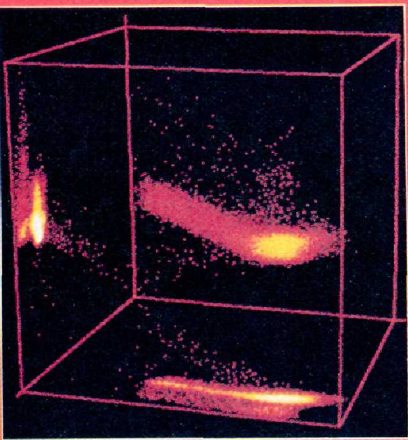

Concentration Histogram Imaging

A Scatter Diagram Technique for Viewing Two or Three Related Images

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In order to characterize the chemistry of materials on a fine scale, we employ compositional mapping techniques. Quantitative compositional mapping produces images that depict the concentrations of elemental and/or molecular constituents of a specimen on a spatial scale of ~ 1 μm or less (*1*). The images can be created from a variety of microanalytical instruments: for example, the electron probe microanalyzer, analytical electron microscope, ion microprobe, ion microscope, and laser Raman microprobe. A common problem is that much of the quantitative concentration information is lost when numerical concentration information is presented in the form of an ordinary gray scale or color scale image. Ideally, we wish to simultaneously present both spatially resolved information and numerical concentration data, often for two or more constituents.

A/C INTERFACE

For one constituent, techniques are available that satisfy the requirements of spatial and numerical display. Depiction of positional information as an image is straightforward—the concentration data are encoded with one of a variety of gray or color scales. Gray scale representation coupled with image processing for selective enhancement provides excellent display of compositional contrast, but it is difficult for an observer to relate specific gray levels to particular numerical concentration values. The human visual process is better adapted to viewing colors. The thermal color scale, which uses the sequence of colors emitted from a black body upon heating (deep red through cherry, orange, and yellow to white), provides a "logical" color scale in which particular colors can be more easily recognized and the approximate corresponding numerical value identified.

When two or more constituents are to be presented simultaneously, the

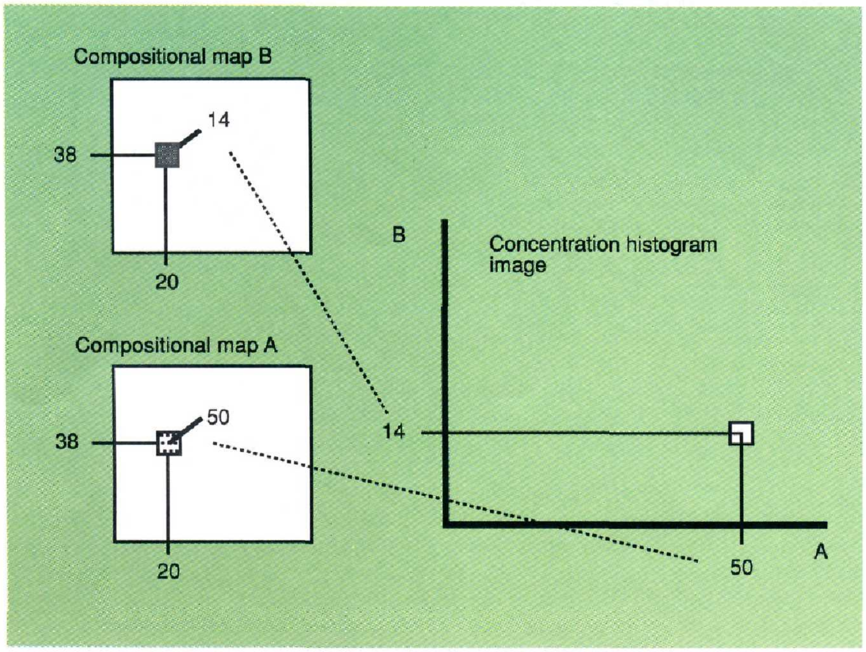

Figure 1. Schematic for construction of a CHI.
Pixels in location (20,38) have intensities 50 and 14. Histogram bin (50,14) is incremented for this image location.

thermal scale cannot be applied. A partial solution to this display problem is to use primary color overlays in which the individual constituent images are superimposed, with each applied as a primary color (i.e., a separate constituent is applied to the red, green, and blue channels of the video display). Spatial coincidence of two constituents is revealed by the appearance of secondary colors; however, information on relative amounts is lost. In principle, color scales with varying intensity can be established to depict relative amounts, but in practice such scales are difficult to use and are even more difficult to transfer to printed media.

