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Radioactivity Measurements Using Storage Phosphor Technology

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Introduction

We propose to apply a recently developed charged particle radiation imaging concept in bio-medical research for fast, cost-effective characterization of radionuclides in contaminated sites and environmental samples. This concept utilizes sensors with storage photostimulable phosphor (SPP) technology as radiation detectors. They exhibit high sensitivity for all types of radiation and the response is linear over a wide dynamic range (>10^5), essential for quantitative analysis. These new sensors have an active area of up to 35 cm x 43 cm in size and a spatial resolution as fine as 50 μm. They offer considerable promise as large area detectors for fast characterization of radionuclides with an added ability to locate and identify hot spots.

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Tests with SPP sensors have found that a single alpha particle effect can be observed and an alpha field of 100 dpm/100 cm^2 or a beta activity of 0.1 dpm/mm^2 or gamma radiation of few μR/hr can all be measured in minutes. Radioactive isotopes can further be identified by energy discrimination which is accomplished by placing different thicknesses of filter material in front of the sensor plate. For areas with possible neutron contamination, the sensors can be coupled to a neutron to charged particle converter screen, such as dysprosium foil to detect neutrons. Our study has shown that this approach can detect a neutron flux of 1 n/cm^2s or lower, again with only minutes of exposure time. The utilization of these new sensors can significantly reduce the time and cost required for many site characterization and environmental monitoring tasks. The "exposure" time for mapping radioactivity in an environmental sample may be in terms of minutes and offer a positional resolution not obtainable with presently used counting equipment. The resultant digital image will lend itself to ready analysis.

The SPP technology has been investigated for applications to medical x-ray imaging and is well proven in molecular biology and radiopharmaceutical research[1-4]. The concept of the technology involves radiation-caused trapping in a sensor plate and release of the trapped energy as light when the sensor is stimulated under the scanning of a laser beam in a reader unit. A photomultiplier tube detects the light and coordinates the light intensity with the scanned positions, creating a digital image of the radiation field. After scanning, the sensor plate can be quickly cleaned and reused again. The separation of the sensor, a semi-flexible plastic plate coated with a thin layer of a special phosphor material, from the laser reader unit means that multiple sensor plates can be used simultaneously for radiation monitoring, thus increasing the effective detector area. For environmental monitoring, this two-step process involved in the SPP application may prove to be advantageous, in that multiple imaging sensor plates can be used to cover a large area of interest, or to assay a group of samples simultaneously. After a short time of exposure the sensor plates can be collected and taken to a data reading station for reading. The reading station consists of a PC-class computer and a laser readout unit which in its current configuration resembles a medium size printer. The station can be placed in a small vehicle or mounted on a cart. The reading process takes about two minutes per sensor plate. Afterwards sensor plates can be quickly cleaned by exposing to a UV source in an eraser and be reused again.

Experiment and Results

A test was conducted on one type of SPP imaging sensors with $BaFBr:Eu^{2+}$ as the phosphor to determine its potential as an alpha particle detector. Two National Institute of Standards and Technology (NIST) standard alpha sources AA840 and AA370 (^{238}Pu, 12.75 cm x 20.25 cm active area, 3.85×10^2 dps and 2.33×10^3 dps respectively) were used. Figure 1 shows the results of 20 second and an 100 second exposure respectively with the source AA840 being held 0.4 cm away from the SPP imaging sensor plate. The 20 second exposure is shown on the left and the 100 second exposure is shown on the right with background area of the imaging sensor plate in the center. A hot spot in the source was detected in the 100 second image and was also visible in the inverted 20 second exposure image. The table in figure 1 lists the integrated relative radiation strength in the three areas defined by the circles. The result of a two second exposure of the same source is shown on the right in figure 2. The 16-fold enlargement of an area defined by the small square in the two second exposure is shown on the left. It is interesting to see the marks left by individual alpha particles. The energy of a ^{238}Pu alpha particle is about 5.5 MeV and it appears to affect about 10 to 15 pixels which can be readily distinguished from background. When an alpha particle entered the imaging plate at an angle far away from normal, a track was formed much like that in a cloud chamber. The two second exposure represents a counting sensitivity of 0.03 alpha/mm^2 and shows a signal-to-noise ratio (SNR) of 15. Figure 3 shows the integrated SPP sensor response as a function of exposure time for the two NIST alpha sources. There is excellent linearity.

The principle of the SPP technology indicates that it is sensitive to the incident energy of radiation. The sensor system, therefore in addition to possessing the capability of imaging individual alpha particles, has the potential to identify unknown alpha emitters through energy differentiation. To explore the possibility of radionuclide speciation by the SPP technology, transmission measurements were conducted with several alpha and beta sources. Filters of various thicknesses were placed between the sources and a SPP sensor plate to measure the attenuation

effect of the filter material as a function of material thickness. The filter material used was DuPont MYLAR polyester film #15 XM555 with a density of 0.552 mg/cm^2.

Figure 4 shows the results of transmission measurements of a group of five alpha emitters by the SPP technology. For each source the SPP imaging sensor responses were normalized to their measurement responses with no filters. The slopes of the response curves indicate the attenuation property of the filter material with respect to different incident energies and follow a predicted pattern that lower energy alphas were being filtered out faster. Notice that the response of ^{241}Am is very close to ^{238}Pu, both emitting alpha particles with energy around 5.5 MeV. However, ^{241}Am also emits a higher percentage of x- and γ-rays and shows an appreciable residual spectrum compared to ^{238}Pu. ^{243}Cm also shows a residual spectrum from radiation other than alpha particles. Figure 5 shows the same type of test with two ^{238}Pu and one ^{239}Pu source. The two ^{238}Pu sources while differing in strength by a factor of 150 produce similar response curves. The ^{239}Pu source with an alpha energy around 5.1 MeV however shows a slightly steeper response slope.

Figure 6 shows the same type of test with two depleted uranium samples. The two samples exhibit very different response curves marked AA101 and AA102 respectively, indicating they have different matrix compositions. Both samples show considerable residual transmissions after 31 μm of MYLAR filter. The test results with ^{238}Pu and ^{239}Pu in figure 5 demonstrate that after 31 μm of MYLAR filter, practically all alpha particles with energies less than 5.5 MeV will be filtered out. Therefore the residual transmissions shown in AA101 and AA102 were from more penetrating radiations such as beta, gamma or x-rays. If these residuals were subtracted from their respective response curves however, the subtracted curves for the two samples marked AA101-X and AA102-X respectively would behave similarly. The residual subtracted curve would fit in between that of ^{208}Po and ^{148}Gd in figure 4, suggesting that the main alpha activities in both uranium sample were from ^{238}U. The energy differentiation property of the SPP sensor system for alpha particles is summarized in figure 7. The slope of attenuation is defined as $(\mathrm{Ln}(A_t)-\mathrm{Ln}(A_0))/t$ where A_t denotes the activity of the source measured by the SPP sensor with a MYLAR filter of thickness t. The residuals have been subtracted from the response measurements of ^{243}Cm, ^{241}Am and ^{238}U sources before their slopes of attenuation were calculated.

The energy differentiation capability of the SPP system to beta particles was also tested and the results are presented in figures 8. An extended and more detailed test with ^{147}Pm is shown in figure 9. That extended test, covering over six orders of magnitude in beta activity measurement clearly demonstrates the high sensitivity and excellent linearity of the SPP sensors to beta particles. These tests with alpha and beta emitters suggest that the SPP sensor system can be utilized as a fast, high spatial resolution and economical radionuclide analyzer for waste characterization applications. Unknown waste samples can be quickly screened to determine the types of radiation and then the specific radioactive isotopes involved can be identified. Radioactivity dosage can be accurately measured when the SPP sensor system is calibrated.

