United States Patent

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[54] **INTERPRETATION METHOD FOR USE IN X-RAY DIFFRACTION AND X-RAY FLUORESCENSE ANALYSIS**

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[52] **U.S. Cl.**.....**33/1 C,** 33/107, 33/111

[51] **Int. Cl.**.....**B43l 7/00**

[58] **Field of Search**.....33/1 C, 1 HH, 1 B, 107, 111

[56] **References Cited**

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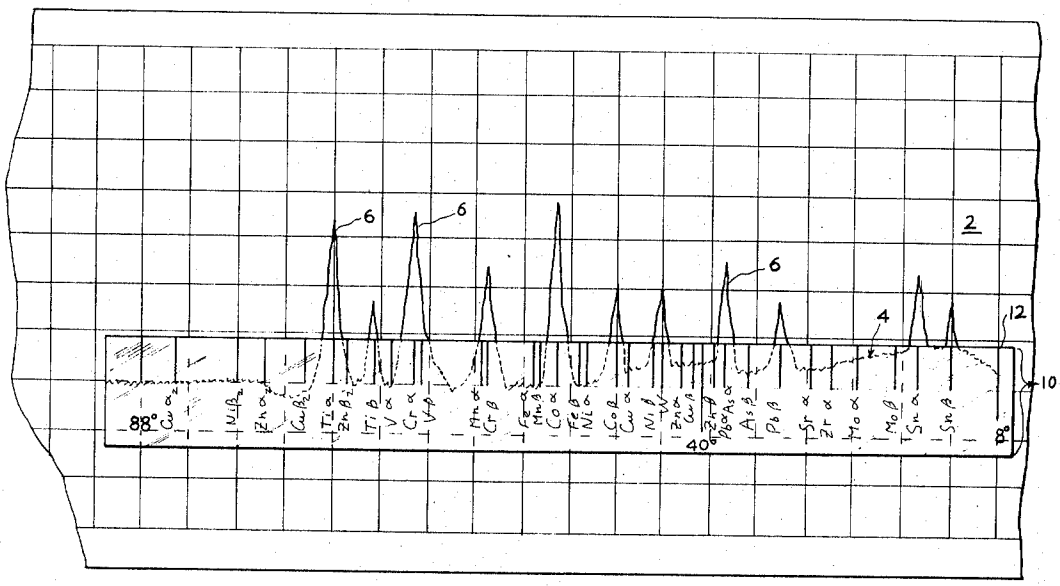
[57] **ABSTRACT**

A simple and efficient means is provided in X-ray fluorescence and X-ray diffraction for deriving interplanar spacings and wave lengths, or characteristic elements, determined from the data by the Bragg equation. The apparatus contemplates a data background sheet and an interpretive sheet. The data on the background sheet is in the form of a plot of either an X-ray diffraction pattern or an X-ray fluorescense pattern. The interpretive sheet is a transparent sheet superimposable over the data sheet with indicia imprinted in scale thereacross.