To overcome these limitations, we have adapted the technique of scatter diagrams and have developed a method in which the compositional maps are transformed from spatial to concentration dimensions. We call this transformation the Concentration Histogram Image (CHI) (*2, 3*).

The CHI is much like a scatter diagram or bivariate histogram, which is more informative than a one-dimensional intensity histogram (*4–6*). Unlike the scatter diagram, the CHI is displayed as an image by encoding the frequency information with the thermal color scale. Also, the image or pixel representation of the CHI lends itself to a reverse operation called the "traceback" function (explained in detail below), which enables clearer rendering of both two- and three-dimensional information.

The CHI provides a direct numerical view of the concentration or intensity relationships between two or three constituents. The traceback algorithm permits rapid determination of the correspondence between features in the numerical display with specific places in the original compositional or intensity maps. CHIs complement the normal primary color overlay composites and are useful for evaluating data, diagnosing experimental problems, and detecting relationships of concentrations. A similar image analysis technique—using scatter diagrams but without traceback—has been applied by Prutton et al. (*7*) to intensity maps generated by Auger electron spectroscopy.

Producing the CHI

Although the CHI is most often applied to images representing concentrations of analyzed constituents, the technique can be applied equally well to any two or three registered images, as it is on our image processing systems (*8*). Consider an $n \times n$ pixel image for constituent A, represented by an array with elements $a(x,y)$, each giving a concentration or intensity value for A at location x,y. Also consider a similar image for constituent B with elements $b(x,y)$ that is in pixel by pixel registration with image A. The CHI is constructed by incrementing an element of the array $h(i,j)$ where i and j correspond to the values a and b of concentration or intensity for every location (x,y) in the images. A FORTRAN code for this procedure is listed in the box on p. 247 A.

Figure 1 shows this procedure schematically. After the CHI array is calculated, it is displayed as an image (described below). Apart from visualization of the array, this procedure is like the construction of a two-dimensional

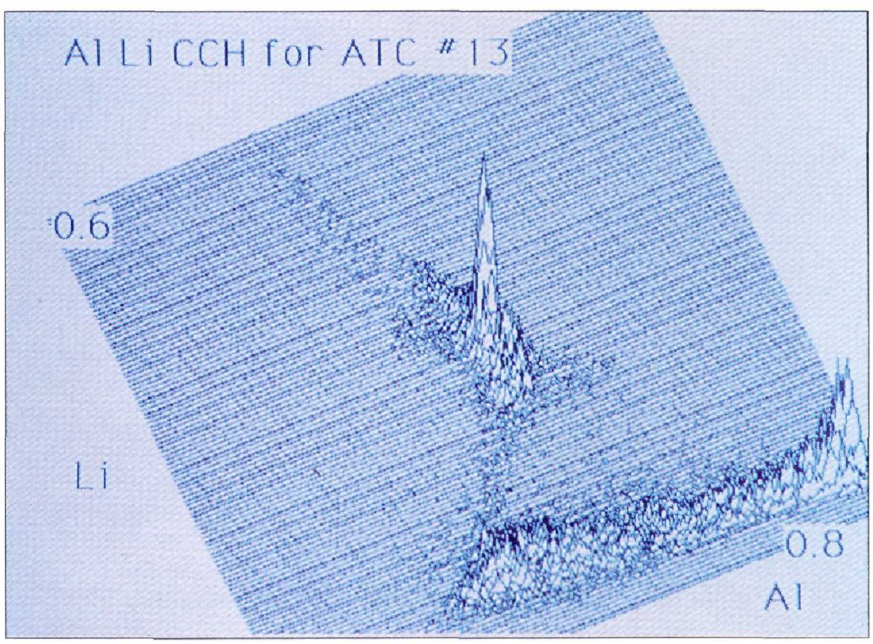

Figure 2. CHI of ion microscope compositional maps of an aluminum–lithium alloy plotted as a mesh plot.
(Specimen courtesy K. Soni and D. Williams, Lehigh University.)