Another test was carried out to confirm the sensitivity of the SPP imaging plate to beta particles and to demonstrate how the new sensor can be used for neutron detection[5] as well. A metal foil with high neutron absorption cross-section and other desired nuclear properties such as having a simple decay scheme and a manageable half-life is used as a neutron to

charged-particle converter in conjunction with a SPP sensor system. A dysprosium (Dy) foil was activated in a nuclear reactor cold neutron beam and the decay history of the foil was subsequently traced by placing the foil in close contact with a SPP sensor in a series of timed-exposures. A commercially prepared 125 μm thick Dy foil about 1.5 cm x 1.5 cm in size was exposed to a cold neutron flux of 3 x 10^7 n/cm^{2}s for 30 seconds at the NIST cold neutron research facility. Figure 10 presents the logarithm of the fraction of radioactivity remaining in the dysprosium foil as a function of time. I(t) is the relative radiation strength of the foil at time t after the neutron activation as recorded by a SPP imaging plate. The imaging sensor plate exposure time to the neutron activated foil was set for 5 minutes for measurements conducted when t is less than 30 hours and was set for 20 minutes when t is greater than 30 hours except that for the last measurement at t = 40 hours then the exposure was set for 1 hour. All data were normalized to a 5 minutes imaging plate exposure time for the graphing in figure 10. The thermal/cold neutron activation of Dy produces a relatively short-lived (half-life 1.26 minutes) metastable state of dysprosium-165 (^{165M}Dy) and the dysprosium-165 ground state (^{165}Dy) with a half-life value of 2.35 hours. ^{165}Dy decays mainly by emitting beta particles. The slope of the decay history line in figure 10 is consistent with the known ^{165}Dy half-life and all measurements were within 1% of the predicted decay values except that for t = 36.2 hours and 37.5 hours, which were within 5%. The slightly larger errors probably were caused by the foil not being in good contact with the imaging plate. The overall excellent linearity of the SPP sensor system response exhibited in figure 10 suggests it can be used in precision quantitative analysis of neutron distributions. Figure 11 shows the SNR of the SPP imaging sensor plate as a function of time in the tracking of the same Dy foil decay. The time is expressed in units of ^{165}Dy half-life.

The time of 40 hours represents about 17 half-lives or a decay factor of more than 130,000 for ^{165}Dy. Figure 11 suggests that at 18 half-lives after activation, a decay factor of more that 260,000, the remaining beta activity in the Dy foil could still be measured with a SNR of 5. The Dy foil was activated in a cold neutron flux of 3 x 10^7 n/cm^{2}s for 30 seconds and generated an estimated ^{165}Dy radioactivity of 2 x 10^4 dpm/mm^2. After decaying for 17 half-lives, the remaining ^{165}Dy radioactivity in the foil would be about 0.15 dpm/mm^2 and equivalent to that of a foil at the end of a 30 second activation with a neutron flux of 2 x 10^2 n/cm^{2}s. If left decaying for 18 half-lives, the remaining ^{165}Dy radioactivity would again be half as much at 0.07 dpm/mm^2, similar to a foil activated for the same 30 seconds with a neutron flux of 1 x 10^2 n/cm^{2}s. A foil exposed to a neutron flux of 1 n/cm^{2}s for about one hour will also generate a similar ^{165}Dy radioactivity. This test demonstrates that the SPP sensor system when coupled with a proper neutron to charged-particle converter can function as a very high sensitivity large area neutron detector. Simple metal foils can be placed in a neutron contaminated area. After a short time the foils can be collected and put in contact with SPP sensors to register the neutron effects.

As an example of how the new sensor technology can be useful in environmental monitoring of radionuclides, we sprinkled a small amount of soil sample from Rocky Flats on a SPP imaging sensor plate with a thin MYLAR foil as a barrier. The soil sample was taken from NIST standard reference material SRM 4353. The SRM certificate states that each bottle of SRM typically contains one or two "hot" particles. Figure 12 shows the image of the soil sample captured by a ten hour exposure with a SPP sensor. A "hot" spot was detected and a 16-fold enlargement of the image area is shown on the right. A threshold was set that only pixels of relative high values, or areas of high radioactivity

are shown. The table in the figure lists the relative radioactivity strength in units of intensity value PSL per 1 mm^2 at the four marked positions in the enlarged area. The table indicates that at position No. 1 there are clusters of particles with relatively high radioactivity, but these are not the only particles with high radioactivity. Figure 13 shows the SPP image of another batch of soil sample from the same SRM, also with ten hours exposure time. At first glance of the image shown on the left of figure 13, there does not appear to be any "hot" spots. We selected an area indicated by the square and enlarged it 16 times. Again by setting the threshold level so only pixels with the highest relative values in the defined area would remain. The table in the figure shows the relative radioactivity strength at the five marked positions. The radioactivities at four positions are "hotter" than the "hot" spot found in figure 12, each shows a level 400 times or more above background. It is estimated that an exposure time of 10 minutes or less will be adequate for the detection of these "hot" spots. The above examples demonstrate that quantitative analysis and mapping of environmental samples can be conducted in an efficient manner using the new sensors. A filter system can be designed to distinguish alpha particle response from that of betas or gamma rays. The proposed approach with the new sensors can reduce the cost and complexity of radionuclide characterization while improving throughput.

Merits of the Technology

As demonstrated in our initial evaluation, the SPP technology shows great potential that it can be developed to be a low operating cost, high sensitivity monitor for environmental radioactivity and site radionuclide characterization. It offers the following advantageous characteristics:

- high sensitivity, is capable of imaging single alpha particles, can be used to measure many types of radioactivities, alpha activity of 100 dpm/100 cm^2, beta activity of 0.1 dps/mm^2, low energy neutron flux of 1 n/cm^2s or x-ray level of 20 μR/Hr can be detected in 10 min. or less;

- energy sensitive, can identify radionuclide through energy differentiation;

- high resolution, detector size can be as large as 35 cm x 43 cm with pixel size as fine as 50 μm, is capable of locating concentrations of radioactivity such as "hot" particles;

- wide dynamic range and highly linear, can be used for quantitative analysis and digital output lends itself for ready image and data processing;

- low secondary waste generation, very thin polyester film can be used for contamination barrier when contact measurements of radioactive samples are required, SPP sensor plates are re-usable;

- low operating cost, one reader can support many re-usable SPP plates;

- quick implementation of the technology, once the performance characteristics are calibrated a laboratory system or field transportable unit can be assembled quickly by using mostly off-the-shelf components, also a microprocessor based portable/remote operable unit can be designed using currently available technology;

- potential for in liquid operation, having a protective polyester cover the SPP sensor

plates are moisture resistant and do not require dark room setup.

Future Activities

We will conduct more detailed measurements to further characterize the SPP technology and compare it to other radiation measurement methods. We will examine soil samples from Rocky Flats and from Oak Ridge to verify the sensitivity and analytical capability of the technology for environmental monitoring. After the sucessful laboratory demonstration, we will assemble a transportable unit for field test at suitable DOE sites. It should provide quick turn-around time for on-site quantitative soil radioactivity analysis. Meanwhile, we will also design and fabricate several large area thin alpha reference sources with various strength for field calibration to ensure quality of measurement results by the SPP technology. The reference sources will be made in collaboration with NIST and the DOW Chemical Co. and be traceable to NIST. We will adapt the technology to field condition and DOE requirements by establishing both GIS and CAD interfaces, hardening and reconfiguring system hardware. Experience gained from the field test will be incorporated into the design of a portable or remote operable unit to further broaden the area of applicability.

