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The development of super-sensitive glass track-etch detectors

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Abstract. Working from a barium-phosphate laser filter glass called VG-13, Buford Price and his colleagues at Berkeley developed BP-1 glass track-etch detectors several years ago. BP-1 exhibits spectacular and unparalleled resolution in the measurement of charge of relativistic heavy ions. Since its development, BP-1 has accumulated an impressive track record of successful applications in experimental astrophysics, in the study of nuclear interactions of relativistic heavy ions, in the study of cluster radioactivity, and even in atomic physics at high Lorentz factor. BP-1 was only very crudely optimized for sensitivity and resolution, so there is no reason to expect that BP-1 happens by chance to have the optimal composition with respect to sensitivity, even among phosphate glasses with identical sets of components. We have two independent sets of evidence—from the analysis of calibration data from the Trek instrument, and from a search for ^{12}C emission from ^{114}Ba—which strongly indicate that a much more sensitive composition exists. We plan to develop the successor to BP-1, which we will call BP-2, by experimentally exploring the glass composition space near the nominal BP-1 composition to find the true maximum in sensitivity. For minimum-ionizing relativistic heavy ions, BP-1 has a detection threshold at $Z = 68$ when etched in the most sensitive etchant (HBF_4). By optimizing the sensitivity, we hope to lower the threshold to $Z < 50$, which would enable us to reach the astrophysically interesting region around the second heavy abundance peak at Sn–Ba.

1 Development and Applications of BP-1

Until the mid-1980s, plastic track-etch detectors (*e.g.,* Lexan) had been in common use in cosmic-ray astrophysics projects. These detectors suffered from rather large dependence of response on registration temperature and on oxygen partial pressure. These characteristics are particularly inconvenient for balloon- and spacecraft-borne missions, since it follows that very precise thermal regulation and pressure-tight containers are required. Motivated by the lack of such difficulties among inorganic track-etch detectors, beginning in 1985 Buford Price and his colleagues (including one of us, AJW) at Berkeley conducted a systematic search for a highly sensitive glass track-etch detector to replace plastic track-etch detectors. They tested numerous silicate, phosphate, arsenate, and vanadate glasses of various compositions, including many laser-filter glasses. One barium phosphate glass, VG-13, manufactured by Schott Glass Technologies as a green laser filter glass, showed unusual sensitivity and charge resolution (Price *et al.*, 1987). The green color was due to a small concentration of uranium, which was undesirable; also, the glass was somewhat hygroscopic. With the help of Shi-Cheng Wang, we manufactured glasses with compositions similar to that of VG-13, but lacking uranium. We found a quaternary glass, BP-1 (Wang *et al.*, 1988), which exhibits spectacular sensitivity and resolution, lacks uranium, is optically clear, and is only slightly hygroscopic. After the discovery of BP-1 in the Berkeley lab, BP-1 has been manufactured commercially by Schott Glass Technologies.

For more than ten years, our group at U. C. Berkeley has used the track-etch detector glass BP-1 in a variety of applications. During that time we have found a surprising number of applications for this detector. One of the first applications of BP-1 detectors was in the study of nuclear collisions of relativistic heavy ions (Westphal *et al.*, 1991, 1992). We have recently conducted a search for cluster radioactivity (Price, 1989) of the proton-rich nucleus ^{114}Ba. Based on the non-observation of any ^{12}C candidates, we were able to place a stringent upper limit on the branching ratio for ^{12}C emission by ^{114}Ba (Guglielmetti *et al.*, 1997). Yudong He and Buford Price searched for the production of Dirac magnetic monopoles and other hypothetical highly ionizing particles produced in relativistic heavy ion collisions, setting an upper limit well below the Drell-Yan cross-section (He, 1997). BP-1 has also found applications in atomic physics. We measured for the first time the cross-sections for electron cap-

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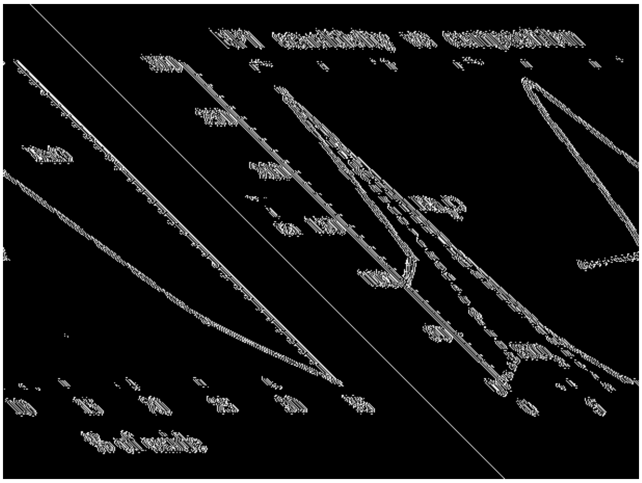