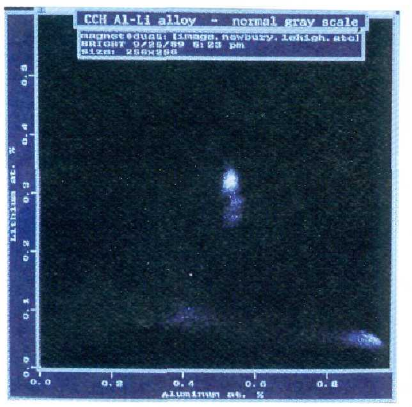

Figure 3. The same CHI as Figure 2, shown as a gray scale image.

or bivariate histogram—or like the plotting of a scatter diagram with limited precision—that is, with clumping the data into bins.

Displaying the CHI

The CHI can be displayed as a mesh plot, in the way bivariate histograms are usually plotted (Figure 2), showing intensity along the vertical axis. However, because the CHI is an array, similar to the images from which it was made, it is natural to display it also as an image (Figure 3).

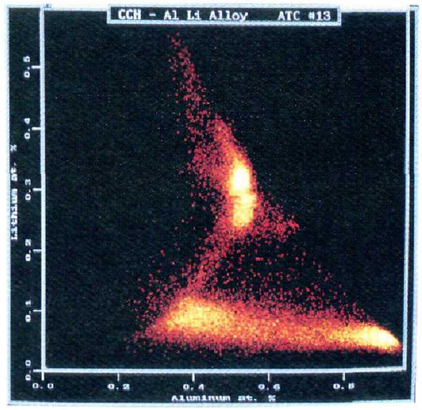

Figure 4. The same CHI as Figures 2 and 3, shown as an image with the thermal pseudocolor scale: 0—black, 1—red, 2–255—dark orange–yellow–white.

When displayed with a gray scale, bins with a few counts are too dim to be seen against the background. Because these more diffuse regions may be as interesting as the more compact and intense regions (higher counts), we can overcome the limitation of the gray

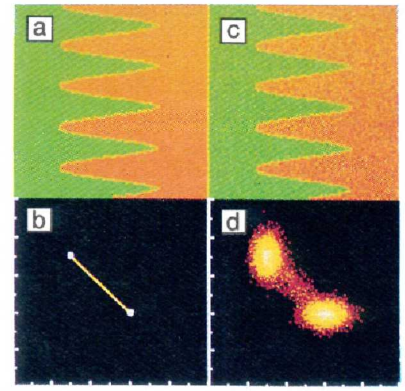

Figure 5. Simulated microprobe maps (a and c) and their corresponding CHIs (b and d).

The maps are shown as color overlays of one image in red and the other in green. Each map has two phases or regions separated by the wavy line. Each region in (a) has a constant amount of the red and the green elements—the color is determined by the predominant element. The map–CHI pair (c) and (d) includes some noise to simulate counting statistics.

