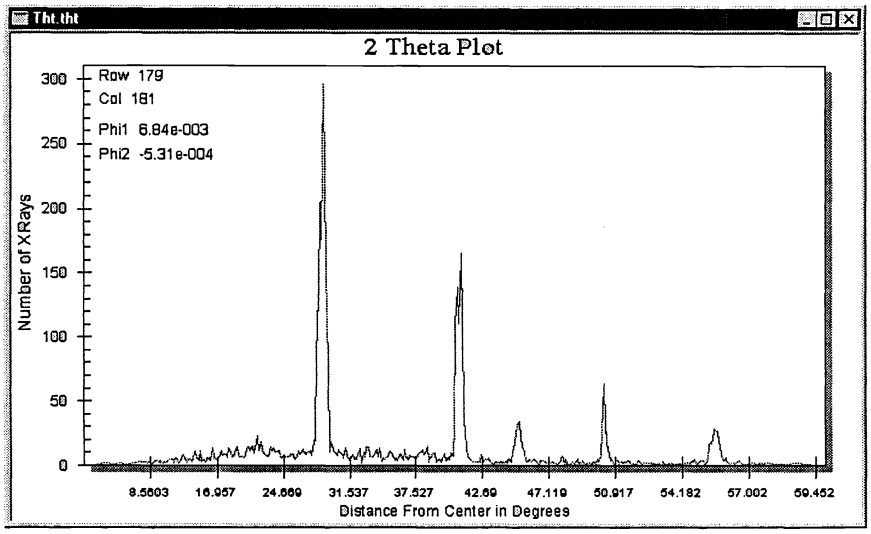


Figure 5. A 2theta plot of a KCl sample

3. Low-Power X-ray Tubes (note: ref. 9 is about the FE tubes)

The low-power tube used to collect data is a small filament tube.

Source	RU-200	Svetlana tube	MOXTEK tube72	MOXTEK tube79	MOXTEK tube79
tube characteristics					
spot size	.1mmx1mm	300 µm	360x600 µm	500x750 µm	500x750 µm
tube current	30 mA	50 µA	50 µA	95 µA	95 µA
tube voltage	30 kV	30 kV	16 kV	17.5 kV	17.5 kV
Optics					
pinhole size near source	300 µm	300 µm	300 µm	75 µm	200 µm
pinhole size near sample	100 µm	200 µm	200 µm	50 µm	100 µm
distance between pinholes	~9 cm	~6 cm	~6 cm	1.38 cm	61 cm
divergence of beam	0.13°	0.24°	0.24°	0.25°	0.14°
CCD angular resolution	~0.6°	~1.3°	~1.3°	~0.5°	~0.7°
Data count rate					
Cl K alpha (2.622 keV)	12.8 cps	1.5 cps	7.6 cps	4.4* cps	7.0* cps
K alpha (3.312 keV)	18.3 cps	2.1 cps	6.4 cps	3.4* cps	6.5* cps
Cu K alpha (8.041 keV) XRD events	4.7 cps	0.33 cps	0.83 cps	0.98* cps	1.2* cps

* These count rates include both single events and reconstructed split events. The split events were not reconstructed for the earlier samples

4. CONCLUSIONS

The breadboard XRD/XRF instrument has been successful. XRD and XRF data have been simultaneously gathered from the instrument. We have been able to investigate the various types of events that may occur on both FSI and BSI CCDs. We have

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found that the FSI CCD is more efficient in capturing single events, and is consequently more suitable for an XRD/XRF instrument. The breadboard's geometrical layout renders a practical device and makes the in-air sample analysis possible. The testing setup has been successful in proving the feasibility of a small compact XRD/XRF instrument that has the potential to be used in future planetary applications and in terrestrial applications. With a small low powered x-ray source we have been able to collected data with minimal power consumption from the tube, which means that soon a prototype will be build that will be portable and able to do both XRD and XRF simultaneously.

ACKNOWLEDGMENTS

NASA is supporting this work under SBIR contract number NAS2-00010. We thank Brigham Young University for student partial-financial support and for the use of equipment.

REFERENCES

[1] Blake D F et al. 1992 *Design of an X-ray diffraction/ X-ray fluorescence instrument for planetary applications* Proceedings of the Lunar and Planetary Science Conf. XXIII 117-118.

[2] Blake D F et al. *X-ray Diffraction Apparatus* 1993 U.S. Patent No. 5,491,738.

[3] Vaniman D, Bish D, Blake D F, Elliott S T, Sarrazin P, Collins S A , Chipera,S 1998 *Landed XRD/XRF analysis of gprime targets in the search for past and present Martian life* J. Geophys. Res. 103(E13) 31,477-31,489.

[4] Cornaby S, Reyes-Mena A, Pew H K, Moody P W, Hughes T, Stradling A, and Knight L V 2001 *AnXRD/XRF Instrument for the Micro Analysis of Rocks and Minerals* Meas. Sci. Techol. **12** 676-683

[5] Cornaby, S 2000 *Using a CCD to gather XRF and XRD Information Simultaneously* Brigham Young University, Dept. of Physics and Astronomy

[6] Ralph P. Kraft, 1995 *Advances in Soft X-ray Spectroscopy Using Charged-Coupled Devices,*Pennsyvania State University, Dept. of Physics and Astronomy

[7] Sweedler J V, Ratzlaff K L, Denton M B, eds 1994 *Charge-Transfer Devices in Spectroscopy*, (New York:VCH Publishers Inc.)

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