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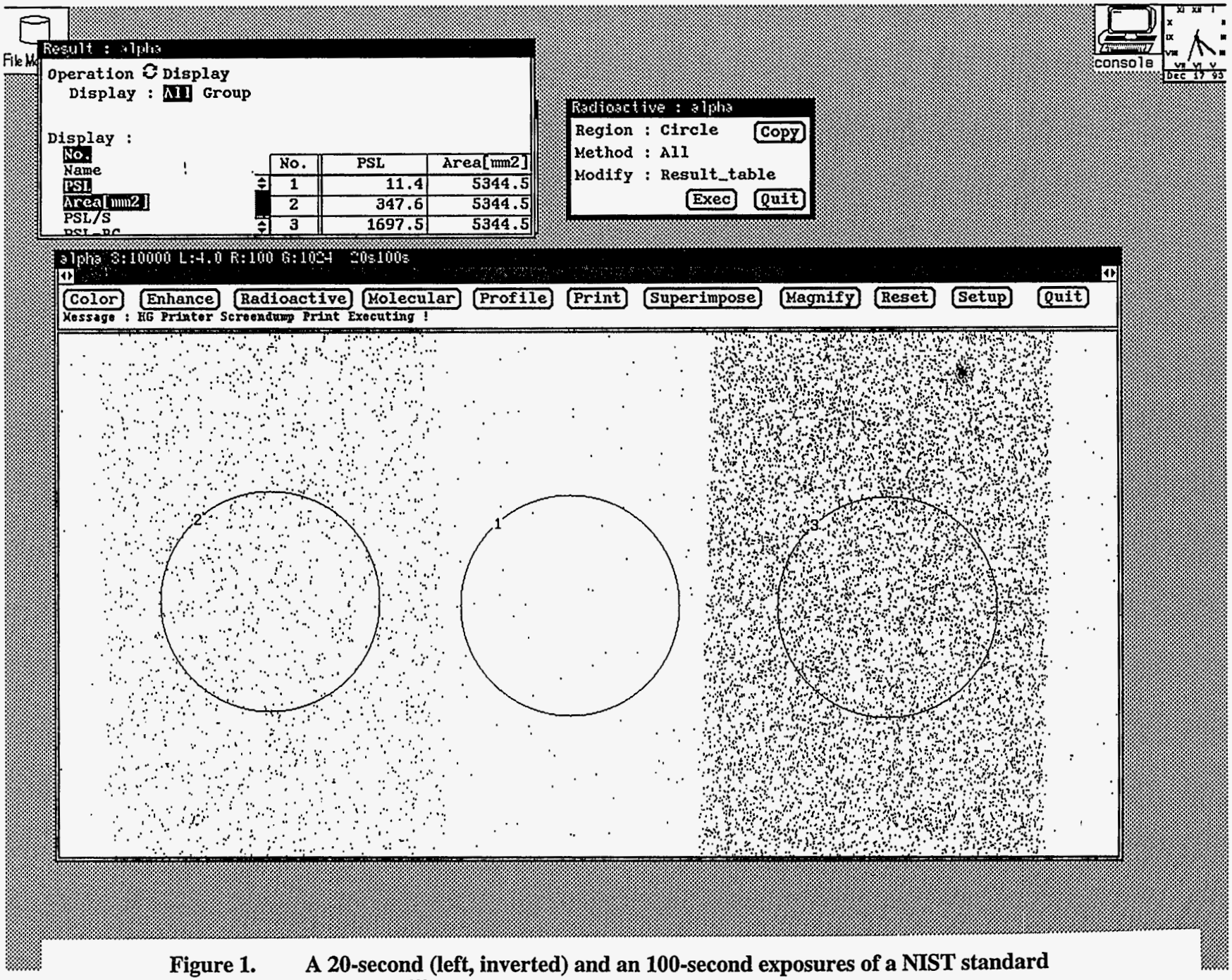

Figure 1. A 20-second (left, inverted) and an 100-second exposures of a NIST standard large area ^{238}Pu source with a SPP imaging sensor plate.

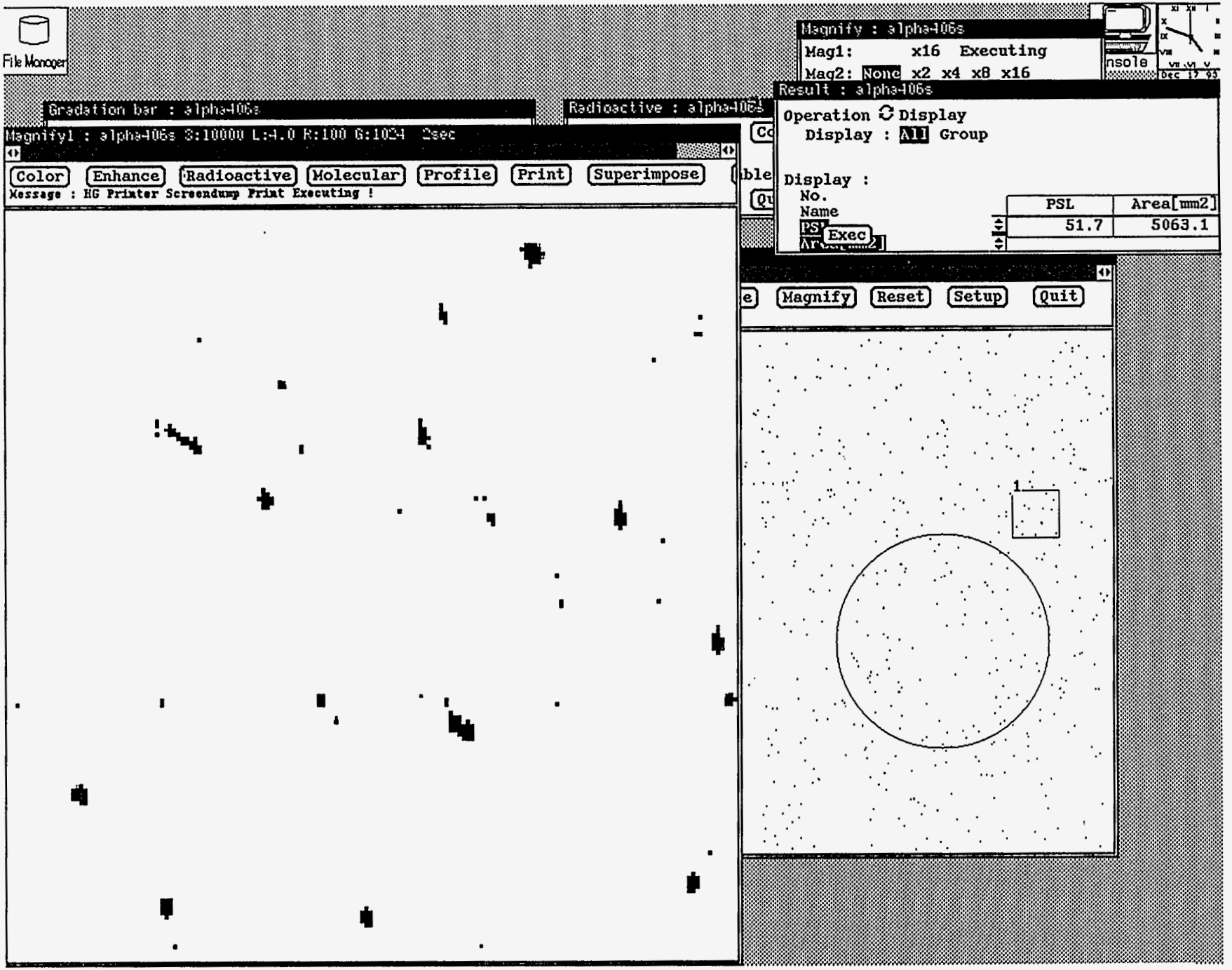

Figure 2. A two-second exposure (right) and detail enlargement of the same ^{238}Pu source with a SPP imaging sensor plate.

