THE HOLE-COUNT TEST REVISITED: EFFECTS OF TEST SPECIMEN THICKNESS

C. E. Lyman, D. W. Ackland, D. B. Williams, and J. I. Goldstein

For historical reasons the hole count, an important performance test for the analytical electron microscope (AEM), is somewhat arbitrary yielding different numbers for different investigators. This was not a problem a decade ago when AEM specimens were often bathed with large fluxes of stray electrons and hard x rays. At that time the presence or absence of a thick Pt second condenser (C2) aperture could be detected by a simple comparison of the x-ray spectrum taken "somewhere in the hole" with a spectrum collected on a "typical thickness" of Mo or Ag foil.2-4 A high hole count of about 10-20% indicated that the electron column needed modifications; whereas a hole count of 1-2% was accepted for most AEM work. Because quantitative x-ray measurements in the AEM are more precise than 10 years ago, "good" values for the hole count on Ag foils are now well below 1%. The absolute level of the hole count is a function of test specimen atomic number, overall specimen shape, and thin-foil thickness. In order that equivalent results may be obtained for any AEM in any laboratory in the world, this test must become standardized.

The hole-count test we seek must be as simple and as nonsubjective as the graphite 0.344nm lattice-line-resolution test.5 This lattice-resolution test spurred manufacturers to improve the image resolution of the TEM significantly in the 1970s and led to the even more stringent resolution tests of today. A similar phenomenon for AEM instruments would be welcome.

The hole-count test can also indicate whether the spurious x-ray signal is generated by high-energy continuum x rays (bremsstrahlung) generated in the electron column (high K-line to L-line ratio) or uncollimated electrons passing through or around the C2 aperture (low K/L ratio).1-6 This diagnostic aspect of the hole-count test is useful for the detection and elimination of the sources of hard x rays in intermediate-voltage (300-400kV) analytical electron microscopes.

This paper discusses the essential parameters for a standardized test of spurious x rays and proposed a candidate test specimen. This new test specimen reduces the experimental variation in the hole-count test caused by specimen thickness effects.

Standardizing the Hole-count Test

The spurious x rays detected in the hole-count test are caused by hard x rays and/or uncollimated electrons generating x-ray photons from regions of the specimen not directly under the electron beam. The most likely material generating these spurious x rays is the thick rim of the conventional electropolished or milled specimen. To make a precise test for this high-energy bremsstrahlung (if present), the test specimen must generate large numbers of spurious characteristic photons from the thick specimen rim when the beam is placed in the hole. This spurious characteristic x-ray intensity depends on (a) electron optical parameters such as probe current, (b) atomic number of the test specimen, (c) specimen rim thickness, and (d) the shape profile of the thinned area including the size of the hole. Since it is difficult to measure the absolute intensity of x rays detected when the beam is placed in the hole, the hole count is usually expressed as a percentage of the x rays generated.

![FIG. 1.---Schematic cross-sectional diagram of typical disk-shaped Mo hole-count specimen. Specimen rim provides greatest volume of material for production of spurious x rays by bremsstrahlung from upper column. Mo K-line x-ray intensity collected when beam is in the hole (electron a) is normally ratioed to that from some "typical thickness" of foil. Tests on Mo specimen of this type showed that hole count can be several times larger if x rays generated by electron b are used in this ratio rather than those generated by electron c. (Not drawn to scale.)](image)

The authors are with the Department of Materials Science and Engineering, Lehigh University, Bethlehem, PA 18015. The authors thank the staff of the Electron Optics Group of Philips Electronic Instruments, especially T. Boden, J. Fahy, M. Otten, M. N. Thompson, and J. van der Jeijden, for discussions concerning the universal hole-count test and assistance with some of the data shown. The authors are also grateful to J. R. Michael of Bethlehem Steel Corp. for helpful discussions and for use of the Vacuum Generators HB501. The Cr film on Ag specimen was fabricated and tested by M. Lyman. The support of DOE through grant DE-FG02-86ER45269 and of NASA through grant NAG 9-45 is gratefully acknowledged.
ated when the beam is placed on the foil itself at a "typical specimen thickness." It is this specimen thickness that makes the test arbitrary since this "typical" thickness may vary with each specimen and with each investigator. Thus, the most important variables that must be specified are the thickness of the specimen rim and the specimen thickness used for the "on foil" measurement, as shown in Fig. 1.