Fig. 1. Study of the sensitivity of test melts of quaternary P_2O_5-BaO-SiO_2-Na_2O glasses as a function of composition (weight percent). The sensitivity data are sparsely sampled, giving only a general indication of the location, magnitude and sharpness of the sensitivity maximum.

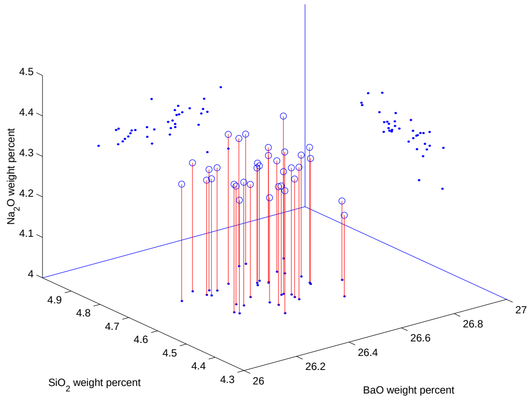

Fig. 2. Scatterplot of composition of 35 BP-1 melts for the manufacture of Trek detectors. The Schott XRF composition data were only recorded with a precision of 0.1 wt.%, so a small amount of random Gaussian noise (width = 0.02 wt.%) was added to the data to separate the individual points from each other.

ture and stripping by ions with Lorentz factor greater than 10 (Westphal and He, 1993). Finally, BP-1 has performed superbly in the measurement of elemental and isotopic composition of Galactic cosmic rays (GCR), *e.g.*, Ni isotopes (Westphal *et al.*, 1996). The Trek detector, deployed in 1991 on the external surface of the Russian space station *Mir*, measured the elemental abundances of the heaviest elements in the GCR. The technical details of this detector were reported by Weaver *et al.* (1998). The composition found by Trek (Westphal *et al.*, 1998) was in dramatic disagreement with the leading model of cosmic ray origin (Meyer, 1985). Recently we have further improved the charge resolution for a subset of Trek cosmic-ray events (Weaver and Westphal, 2001a,b).

2 Evidence for Super-Sensitive Glasses

BP-1 was only the most sensitive of the glasses made in a very coarsely-spaced study of the sensitivity as a function of composition. We show the results of the study in Figure 1. In addition to the obvious sparseness of the data sampling (the curves are arbitrarily drawn to smoothly go through the data), we also point out that the sensitivity was optimized using low-energy fission fragments. This was done for reasons of expediency, and of course the resulting glass turned out to perform superbly — perhaps by accident — in applications involving relativistic heavy ions. But there is absolutely no reason to assume that the sensitivity maximum for fission fragments coincides with that for relativistic heavy ions. In addition to the *a priori* unlikelihood that BP-1 happens by accident to have the optimal sensitivity among all possible P_2O_5-BaO-SiO_2-Na_2O glasses, we have three independent pieces of evidence that point to a more sensitive detector.

First, in order to determine composition tolerances for the anticipated manufacture of more than two tons of BP-1 for next-generation GCR detectors, we have studied the dependence on response as a function of composition for BP-1 detectors which constituted the Trek instrument. Each stack of Trek glass detectors was calibrated with a 10.8 A GeV Au beam at the AGS at Brookhaven. For the so-called Extended-Trek analysis (Weaver, 2001), we have analyzed glass detectors which were calibrated with Au at two angles. This allows us to measure the detector response without reference to the amount of material removed during etching. For Extended-Trek, we physically removed a circular wafer from each detector, centered on the GCR track. Extended-Trek wafers have been etched in two batches, in lots of 640 wafers each. Each wafer was scanned using our automated microscopic scanning system, and the calibration signal for the 10.8 A GeV Au events was measured. We then examined the correlation of the calibration signals with the composition of each wafer. Individual wafer compositions were measured batch-by-batch by XRF at Schott at the time of the Trek detector manufacture. In Figure 2 we show a scatterplot of the individual compositions. In Figures 3 and 4, we show the median calibration signal, with errors estimated from the dispersion in the data, versus XRF composition for the two major components. Two independent sets of data from two separate etches are shown; the agreement between the datasets is remarkable.