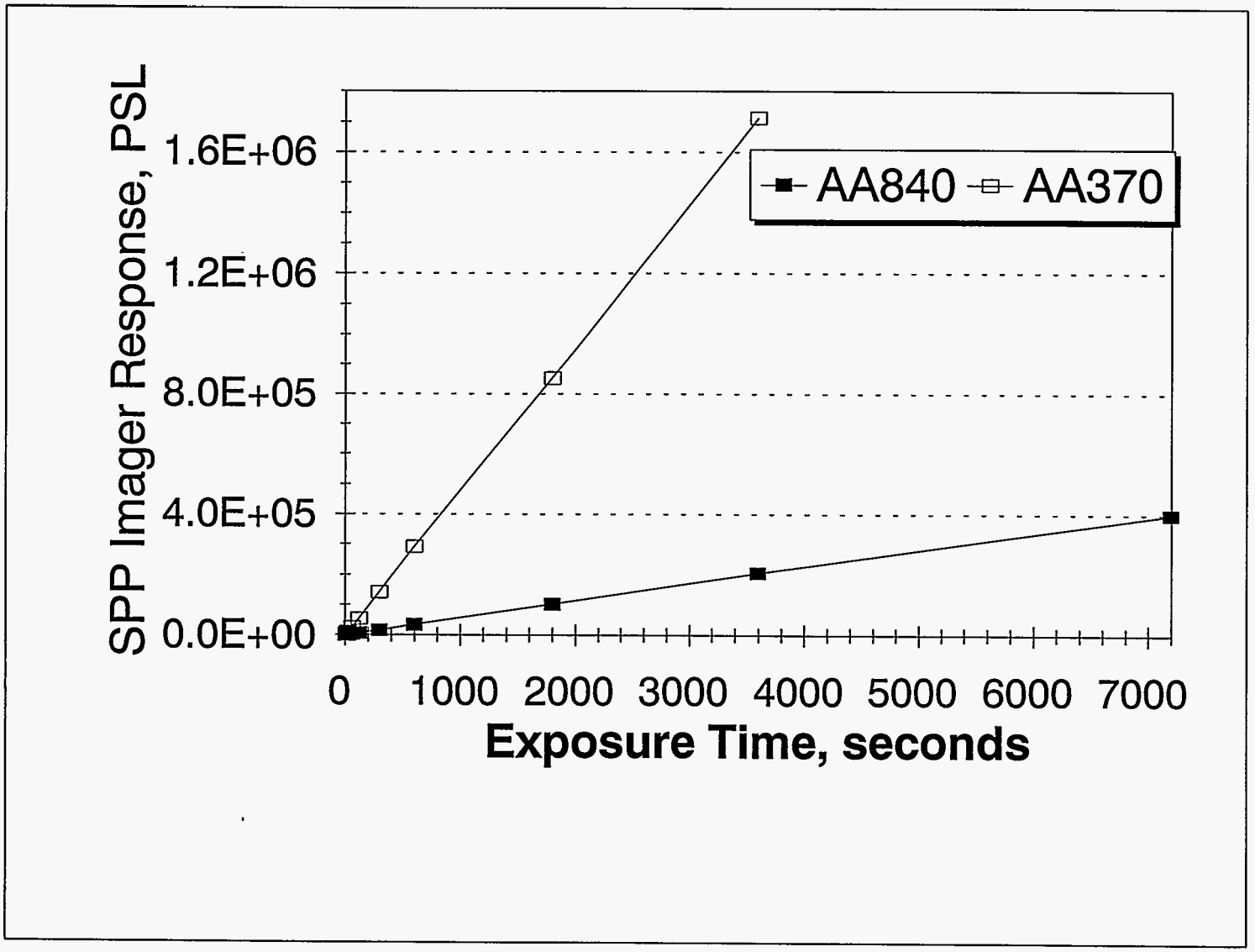

Figure 3. SPP sensor response as a function of exposure time to NIST large area ^{238}Pu sources AA840 and AA370.