Electron Optical Parameters

The reappearance of the hole count as an issue in AEM performance is a consequence of the higher electron accelerating voltages of modern AEMs. Since 300-400kV electrons can generate considerably more high-energy bremsstrahlung (typically 20-200 keV) at the C2 aperture than 100kV electrons, the electron column modifications necessary to produce an adequately low hole count may prove to be more difficult than those employed at 100 kV. At 300-400 kV, hole counts that might appear adequate for AEM analysis of Fe-based samples may indicate serious problems for the analysis of Pt/Al2O3 catalyst specimens because high-energy bremsstrahlung excites Pt much more efficiently than Fe.

A major electron-optical parameter to be controlled at any accelerating voltage is the beam current. The hole-count test should be conducted with the same emission current and the same electron probe current each time. One may achieve this by measuring the current before each test with a Faraday cup and a picoammeter. Since many microscopes are not yet equipped with these devices, a secondary measurement such as screen current may be used if properly calibrated by the microscope manufacturer. Although the absolute level of probe current is not very important, this variable must be held constant for comparable tests.

Another important electron-beam parameter is the setting of the first condenser lens. Since the probe will be placed well into the hole, the actual spot size is not important. However, the first condenser lens also governs the extent to which the beam impinges on the second condenser aperture and the liner tube in the upper column. The hole-count test should be performed with the same setting of the C1 lens. In this case the probe tails, if present, may be sensed. Correct aperturing at the C2 lens is imperative for adequate performance in this test.

Test Specimen Material

The intensity of a characteristic x-ray line excited by electrons is given by

$$I = \frac{i(V - V_K)}{e^2}$$

where $i$ is the electron beam current, $V$ is the electron accelerating voltage, and $V_K$ is the excitation potential for the element of the test specimen. However, in most situations encountered with 300-400kV microscopes, the spurious x rays are generated by high-energy bremsstrahlung. For these cases, the intensity of the spurious characteristic x rays follows the mass absorption coefficient. The expression to predict the variation of $(\mu/\rho)$ between absorption edges is

$$\left(\frac{\mu}{\rho}\right) = 2Z^\lambda,$$

where $Z$ is the atomic number of the absorber (in this case, the test specimen) and $\lambda$ is the wavelength of the radiation exciting the absorber. For example, under the same test conditions in a Philips EM400T at 120 kV, Ni ($Z = 28$), Mo (42), and Ag (47), thin-foil specimens exhibited K-line hole counts of 3%, 6%, and 9%, respectively. Thus, the test specimen should be made of high-Z material for the generation of a high intensity of spurious characteristic photons at any fluorescing bremsstrahlung wavelength if the bremsstrahlung intensity is significant. This insures that if spurious x rays are generated, the test specimen will allow their detection.

If both a K and an L line from the specimen are to be detected with a Si(Li) EDS detector, the energies of both lines should be within the high-efficiency range for this type of detector: about 20 keV. Molybdenum with an L-series at 2.3 keV and a Kα line at 17.4 keV is an obvious choice. Mo has the additional advantage that most EDS systems are usually set to the energy range 0-20 keV for normal analysis. One important use of intrinsic Ge detectors in the AEM may be to determine whether the specimen stage is free of 20-200 keV bremsstrahlung that will fluoresce heavy elements in an analysis sample. These high-energy x rays would not be detected with a Si(Li) detector. Considering the desirability of a single test specimen for both Si(Li) and Ge detectors, the original proposal of silver as a hole-count test specimen still appears attractive, even though the x-ray spectrometer must be operated over the energy range 0-40 keV, which is not normally used for analysis with Si(Li) detectors (Ag Kα at 3.0 keV and Ag Kβ at 22 keV). Intrinsic Ge detectors, with their wider range of efficiency (roughly 2-100
keV), make even higher-atomic-elements suitable as test specimens, such as gold ($\alpha$ at 2.1 keV; $\beta$ at 9.7 keV; and $\gamma_1$ at 68.7 keV).