There are three points to be made about these measurements. First, we are sampling over only a very small region of composition space, so the variations in sensitivity are correspondingly small. The largest difference in sensitivity is the equivalent of $< 0.5e$ charge shift. Second, it is obvious that the dispersion in the data is much larger than the estimated errors. This is to be expected, because for any given variation in composition (*e.g.*, the weight fraction of P_2O_5 in Figure 3) *we are not holding the other fractions even ap-*

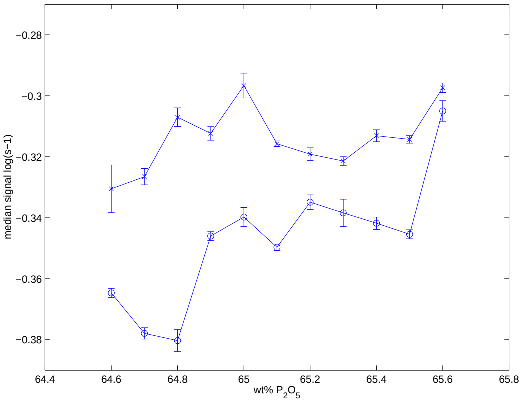

Fig. 3. Median 10.8 A GeV Au calibration signal versus P_2O_5 weight fraction.

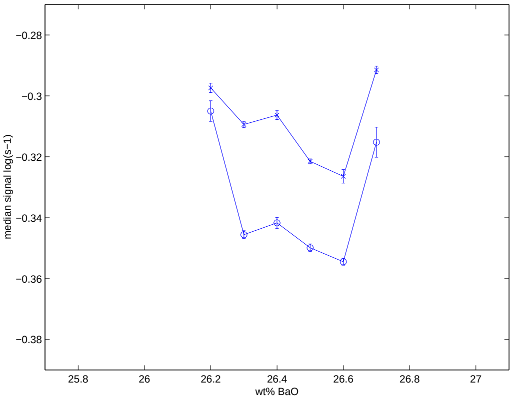

Fig. 4. Median 10.8 A GeV Au calibration signal versus BaOweight fraction.

proximately constant. This is because we are dealing with a very small number (35) of distinct melts. Third, despite the limitations of the small sampling statistics, the trends are clear in the two independent data sets — and we emphasize that these datasets come from two distinct sets of detectors which were etched at two different times. (The overall shift in sensitivity is due to variations in etching conditions.) The data strongly suggest that sensitivity improves with increasing P_2O_5, decreasing SiO_2, decreasing Na_2O. The surprise is that BP-1 may be near a *minimum* in sensitivity with respect to variations in the fraction of BaO! We recognize, of course, that these data are not definitive, since the precision of the XRF data is poor, and we are only taking the melts that we happened to have received from the manufacture of the Trek detectors — this is not a controlled study, in which only one parameter is varied at a time. But the data strongly support the proposition that, as one might expect, we did not happen by pure luck to find in BP-1 the optimal composition among all possible P_2O_5-BaO-SiO_2-Na_2O glasses.

Second, as mentioned above, in 1996 we conducted a search for a mode of radioactivity in which ^{114}Ba decays to ^{102}Sn by emission of ^{12}C. The experimental apparatus consisted of an Al sphere which was tiled on the inside with BP-1 detectors. ^{114}Ba was produced at the UNILAC at GSI, then electromagnetically separated and implanted in a carbon catcher foil in the center of the sphere. After the exposure, the BP-1 detectors were etched and scanned both automatically and by eye. No etch-pits consistent with 15–17 MeV C ions were detected, so we placed an upper limit of 3.4×10^{-5} for the branching ratio of ^{12}C with respect to α-decay.

