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Chemical treatment of CdZnTe radiation detectors using hydrogen bromide and ammonium-based solutions

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ABSTRACT

Surface damages occur in Cadmium zinc telluride (CdZnTe) wafers for radiation detection devices during dicing and polishing. This often results in increased leakage current that limits the performance of the detector. An effective method of removing the surface damage and thus reducing the leakage current is through the use of chemical treatments. The effects discussed in this study include: chemical polishing with a mixture of hydrogen bromide solution followed by passivation with ammonium fluoride in a hydrogen peroxide solution. The effects on the current-voltage measurements and the spectral response were monitored over a 2-week period. X-ray photoelectron spectroscopy (XPS) was also obtained to observe the formation of chemical species on treated surfaces. The resistivity of the treated CdZnTe samples is on the order of 10^{10} ohm-cm. The current in the I-V measurements increased rapidly immediately following the chemical polishing and surface passivation, and decreased steadily afterwards. The spectral response showed that the 59.5-keV peak of Am-241 was stable in the same position over the test period.

Keywords: Cadmium Zinc Telluride, radiation detectors, surface passivation, X-ray photoelectron spectroscopy, current-voltage characterization, spectral response.

1. INTRODUCTION

Cadmium zinc telluride (CdZnTe), a prime material for room-temperature radiation detectors as used in national security and medical imaging applications, is increasingly in demand [1,2,3]. Despite their high value, the detector performance is limited by processing procedures and products. To produce higher quality detectors, surface treatment processes are applied to reduce fabrication-induced defects that need to be systematically removed. One such process is the use of chemical treatments in techniques known as etching and passivation. In etching CdZnTe, researchers have investigated the use of bromine-based solutions that produce comparatively smoother surfaces compared to mechanically polished crystals.

Etching also lessens the problem of trapped charges [4]. This treatment also generated surfaces low in cadmium and rich in tellurium that increased the surface conductivity, and thus, leakage current. This finding stresses the importance of passivation [4,5]. Ammonium-based solutions reportedly are favorable chemicals for surface passivation [6,7]. In this paper we detail the results of the immediate effects and long-time effect of passivation with a mixture of ammonium fluoride in hydrogen peroxide and water. The chemo-mechanical polishing was conducted with a hydrogen bromide solution rather than the conventional bromine-methanol etchant.

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2. EXPERIMENTAL PROCEDURE

2.1 Surface processing and x-ray photoelectron spectroscopy

In this experiment, we used three 6.4 mm x 6.4 mm x 2.8 mm samples sliced from a detector-grade CdZnTe wafer grown by the Bridgman method to remove any apparent surface damage produced during slicing. The samples were polished mechanically, first with 800-grit- and 1200-grit-silicon carbide abrasive- papers fixed to a polishing machine. Subsequently, the crystals were fine polished on a smooth disc with 3-micron and then 0.9-micron alumina powder. The samples next were rinsed thoroughly with distilled water and methanol to remove any powder residues.

The samples were labeled A1, A2, and A3. A1 was reserved as the control, while A2 and A3 were chemo-mechanically polished on a smooth disc with a 48%hydrogen bromide in 30%hydrogen peroxide and ethylene glycol ($HBr + H_2O_2 + C_2H_6O_2$ at a volumetric ratio of 5:1:12). After this, the residual bromine was removed by rinsing them with methanol and distilled water. Finally, sample A3 was passivated by immersion in a mixture of ammonium fluoride, 30% hydrogen peroxide, and water ($NH_4F + H_2O_2 + H_2O$) for 1 minute at a volumetric ratio of 2.86 g: 8 mL: and 17 mL.

Passivation was accomplished at a room temperature of 25 °C. X-ray photoelectron spectroscopy (XPS) experiments then were conducted using a RHK Technology UHV 7500 system in an ultrahigh-vacuum setup at a pressure below 8 x 10^{-10} Pa. XPS measurements were acquired through an Aluminum (Al) X-ray source immediately after each chemical treatment. We scanned for peaks of cadmium (Cd), tellurium (Te), and tellurium oxide (TeO_2), and also analyzed the chemistry of the surface chemistry. XPS measurements were repeated on all the samples 24 hours later.