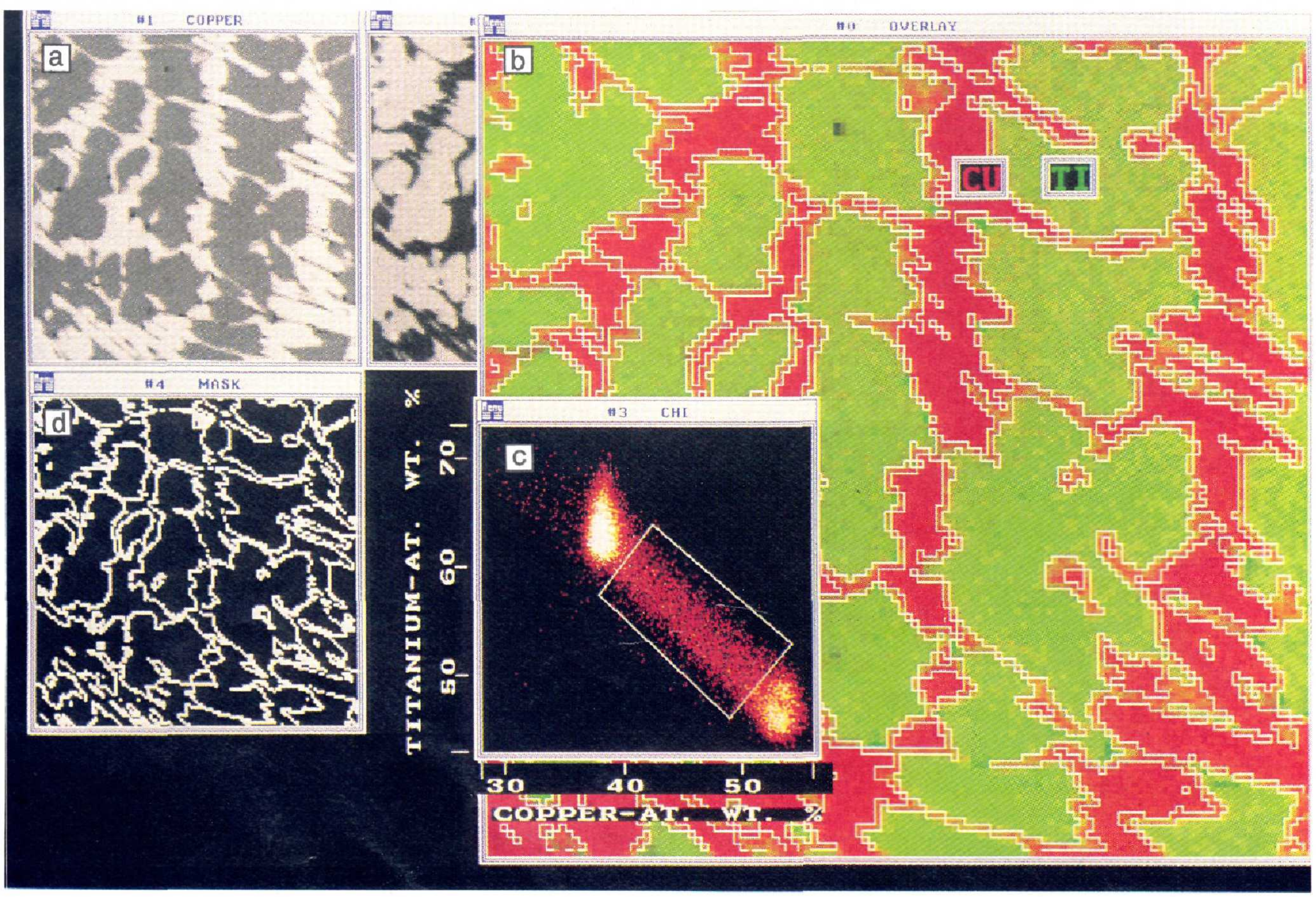

Figure 6. Electron probe maps and corresponding CHIs of a Cu–Ti alloy.

(a) Left: copper map, 128 × 128 pixels. Edge of field is 125 μm. Right: Ti map (partly hidden). (b) Color overlay: Cu is red, Ti is green. Edge of field is 125 μm. (c) CHI. (d) Traceback mask image using outlined area in CHI (bottom right). Selected pixels outlined in white in color overlay.

FORTRAN code for producing a CHI

```
do y = 1,n
do x = 1,n
   i = a(x,y)
   j = b(x,y)
   h(i,j) = h(i,j) + 1
end do
end do
```

FORTRAN code for the traceback function

```
do y = 1,n
do x = 1,n
   i = a(x,y)
   j = b(x,y)
   if (i,j) fall within the selected region
   of the CHI then
      m(x,y) = 1
   else
      m(x,y) = 0
   end if
end do
end do
```

scale by displaying the CHI with a thermal scale (Figure 4) constructed so that zero count is black, one count is red, and higher counts range from orange to yellow to white. In this way, the centers of the high-intensity clumps are visualized as well as the scattered bins with only one count each.

This display method also works well for higher dimensional CHIs. Although the mesh plot (Figure 2) shows, in a quantitative way, the relative counts per bin or relative areas in the original images, Figure 4 shows more clearly the relative positions and sizes of clumps and the individual bins with few counts. It is also easier to determine concentration values from Figure 4 using the axes, and to outline selected areas for applying the reverse transformation (traceback).