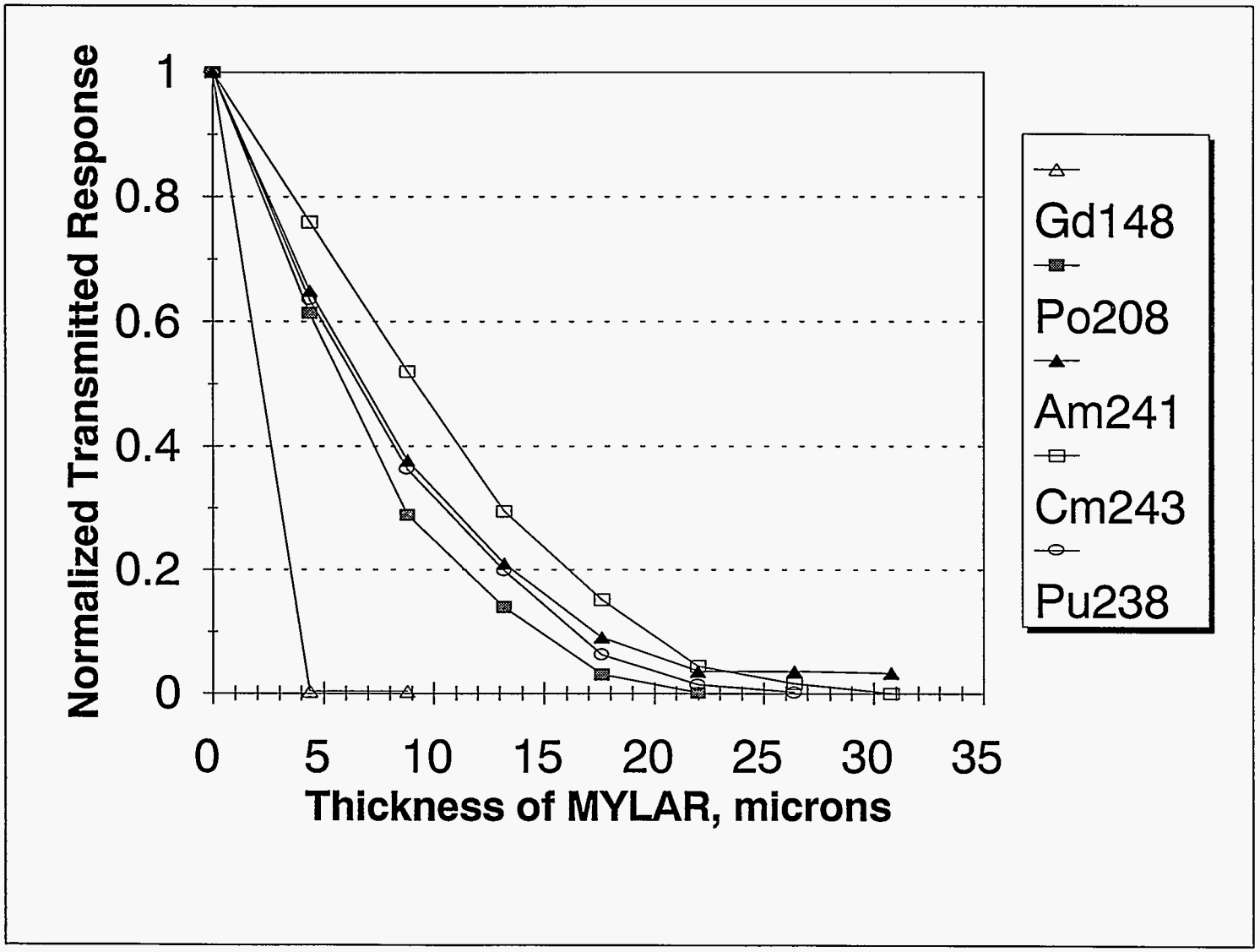

Figure 4. **Radioactivity attenuations of five alpha sources in MYLAR.**

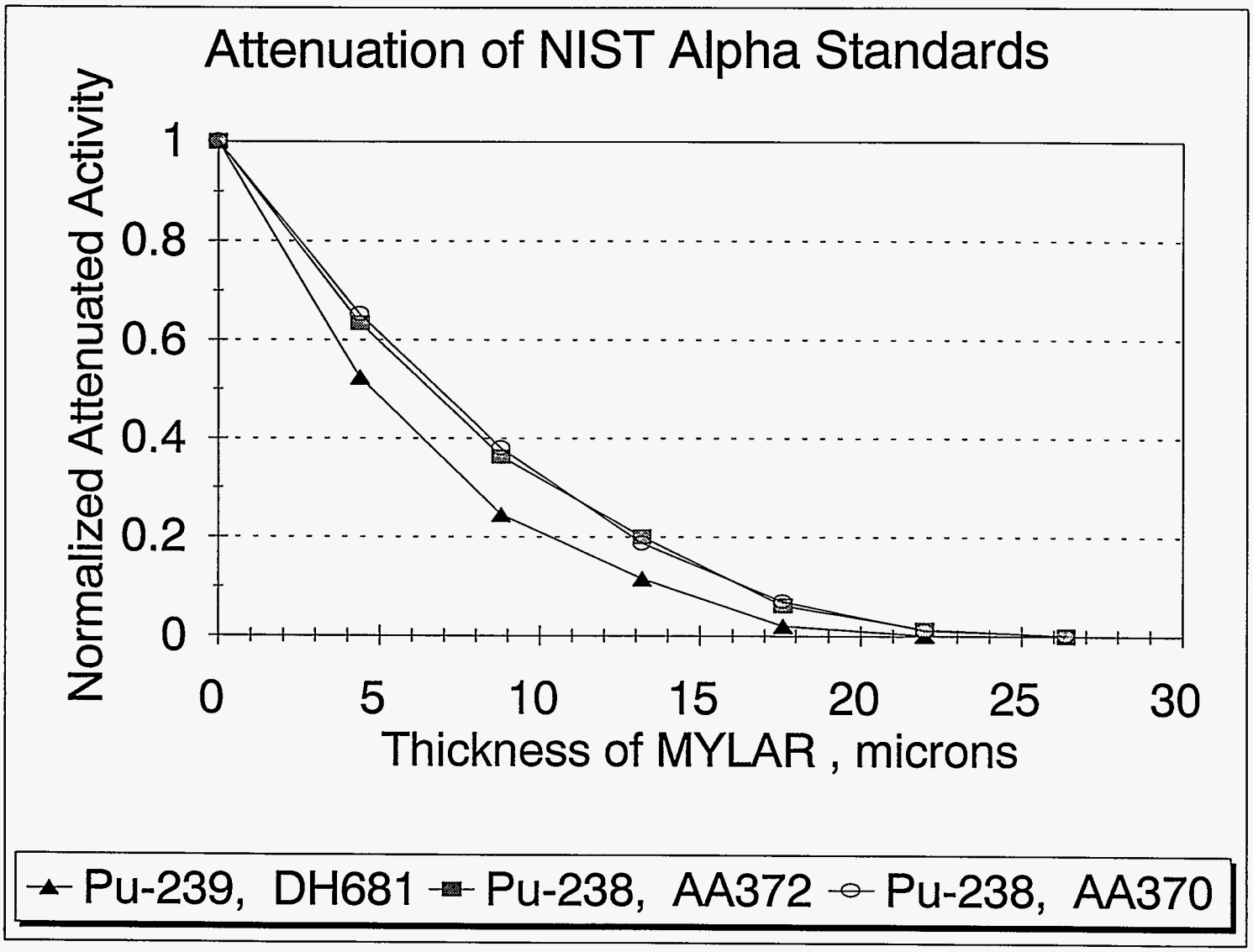

Figure 5. **Comparison of ^{238}Pu radioactivity attenuation in MYLAR to that of ^{239}Pu with a SPP sensor.**

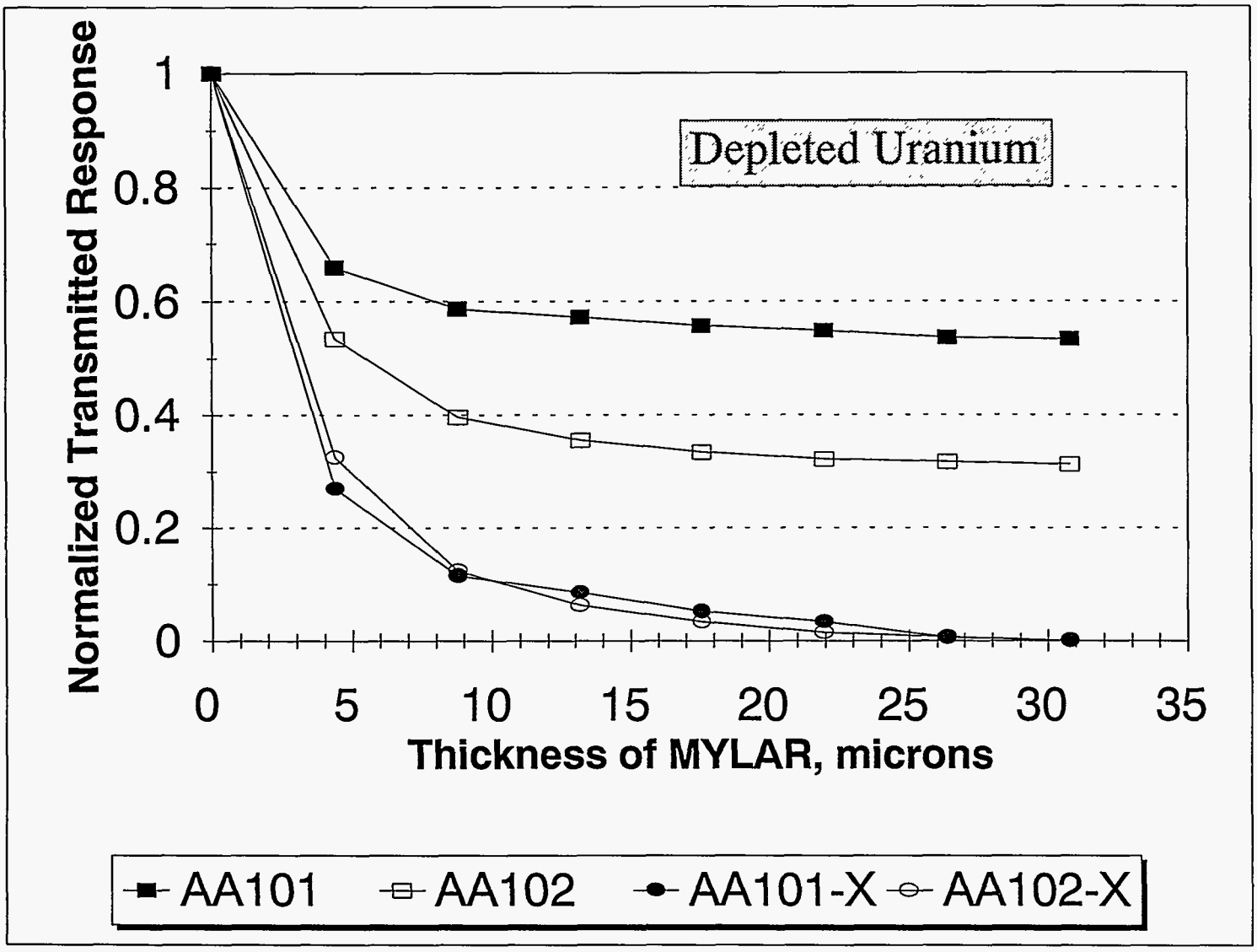

Figure 6. **Radioactivity attenuation of two depleted uranium samples in MYLAR.**

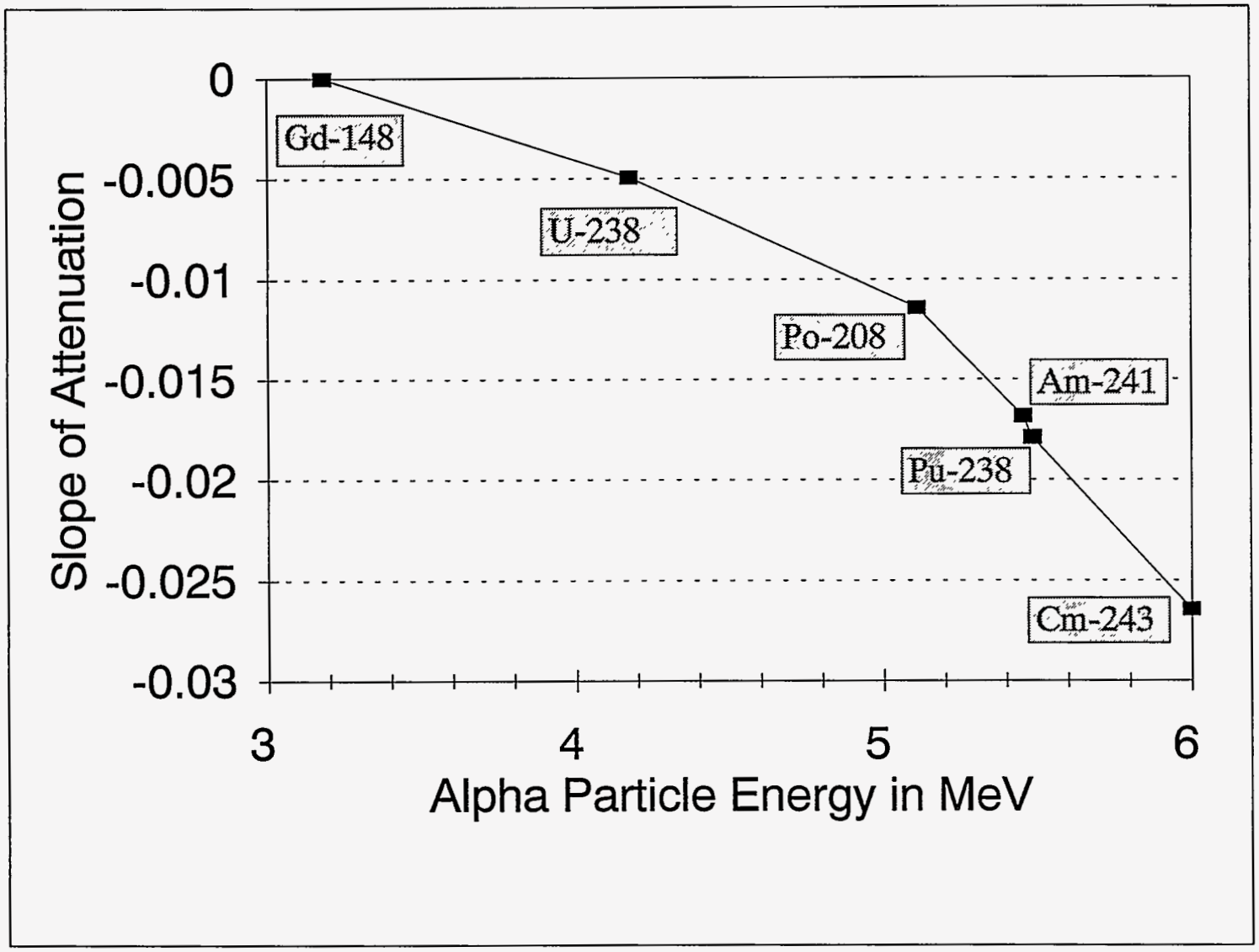

Figure 7. **Slopes of attenuation curves of alpha emitters vs. alpha particle energy measured by the SPP technology.**