**Test Specimen Shape**

The shape of the test specimen has a significant effect on the efficiency of spurious x-ray generation and is probably responsible for much of the lab-to-lab variation in hole counts. Ideally, the test specimen should have a rim as thick as possible. A thick rim maximized the amount of spurious x-ray generation. For thin specimens with rim thicknesses of 80 and 120 μm, the hole counts were found to be 2.6% and 5.9%, respectively (Philips EM400T at 120 kV). Thus, the 100μm-thick specimen of an electropolished foil would be more useful than the somewhat thinner rim usually employed for ion-beam-milled specimens in order to reduce milling time.

A specimen of well-controlled shape can be fashioned with standard techniques by cutting disks from 100μm-thick Mo sheet, dimple grinding (15mm wheel) each side to a few micrometers total thickness, and finally ion-beam milling. But the time and effort to make such specimens and their fragility mitigate against their widespread acceptance as a standard test specimen. Even with such a carefully fabricated specimen, the "typical" thin-foil area used to obtain the denominator in the hole-count ratio usually exhibits a range of thicknesses from 50 to 500 nm, depending on the material. With the old hole-count tests, counts on the specimen were collected until 10,000 counts were obtained in the "typical" foil thickness. If the on-foil x-ray count is taken in a thick region of foil, the hole-count ratio will be lower than if it is taken on a thin region. Some method (such as direct thickness measurement or detection of the onset of Kikuchi lines) must be used to indicate a repeatable foil thickness for this test. The alternative to this cumbersome procedure is to use a specimen with a thin foil of constant thickness, as discussed below.

**Specimen Environment**

Components of the microscope surrounding the specimen also can generate spurious x-rays. Many AEMs based on the TEM column have cold fingers and objective aperture mechanisms located within a few millimeters of the specimen which may contribute to the overall extraneous background. Different lens designs also result in different amounts of metal around the specimen. For example, hole counts measured on the same specimen in a symmetrical condenser-objective lens (Philips EM400T) and in an unsymmetrical top entry lens on a cooled column (Vacuum Generators HB-501) were 5.9% and 0.25%, respectively. The low hole count in the VG HB-501 may result partly from the use of a wide second bore for the objective lens (less metal near the specimen).

**X-ray Counting Problems**

The detection of low x-ray fluxes when the beam is placed in the hole can be difficult with some x-ray spectrometers. For example, if very few x-rays are detected when the beam is in the hole, the spectrometer should faithfully record the exact number of photons entering the detector. In some situations, the setting of the fast discriminator or other deadtime circuit parameters can affect the number of x-rays apparently detected when the count rate is very low. A standard specimen with the potential for generating large intensities of spurious x-rays should relieve this problem.

**K-to-L-line Ratios from In-hole Spectra**

If the K-to-L-line intensity ratio from the hole is larger than that obtained when the beam is placed on the specimen, the spurious x-rays may be assumed to be generated by high-energy bremsstrahlung. Obviously, it is this component of spurious radiation that is of most concern for 300-400kV instruments. Figure 2 shows an example of this type of analysis for 120 and 300 kV. The 120kV data of AEM #1 show that the K/L ratio in the hole and on the specimen are essentially identical, as might be expected for an electron-generated hole count. The 300kV data show a K/L ratio about 20 times higher for AEM #2 and 4 times higher for AEM #3. These data indicate that the design of the electron column should be improved at 300 kV to reduce this excessive level of high-energy bremsstrahlung impinging on the specimen.