Properties of the CHI

To introduce some properties of the CHI, Figure 5 shows compositional X-ray maps simulated on the computer to specify the counting statistics. The first synthetic image consists of two "phases," with no variation in apparent concentration from counting statistics (Figure 5a). The two phases have a predominance of either the "red" or the "green" element and a wavy boundary between the two. A pixel through which the boundary passes has "concentrations" calculated from the relative fractional area of the pixel that is in either phase. The resulting CHI in Figure 5b has one point at each of the concentration values for the two phases and a line between the points corresponding to pixels on the boundary. The upper left point in Figure 5b is high in element green and corresponds to the green phase. Parts of the yellow line that are near this point represent boundary pixels that are mostly in the green phase. The map–CHI pair, Figures 5c and 5d, is the same as that on the left except that counting statistics have been added. The two points spread out into clumps, and the line spreads into a diffuse band (Figure 5d). This CHI looks very similar to actual experimentally measured systems of two phases (see Figure 6c).

Traceback

Having generated the CHI, the observer is likely to be interested in the relationship of features recognized in the CHI to features in the original maps. This relationship can be discovered with a modification of the CHI algorithm, which we call the traceback function. The traceback function selects pixels in the original images on the basis of intensity correlations; this is analogous to selecting pixels in a single image by setting an intensity threshold. This function is implemented by marking a mask image, $m(x,y)$ (Figure 6d), on the basis of regions previously selected from the CHI. A pixel in the mask is marked if the corresponding CHI bin lies inside the selected region of the CHI. The FORTRAN code for the traceback function appears in the box.

Mask m is a binary image that is in registration with images a and b and

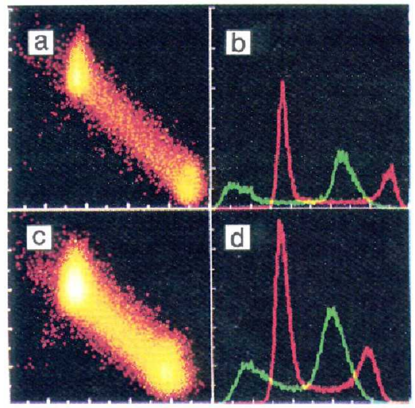

Figure 7. The effect of resolution on CHIs and one-dimensional histograms.
(a) CHI for the lower resolution (128 × 128 pixels) maps in Figure 6a. (b) One-dimensional histograms for these maps (colors in overlay and those of histograms correspond). (c,d) Maps of same sample area as top, taken at higher resolution (256 × 256 pixels). Cu—x axis on CHI and red plot: 44–98 atomic percent. Ti—y axis on CHI and green plot: 37–67 atomic percent.

that labels the pixels in both images as corresponding to the selected region of the CHI. The mask thus selected can be viewed as an image in its own right, or it can be used to mark the original maps.

Practical details

Scaling. Many images, especially images representing concentrations, do not have pixels with integer values. To construct a CHI, the pixels are therefore scaled to integers ranging from 0 to 255, or to a range matching the dimensions of the CHI array. If the images are originally integer images with a small range of values, then care must be taken when scaling to avoid stripes in the

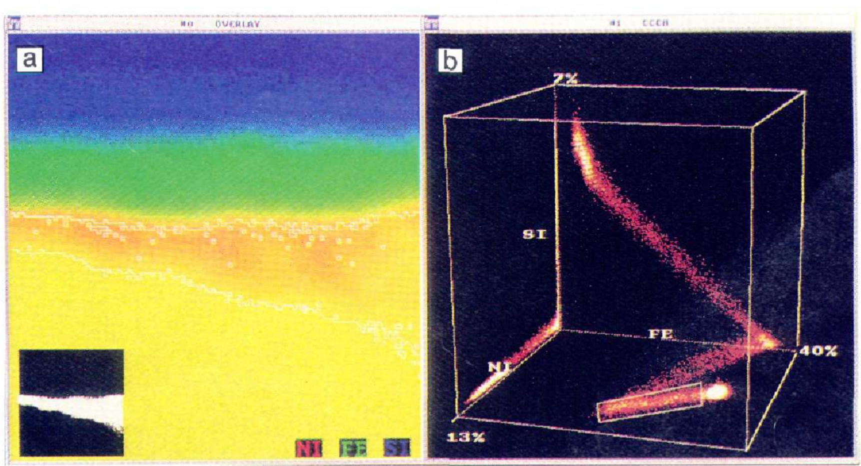

Figure 8. (a) Color overlay of three maps (Ni—red, Fe—green, Si—blue) of electron probe microanalyzer compositional maps of a diffusion zone at a metal–glass seal and (b) corresponding CHI.
Edge of field is 25 μm. Areas outlined in white on overlay correspond to outlined feature of CHI. Inset: traceback mask image. (Specimen courtesy J. Mecholsky, Jr., University of Florida.)