2.2 Current-Voltage and Spectral Measurements

To measure the leakage current and also record the spectroscopic performance of CdZnTe, the surface-processing steps undertaken before the XPS experiments were repeated on samples A2 and A3, still leaving sample A1 as the control. Thereafter, we deposited a gold chloride ($AuCl_3$) solution at the center of the two planar CdZnTe surfaces using the electroless-pipette method. Adequate care was taken to avoid the $AuCl_3$ solution from spilling over to the sides. After 10 seconds, the excess $AuCl_3$ solution was blotted off, using the tip of a clean wiping cloth, and the samples were dipped in distilled water to remove the excess $AuCl_3$ solution.

The bulk leakage current of samples A1, A2 and A3 were measured by using a customized current-voltage probe in a metal box coupled to a Keithley Picoammeter/Voltage Source. Up to 100 volts was applied to the detectors. We monitored the samples' spectroscopic response over two weeks by mounting the detectors on to a standard eV Product's brass holder, securing the detector against a beryllium window with a gold-plated spring contact. The surface in contact with the beryllium window acted as a cathode. The brass holder was then connected to a multi-channel analyzer (MCA) through a pre-amplifier and a shaping amplifier. The spectral response from a sealed Am-241 gamma ray source was recorded at 600 seconds with 200 V applied, and a shaping time of 2 μs. The data were stored for further analysis.

3. RESULTS AND DISCUSSION

It is important to note that the chemical treatments applied in this study do not improve the detector's performance when carried out on rough surfaces, because rough crystal surfaces create additional trapping centers and increased the leakage current [4, 8],especially for low energy x- and gamma-rays. In this study, the treatments applied to all the samples were analyzed assuming that they have comparable properties, because they were cut from the same CdZnTe wafer. Figures 1 and 2 show the XPS analysis results of the surface chemistry of the chemo-mechanically polished sample (A2) and the freshly passivated samples (A3). Prominent $Cd3d_{3/2}$ and $Cd3d_{5/2}$ peaks were evident at approximately 402 eV and 405 eV, consistent with the elemental peak values for cadmium. The cadmium peaks remained stable with no significant shift before and after the passivation as shown in Figure 1.

Further XPS analysis revealed two peaks in the Te3d region at approximately 573 eV and 584 eV, consistent with the elemental peak for tellurium [9]. The TeO_x peaks at approximately 577 eV and 588 eV were evident on freshly chemo-mechanically polished sample as shown in Figure 2. The tellurium peak was however more prominent on chemo-mechanically polished sample A2 indicating the presence of a tellurium-rich surface. The effect of passivation with ammonium fluoride passivating agent on the hydrogen bromide treated sample can be seen in the shift in the peaks in the Te3d region. The tellurium elemental peak disappeared following passivation while the TeO_2 peak increased.

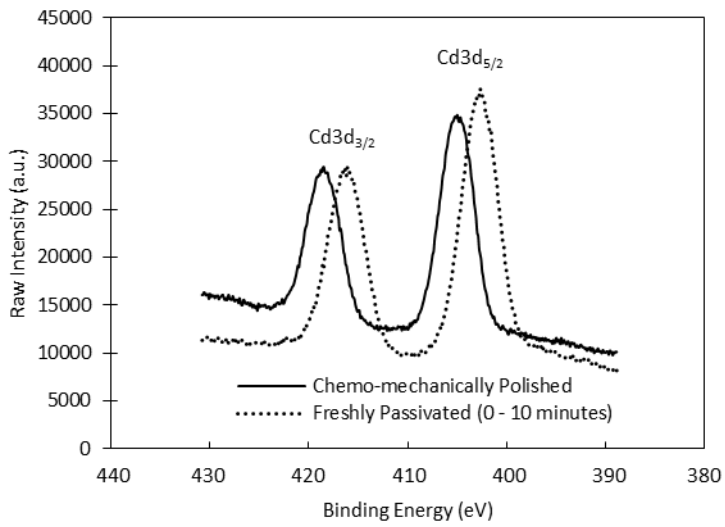

Figure 1. XPS spectra showing $Cd3d_{3/2}$ and $Cd3d_{5/2}$ peaks before and after passivation