![Figure 2](image)

**FIG. 2.--Molybdenum K-line to L-line ratio integrated intensity ratios from "in-hole" spectra (solid symbols) and "on-specimen" spectra (open symbols) vs hole count for three AEMs. Higher hole counts for AEM #1 (triangles) and AEM #2 (squares) represent measurements made on thinner regions of foil.**

**Proposed Test Specimen for Spurious X-rays**

Since the main goal of this test specimen is to generate large numbers of spurious photons (when a bremsstrahlung source is present),
there is no reason to employ a specimen of conventional shape. A new hole-count test specimen might consist of an Ag support grid covered with a uniform thickness of Cr sputtered onto a thin film of carbon (Fig. 3). Holes in the Cr film or open-grid squares not covered by Cr may be used to collect the "in-hole" count, and the constant-thickness Cr layer allows a consistent "on-specimen" x-ray intensity measurement.

Preliminary data from a Cr film of unknown thickness on a 400-mesh Ag grid are encouraging. With this specimen in a low-background holder of the Philips EM 430T (200 kV, STEM mode, spot size 5, 0.5 nA probe current, 100s clocktime, 10 000 counts in Cr Kα) the ratio (converted to %) of the Ag Kα line (FWHM) collected in the hole to the Cr Kα line (FWHM) collected on the Cr film ranged from 1.9% to 2.4% and averaged 2.1%. These seven trials included data from three different Cr-covered grid squares and three different open-grid squares. This 24% variation in hole count should be compared with the 100-300% variation in hole count measured when the test was performed with different foil thicknesses in a Mo disk-shaped specimen. The conditions quoted above must not be considered a standard procedure for the new hole-count test. The hole count values measured above with the new test specimen cannot be related to traditional hole count values at the present time. Standard test conditions and acceptable hole count values with this test will emerge in the near future.

The major advantage of this type of specimen is the consistency with which the test can be performed. Since all the thin Cr areas have the same thickness, there should be no significant variability in the test between different operators. This type of specimen should be easy to fabricate in large numbers at reasonable cost.

![FIG. 3.--Schematic diagram of proposed hole-count test specimen consisting of silver grid supporting thin film of chromium on carbon.](image)

This test specimen has additional advantages in that, once the hole count is determined to be acceptably low, other standardized tests may also be performed on the same specimen. These include tests of Cr peak-to-background, estimated minimum detectability of Cr, and comparative x-ray detector collection efficiency.

Spurious X Rays in Relation to Experimental Analysis Data

The above Ag/Cr spurious x-ray test, like the older hole-count test, gives an important figure of merit for an AEM. Of course, the values to be quoted as acceptable for this "Ag/Cr spurious x-ray test" will be different from those quoted in the old hole-count test. The desired value will again be 0%. Acceptable values for quantitative analysis will depend on the type of sample analyzed. The criterion to be used is whether the characteristic peak of interest, detected when the beam is placed in the hole, is well below the electron-generated background from the analysis area under the beam. This "hole count from the analysis specimen" is a small spectrum related to the average composition of the entire specimen which is added to each individual spectrum taken in regions localized by the focused electron beam. Subtraction of this unwanted signal from the analysis peak is often suggested. However, progress toward use of the AEM as an "electron nanoprobe" for thin specimens, with precision and accuracy approaching that of the "electron microprobe," demands that all extraneous sources of x rays be eliminated at their sources.

Conclusion

A hole-count test specimen consisting of a thin film of chromium over a silver support grid has been shown to have significantly better repeatability than previous specimens used for this test. Whereas the old test was prone to variations both in rim thickness and in the foil thickness selected for the "on-specimen" spectrum, the thicknesses of the Ag grid and the Cr film in this new specimen are nearly constant—eliminating these sources of error. When a standardized test specimen of this type is available in reasonable quantity, further progress can be made toward the reduction of spurious x-ray background in the AEM.

References

8. N. J. Long, described at the 1981 VG Microscopes users' group meeting.

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