CHI caused by periodically missing bins or doubling counts in bins. If the intensity range of the image is on the order of, or smaller than, the number of available bins along a CHI axis, it is best to use the intensities directly or to scale the images with care.

Comparison of one- and two-dimensional histograms at two resolutions. Does the resolution at which the images are taken affect the appearance of the CHIs? Figure 6a shows Cu and Ti electron probe compositional maps (128×128 pixels) of a binary alloy and Figure 6b shows a color overlay with Cu in red and Ti in green. We often display corresponding images as a color overlay to examine the spatial relationships of the constituents. The color overlay shows a boundary between the two phases that has moderate concentrations of both Cu and Ti. Whether the boundary is a diffusion zone or an instrumental artifact caused by the finite width of each pixel and the finite electron interaction volume in which the X-rays are generated cannot be determined from either the images or the CHI. This is because the boundary is one pixel [1 μm] thick or less. The width of the interaction volume for the Cu–Ti alloy at 20 keV is about 1 μm.

Figures 7a and 7b show standard one-dimensional histograms and CHIs that correspond to maps in Figure 6a, whereas Figures 7c and 7d derive from maps of the same area that were taken at twice the spatial resolution (256×256 pixels). (The higher resolution maps are difficult to distinguish visually from the lower resolution maps and are not shown in Figure 6.) Because the higher resolution maps have four times the number of pixels, the corresponding CHI has four times the number of counts and appears broader, but the features are essentially the same. Note that with conventional one-dimensional histograms alone, it cannot be determined which phase in one histogram corresponds to a particular phase in the other histogram. The CHI shows this type of correspondence immediately.

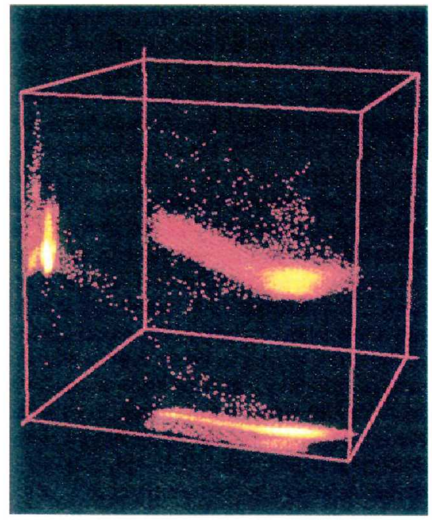

Figure 9. CHI for electron probe microanalyzer compositional maps of an Fe–Ni–Cr alloy with one major phase.
A secondary cluster is seen in front of the primary cluster in projections on the bottom and left face of cube; this secondary cluster is not visible from this viewpoint of the three-dimensional presentation within the cube.