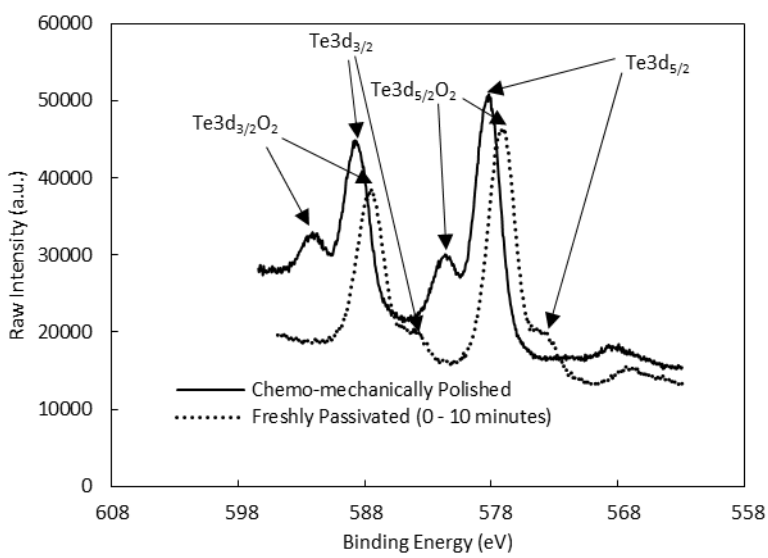

Figure 2. XPS spectra showing the TeO_x peaks before and after passivation

The results of the I-V measurements over a 14-day period are shown in Figure 3. The resistivity of the CZT samples varies between 1.3 x 10^{10} Ω-cm and 7.2 x 10^{10} Ω-cm in-between treatments. Chemo-mechanical polishing (sample A2) and fresh passivation (sample A3) increased the leakage current immediately after the process, compared to mechanically polished water (sample A1). After 14 days, the current values for the passivated sample returned close to that of the mechanically polished wafer.

The results of Am-241 spectral responses for the mechanically polished (sample A1), chemo-mechanically polished (sample A2) and freshly passivated (sample A3) CZT wafers are shown in Figure 4. The chemo-mechanical polishing and surface passivation processes do not show significant channel shifts in the 59.5-keV peak of Am-241. The Am-241 spectral responses for the passivated CZT wafer (sample A3) measured over a 14-day period are shown in Figure 5. These results show that the 59.5-keV peak of Am-241 was fairly steady under the same channel over the test period.