During scanning, we found three events which were definitely not due to decay of ^{114}Ba — they did not point back to the catcher foil — but were of some interest since similar events which did happen to point in the right direction could in principle produce an irreducible background. (We have since concluded that these three events were caused by rare light cosmic rays.) We scanned the reverse sides of the BP-1 detectors to measure this background. We found no events of the type found on the obverse sides of the detectors, but one of us (PBP) found several intriguing clusters of etch-pits, one example of which is shown in Figure 5. Having done calibrations with lithium, beryllium, and boron ions, he judged that these were similar in general appearance, but must have been due to α-particles, which could only be true if the glass were locally much more sensitive than overall. (The reason for ruling out Li, Be, or B in favor of He was simply that only α-particles could provide a plausible explanation for the very high density of tracks.) Using an electron microprobe, we scanned the area to try to detect compositional changes in the sensitized region, but the results were inconclusive, probably because of the irregular geometry of the etched surfaces. We searched for more examples of such etched tracks in similar detectors held in cold storage, and found only a few examples of regions of locally high sensitivity out of several hundred cm^2. If the hypothesis of sensitivity to α-particles is correct, it points the way toward development of a highly sensitive glass detector which could be used in place of CR-39. Such a detector would have advantages over CR-39 in being functional *in vacuo* and immune to the effects of sunlight. Also, if our experience with BP-1 is any indication, it might have far superior charge resolution.

Third, very recently we have found additional evidence of super-sensitive regions in BP-1. During a routine scan of a sheet of glass from the Trek detector, one of us (AJW) found the region shown in Figure 6. This region appears to be almost saturated with tracks. The density of 10.8 A GeV Au nuclei to which this sheet was exposed was far to low to account for these tracks. We are still investigating, but tentatively we conclude that this region is recording the tracks of very light particles.

3 Conclusion

We are convinced that a more sensitive barium-phosphate glass exists. We are now preparing to develop BP-2, the optimized successor to BP-1. As we have shown in the review

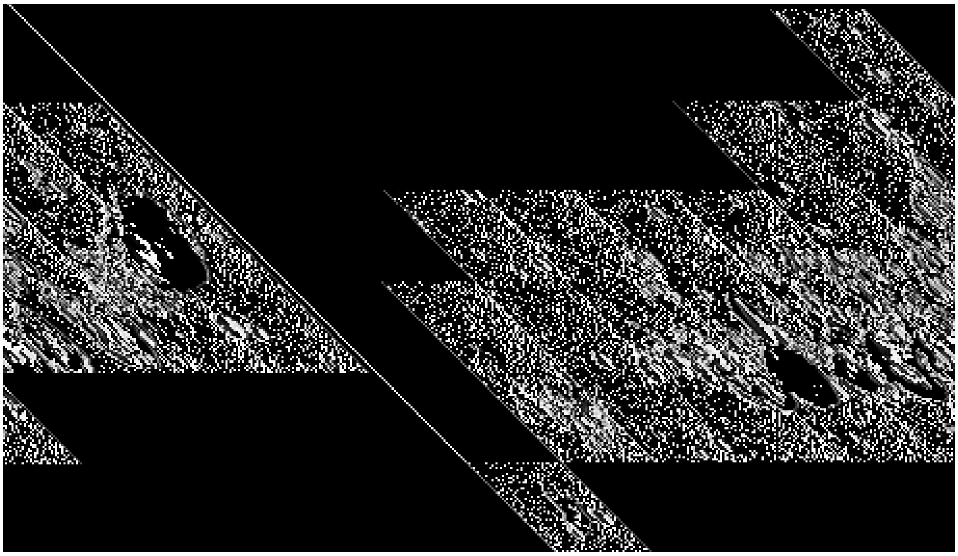

Fig. 6. A recently discovered apparent super-sensitive region of BP-1 glass. The large elliptical etch-pit in the left-center is due to a Au calibration track. The region is approximately 1 mm in width. A larger version of this figure is at http://ultraman.ssl.berkeley.edu/˜domi/.

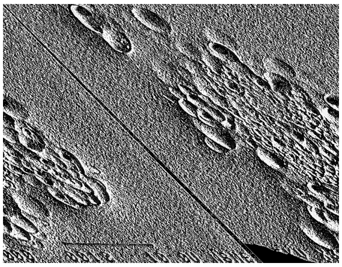

Fig. 5. SEM image of a cluster of apparent etched nuclear tracks in BP-1 detectors.

of applications of BP-1, the applications of a super-sensitive glass detector cannot be fully anticipated. However, we have already identified an application in cosmic-ray astrophysics. For minimum-ionizing relativistic heavy ions, BP-1 has a detection threshold at $Z = 68$ when etched in the most sensitive etchant (HBF_4). By optimizing the sensitivity, we hope to lower the threshold to $Z < 50$, which would enable us to reach the astrophysically interesting region at and just above the second heavy abundance peak at Sn–Ba. We think that this goal is actually rather modest; a shift in sensitivity of this magnitude (albeit in the other direction) is currently achievable in BP-1 just by choosing a different etchant.

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