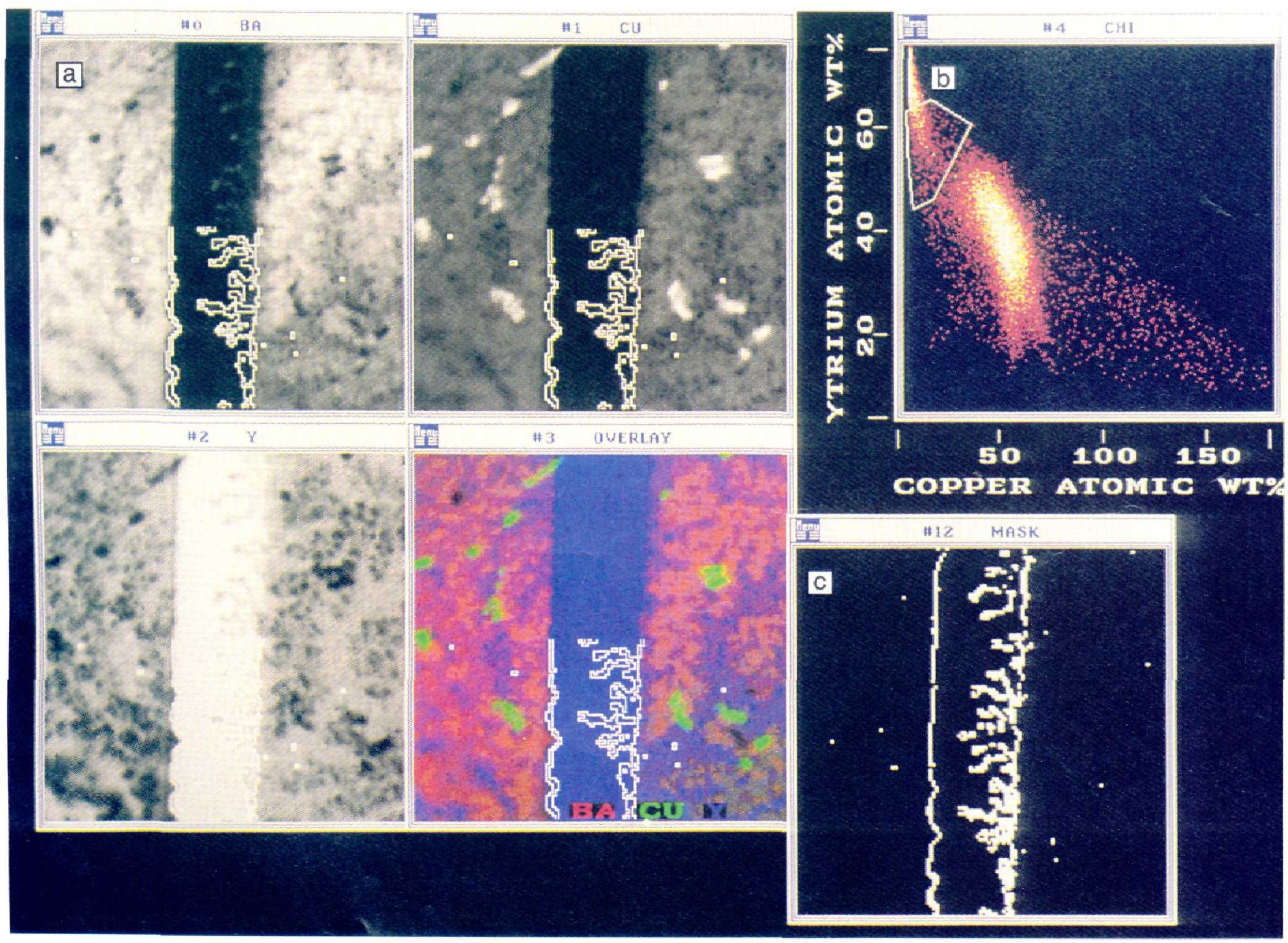

Figure 10. Element maps and the CHI for a small element of a superconducting ceramic integrated circuit.
(a) Ba, Cu, and Y maps and color overlay (labeled in banners). Center strip is conduction barrier. Image width is 125 μm. (b) CHI (Y vs. Cu) for two of the maps on the left. Outlined region corresponds to tendrils. (c) Traceback mask image corresponds to outlined region in CHI. Matching pixels are outlined in white in bottom half of maps and overlay. Top halves are not outlined to show tendrils better. (Specimen courtesy R. Ono, NIST.)

Three-dimensional CHIs. The pseudocolored display of the CHI lends itself directly to adding another dimension. Just as the two-dimensional histogram is plotted inside a square, the three-dimensional histogram is plotted inside a virtual cube and is rendered as a projection. To present three axes of concentration data, the only choice possible for the frequency information is color encoding. Figure 8a shows a color overlay for electron probe compositional maps of a diffusion zone at a metal–glass seal and the corresponding CHI is shown in Figure 8b. The CHI is snakelike in shape and is three-dimensional, taking two sharp turns.

When the histogram is viewed in projection, there is always a danger of missing some structure along the line of sight. Viewing the histogram from several angles helps, and presenting the CHI as a stereo pair (*9*) works even better, although the need for special optical devices to view the stereo pair makes this approach somewhat impractical. A practical way to view the histogram is to use one, two, or three ancillary projections onto the rear faces of the virtual cube. A projection on the left face is shown in Figure 8b, which makes it easier to see that the part of the snake ending near the bottom face is in fact lying on the bottom face and not heading upward (increasing in Si concentration), as might be mistakenly deduced without the projection. The white outlines and mask (inset) illustrate the traceback function (see below) for this feature.