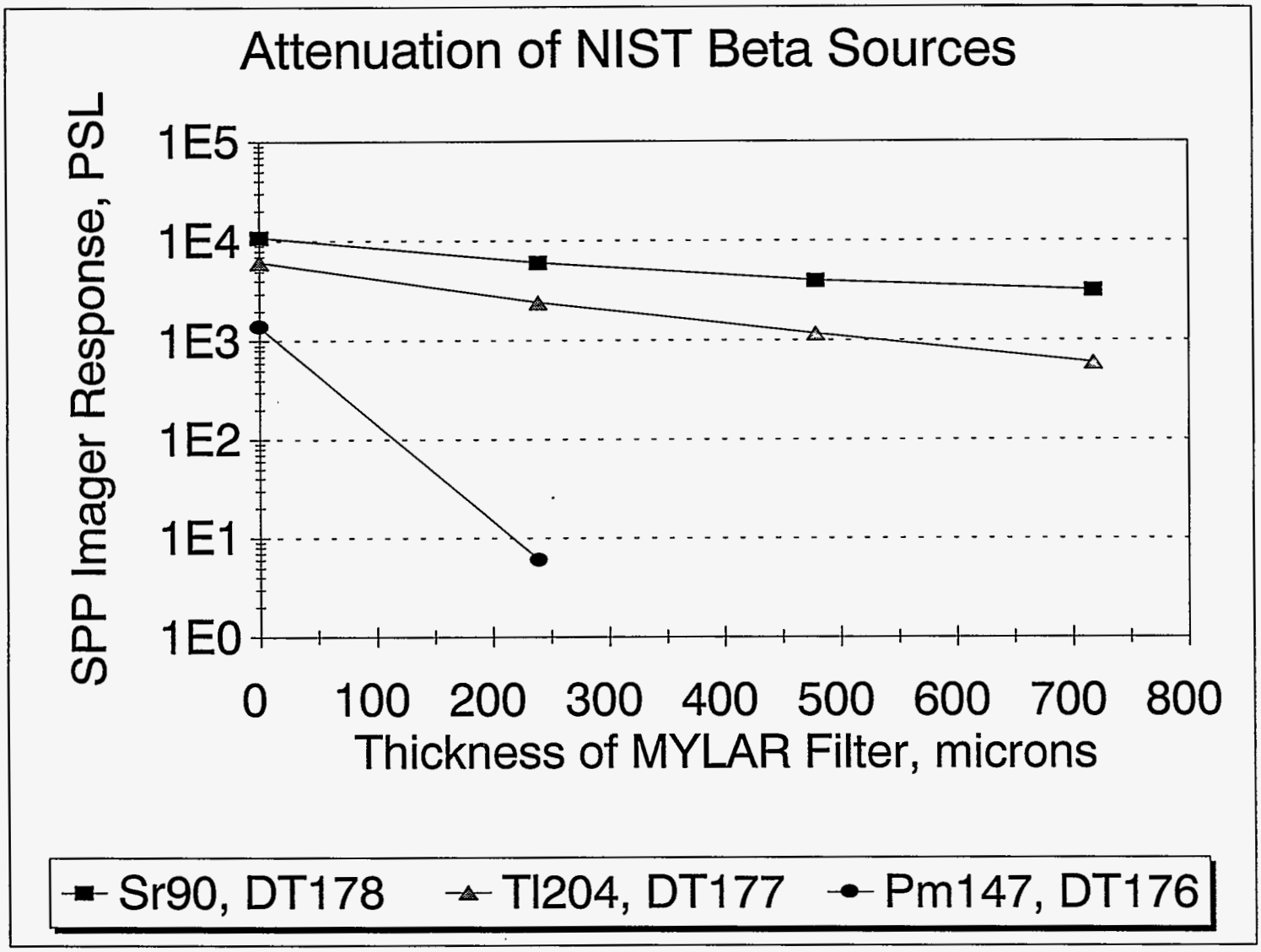

Figure 8. Attenuation of ^{90}Sr, ^{204}Tl and ^{147}Pm radioactivity in MYLAR.