6 Claims, 2 Drawing Figures



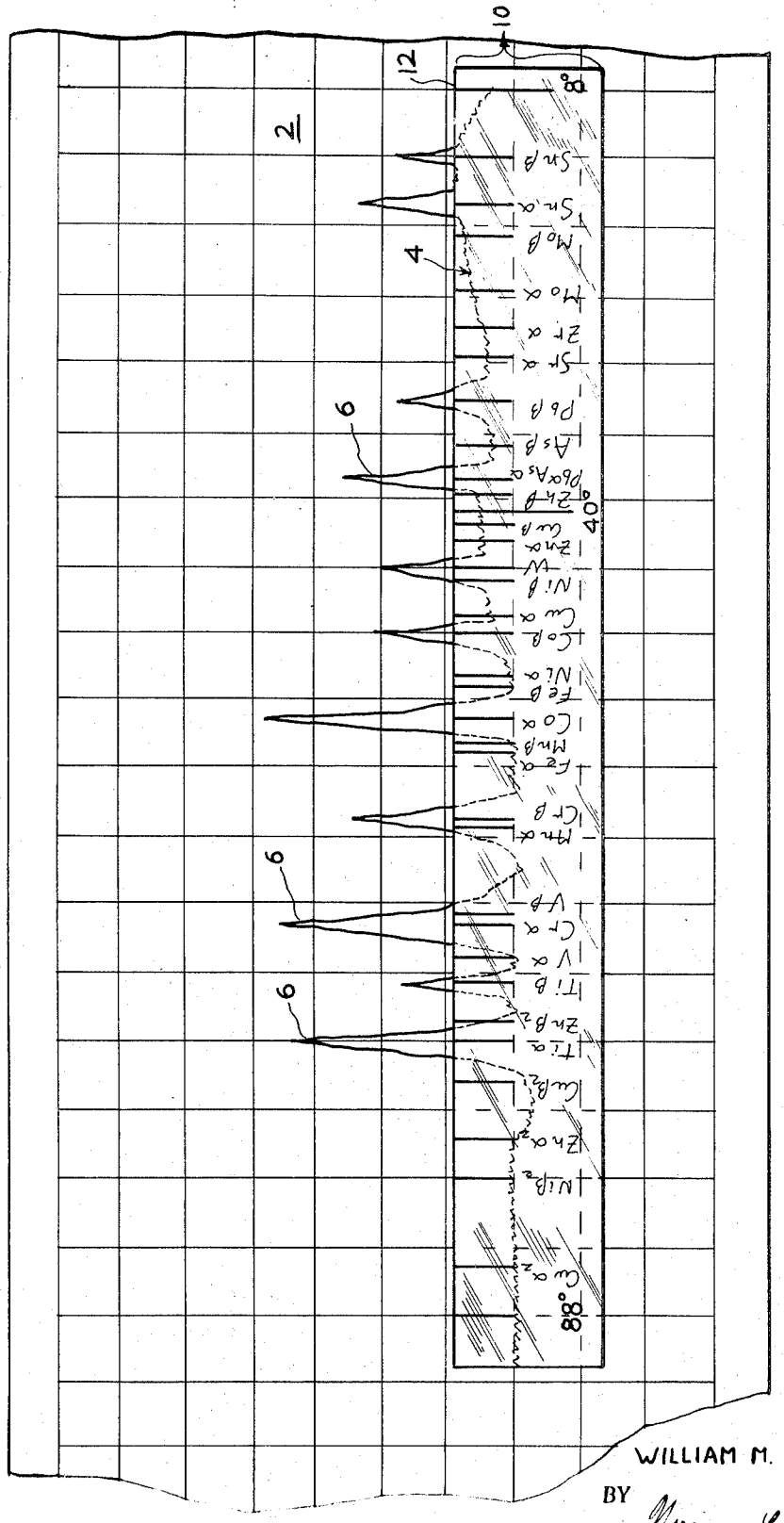


FIG. 1

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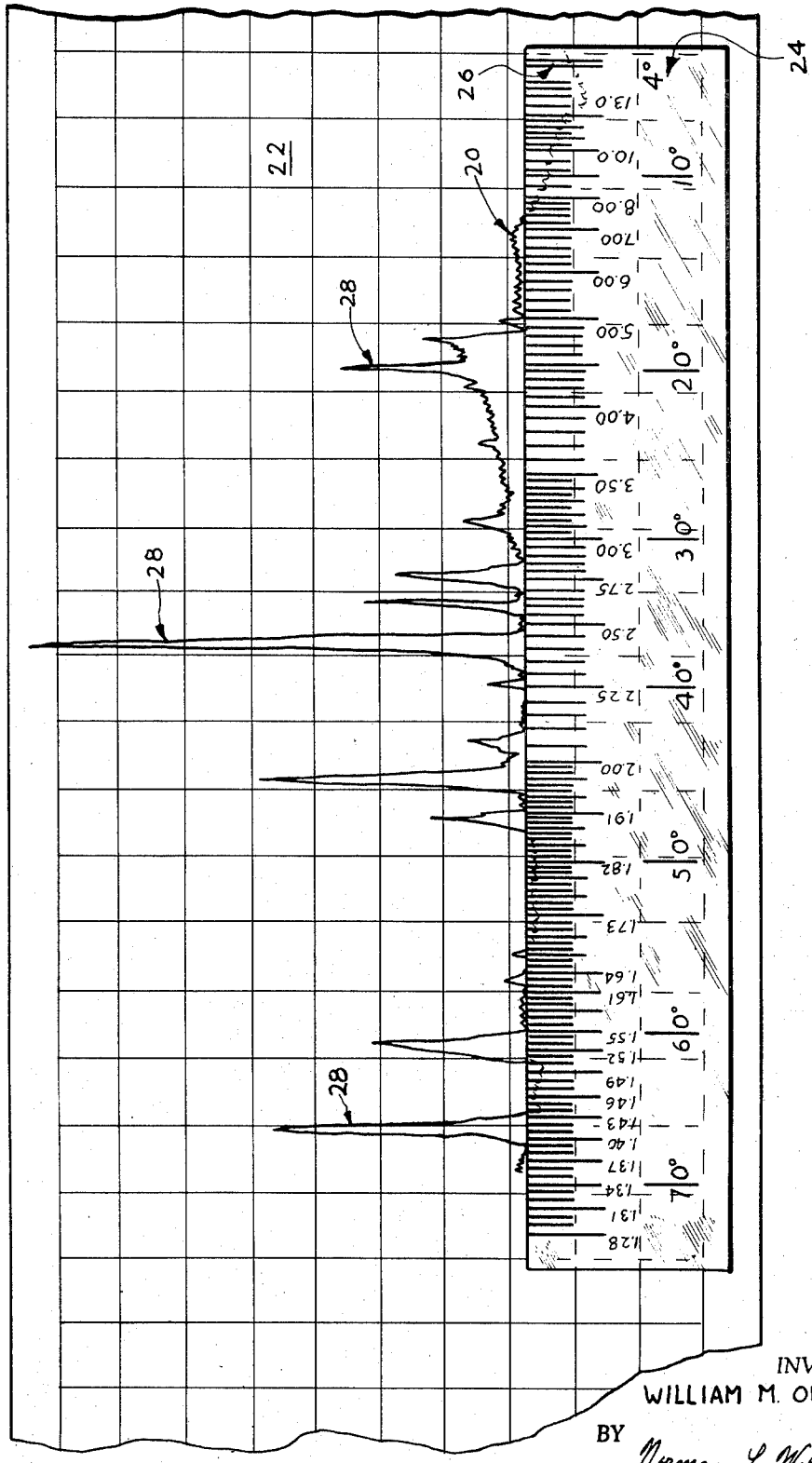