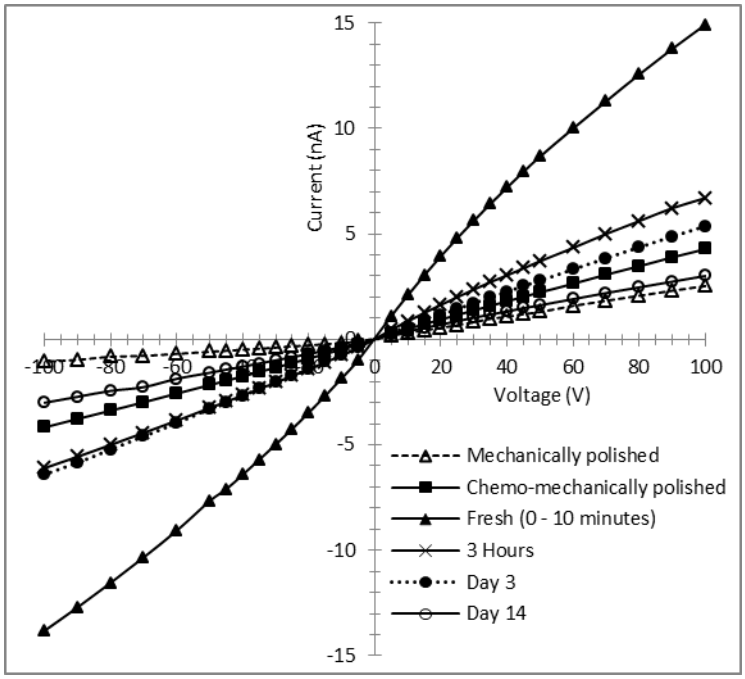

Figure 3. The results of the I-V measurements up to 14 days after surface processing.

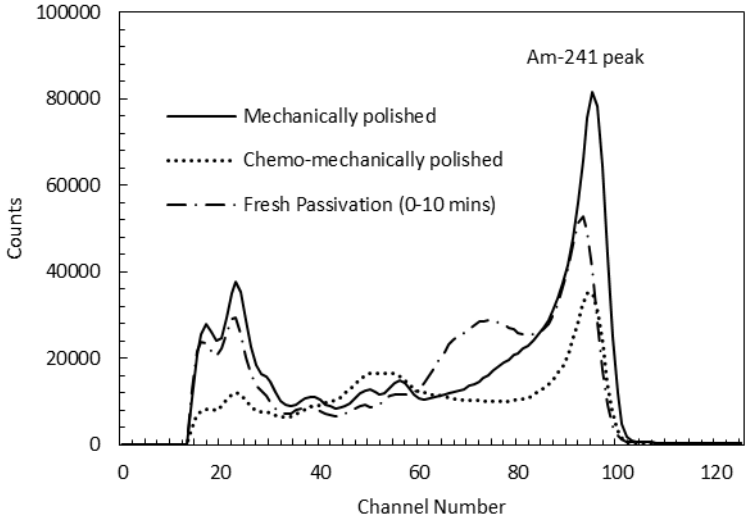

Figure 4. Am-241 spectral responses for the three CZT wafers: mechanically polished (sample A1), chemo-mechanically polished (sample A2) and freshly passivated (sample A3).

4. CONCLUSIONS

The CdZnTe crystal exposed to chemo-mechanical polishing using hydrogen bromide solution and passivation treatment for 1 minute showed the formation of tellurium-rich surfaces. The results presented in this paper provide data that validate previous studies in the fabrication of radiation detectors, specifically CdZnTe, with the aim of improving the processes that affect device performance.

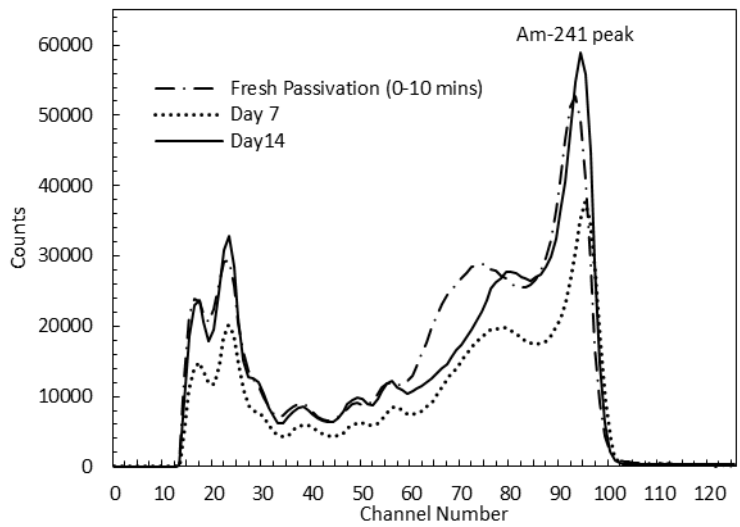

Figure 5. Am-241 spectral responses for the passivated CZT wafer (sample A3) over a 14-day period.

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