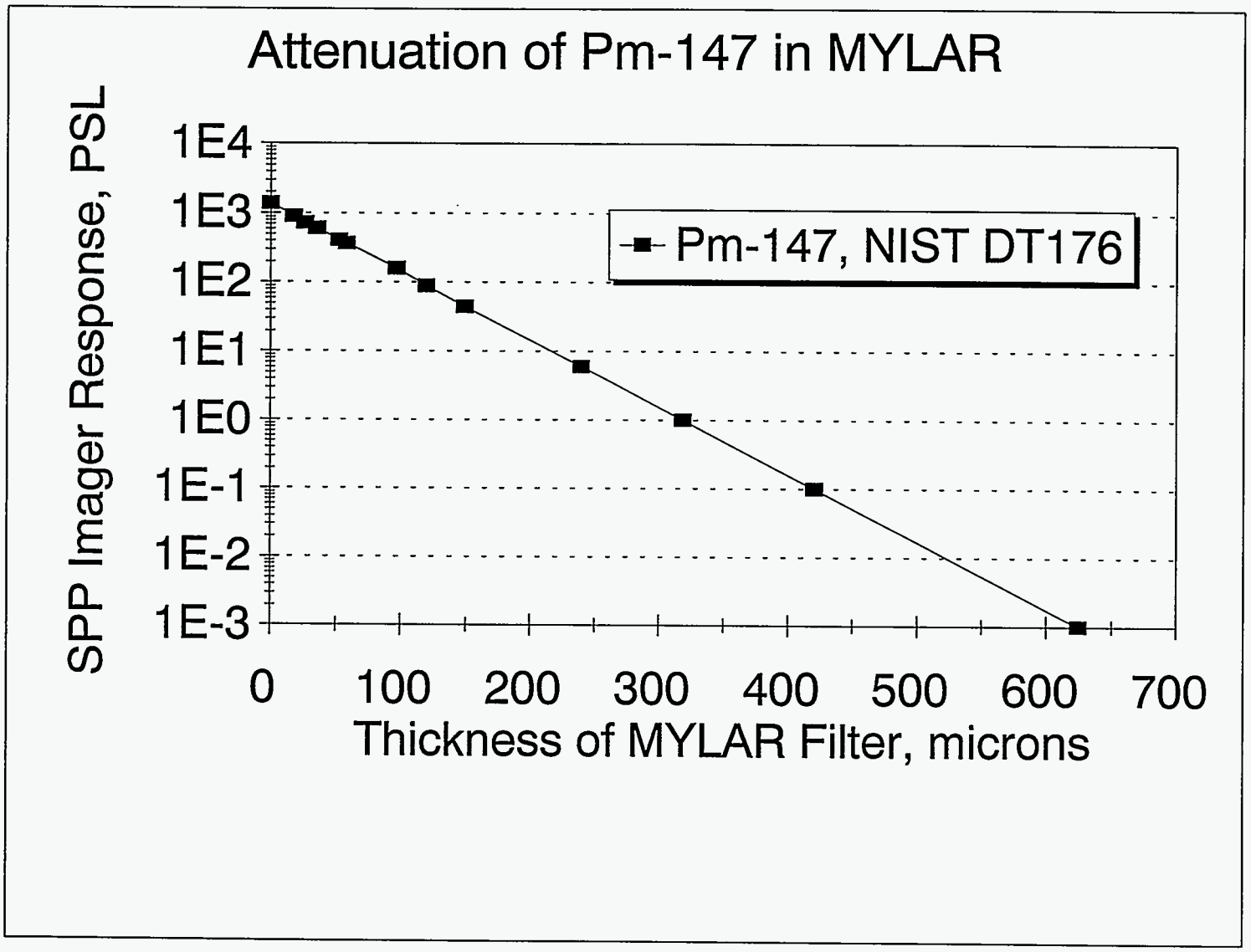

Figure 9. **Attenuation of ^{147}Pm radioactivity in MYLAR.**

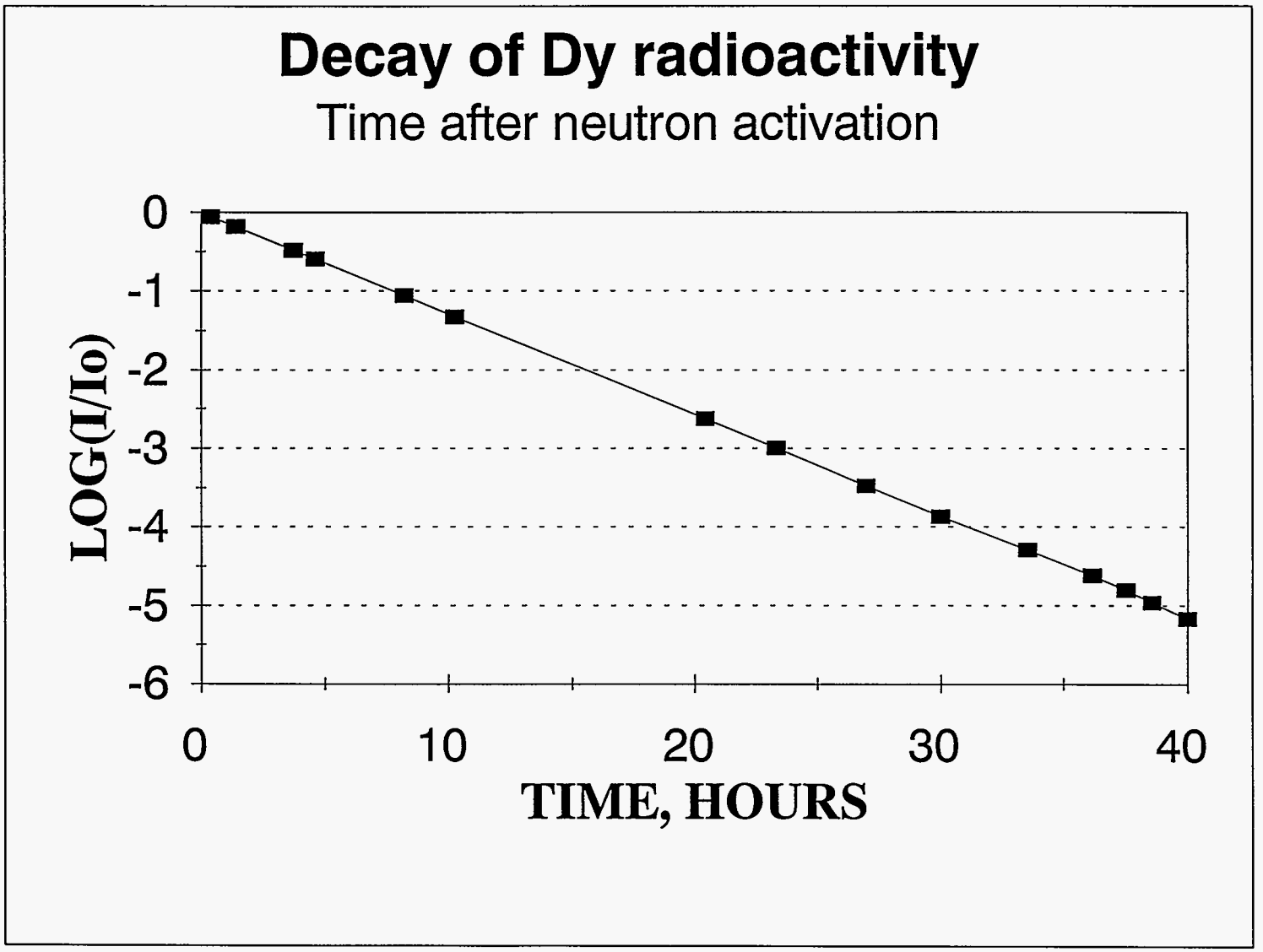

Figure 10. **The decay of a neutron-activated dysprosium foil monitored with a SPP sensor plate.**

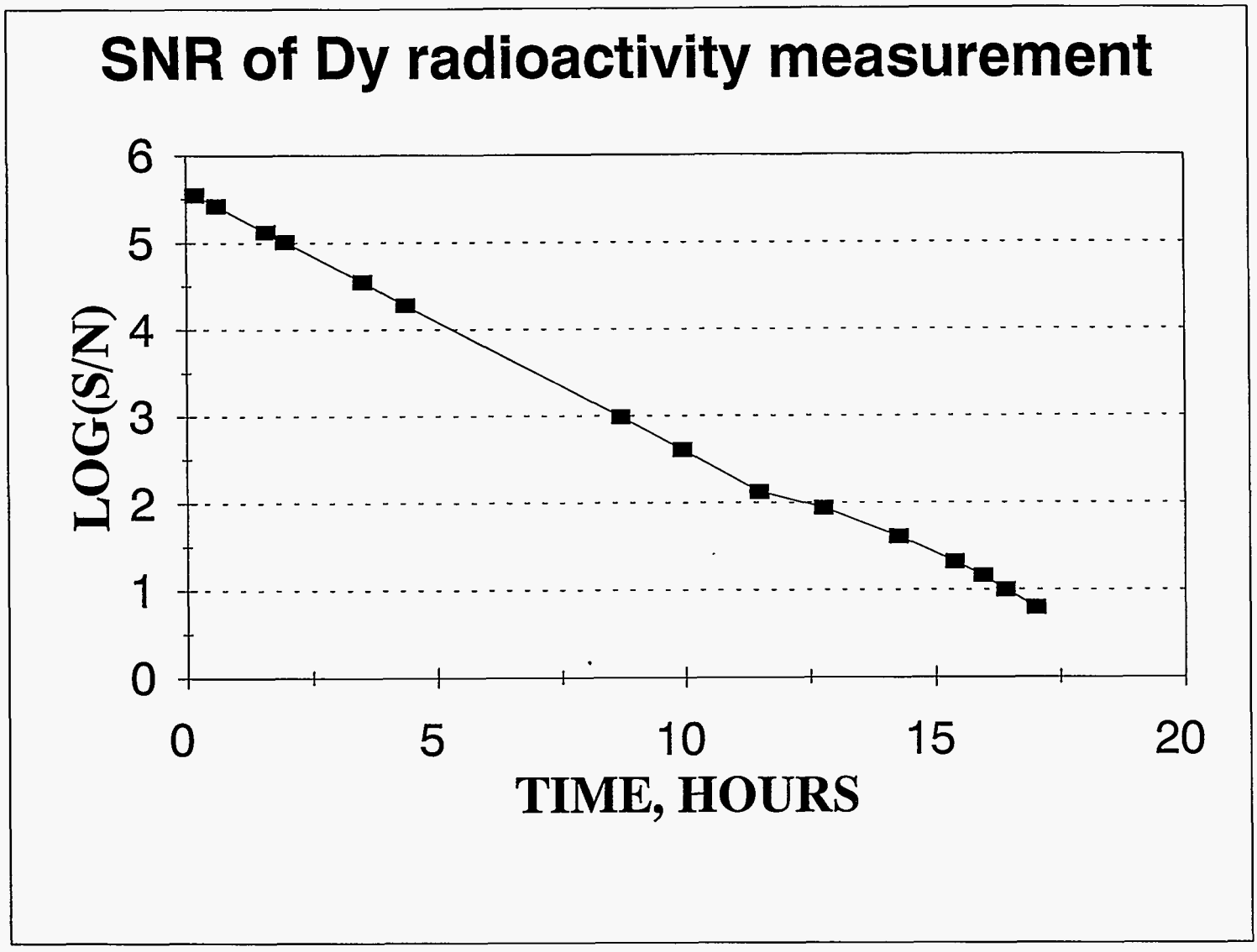

Figure 11. **The radioactivity signal-to-noise ratio as measured with a SPP sensor of a neutron-activated dysprosium foil at different half-lives after activation.**