Another example is given in Figure 9, which depicts a CHI of a set of maps for a Fe–Ni–Cr alloy. A small clump of points is visible in the projections on the bottom and left face of the cube but is hidden in the interior of the three-dimensional presentation within the cube.

A third possibility is to view a three-dimensional CHI in a display that permits real-time rotation of the plot. Such a display, when in motion, provides a feel for the shape of the plot and allows searching from arbitrary viewpoints for features of interest.

Examples of the traceback function

The traceback feature identifies which areas of the image correspond to which features of the CHI. As an illustration, Figure 10a shows Ba, Cu, and Y electron probe microanalyzer compositional maps and the color overlay for a superconducting thin-film circuit element. There are tendrils of Cu stretching across the vertical band in the center that is supposed to be Cu-free. These tendrils are not seen on the color overlay and are barely visible as depletions in the Y map. We wished to select the tendrils from the rest of the image. Inverting the Y map of Figure 10 shows the tendrils to a degree, but it also shows the large darker areas on both sides of the band.

From inspection of the maps, the tendrils were presumed to be high to intermediate in Y and low (but not zero) in Cu. The outline for such an area is shown in white on the CHI (Figure 10b), and the traceback is shown as a mask in Figure 10c and as outlines in Figure 10a. The tendrils are outlined along with pixels on the other side of the strip, which suggests a similar process at an earlier stage; this feature was missed in the previous figures.

Figure 11 shows the traceback applied to the projection of a CHI. Although (strictly speaking) one should outline parts of this three-dimensional object with a surface (or box), the use of

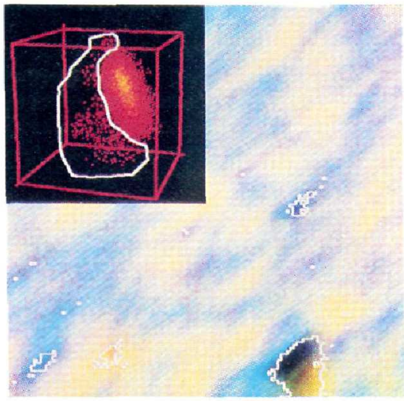

Figure 11. Traceback applied to the projection of a CHI.

Color overlay for Ba (red), Cu (green), and Y (blue) maps for superconducting ceramic thin film. White boundary surrounds regions corresponding to outlying points in CHI. Image width is 50 μm. Inset: CHI with white line surrounding outlying points. Ba—lower left axis, 21–38 atomic percent. Cu—lower right axis, 12–23 atomic percent. Y—vertical axis, 11–19 atomic percent.

the projection shown here often works well. This CHI is of a superconducting thin film and appears as an ellipsoid with outlying bins (outlined in white). The traceback function applied to most of the outlying bins is seen outlined in white on the color overlay and reveals discontinuities in the film.

The raw X-ray count maps measured with the electron probe microanalyzer require several steps in the calculation process to convert the X-ray counts into concentrations. Sometimes the changes on quantitation of the relative image intensities are subtle. A CHI corresponding to Figure 7a before the X-ray counts in the maps have been corrected for instrumental effects shows fragmentation of the two clumps. The traceback feature shows that the fragments correspond to the high Cu phase on the right side of the map in Figure 6b, suggesting that the clump fragmentation is caused by spectrometer defocusing (*10*). The secondary clump in Figure 9 similarly seems to be attributable to instrumental artifacts that have not been completely corrected.

Conclusion

The CHI is a useful diagnostic tool for examining compositional maps. This technique of image presentation is a useful adjunct to conventional images and is especially effective for detecting inhomogeneities in materials, recognizing and highlighting rare events, checking quantitative correction procedures needed to produce compositional maps, and evaluating sampling and measurements statistics. The CHI is also useful for selecting features of interest for further analysis.

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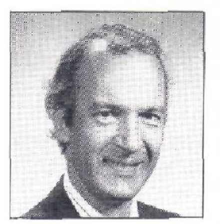

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