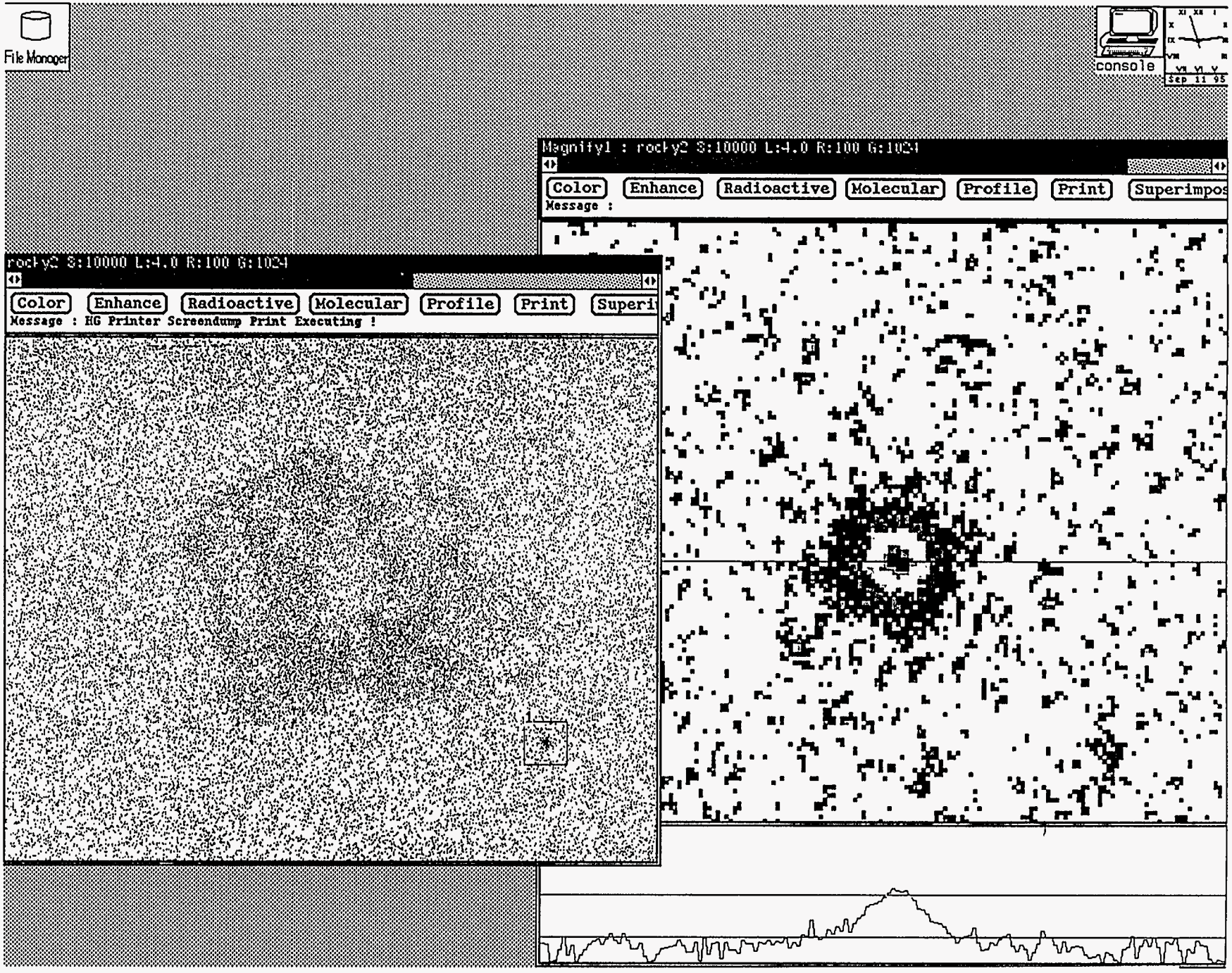

Figure 12. 10-hour exposure image of a sample taken from NIST SRM 4353 on a SPP sensor.

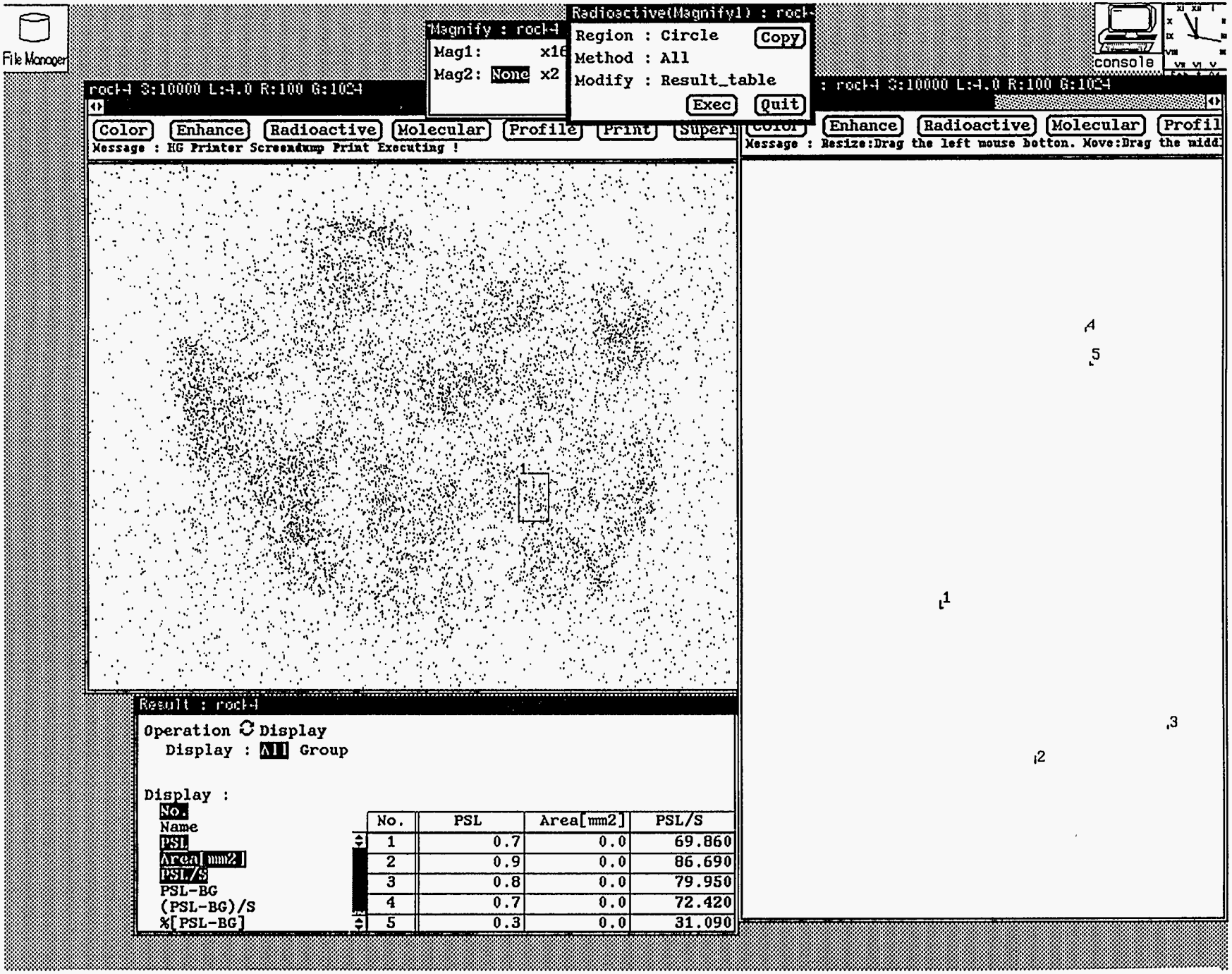

Figure 13. 10-hour exposure image of a second sample taken from NIST SRM 4353 on a SPP sensor.