FIG. 2

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INTERPRETATION METHOD FOR USE IN X-RAY DIFFRACTION AND X-RAY FLUORESCENSE ANALYSIS

BACKGROUND OF THE INVENTION

This invention relates to interpretation apparatus for use in identifying unknown substances from X-ray diffraction and X-ray fluorescence data. The invention provides a simple and efficient means, in X-ray fluorescence and X-ray diffraction, for deriving interplanar spacings, and wave lengths, or their characteristic elements, determined by the Bragg equation.

The use of X-rays as a physical tool for chemical analysis has increased markedly over the last few years. Whereas the primary emission spectrum with the sample serving as the target of an X-ray tube is no longer frequently used in X-ray analysis, X-ray fluorescence and X-ray diffraction methods are widely employed. In X-ray fluorescence the primary X-rays emitted by the anticathode (target) are allowed to strike the sample to produce a secondary or fluorescent radiation. This radiation passes through a collimating slit onto a single crystal analyzer and goniometer and then to a detector.

It is apparent that the analyzer crystal plays an important role. Reflection by an analyzing crystal yields virtually monochromatic radiation. Analyzing crystals of mica, quartz and sodium chloride have particularly strong reflections. The fluorescent X-rays impinge upon this single crystal and the crystal acts as an analyzer or spectrometer and reflects the various wave lengths according to Bragg's law, $n\lambda=2d\text{Sin}\theta$ where n is a small integer giving the order of reflection, λ is the wave length of the fluorescence X-rays, d is the lattice spacing or spacing between the various planes of the crystal and θ the glancing angle of incidence. The crystal is rotated slowly and reflects each wave length, one at a time, at an angle 2θ. Thus each wave length from the specimen is reflected successively at separate discrete angles as the crystal, and goniometer, rotate. Since the analyzing crystal has a known d value, the various wave lengths are determined from the Bragg equation. It is necessary to select a crystal with the interplanar or d spacing small enough to provide adequate angular separation of spectral lines, usually 0.5 to 5°2θ, depending on the scanning speed.

In X-ray diffraction the wave length of the X-rays is known, along with the angle θ and the interplanar spacings must be determined by the Bragg equation. The diffraction patterns are identified by comparing the d spacings (generally the three strongest lines) and relative intensities of the unknown with these data for known specimen. X-ray diffraction is used primarily for qualitative work. Fluorescent X-ray analysis can be quantitative, offering the greatest advantage for elements having atomic numbers larger than 10 present in amounts of from a few parts per million to 100 percent.

It will be appreciated that the calculation of wave lengths or interplanar spacings would be a very tedious and time consuming job. For this reason sets of tables have been developed which give interplanar spacing values for the angle 2θ for the types of radiation commonly used. Similar tables are available for use with X-ray fluorescence methods giving wave lengths and elements in the case of known analyzing crystals and angles. It is apparent however than even with the availability of tables each peak has to be located. Taking

these values from tables takes time, training, and is also subject to error. Thus a tabulary compilation of a great number of d spacings and wave lengths is rather voluminous and complex. It is this data interpretation problem with which my invention is concerned.

SUMMARY OF THE INVENTION

In order to facilitate the derivation of d spacings from X-ray diffraction data or elements from X-ray fluorescence data the interpretation apparatus of this invention is provided. The interpretation apparatus of the invention utilizes data obtained by direct recording X-ray spectrometers. The apparatus herein contemplates a data or background sheet and an interpretive sheet. The data on the background sheet is in the form of a plot of either an X-ray diffraction pattern or an X-ray fluorescence pattern. The interpretive sheet is a transparent sheet superimposable over the data sheet with indicia imprinted in scale thereacross. The indicia on the interpretive sheet is in the form of an arrangement of elements when the data on the background sheet is in the form of an X-ray fluorescence spectrogram and in the form of interplanar spacings when the data on the background sheet is in the form of an X-ray diffraction spectrogram. The interpretive sheet is adapted to be aligned with the data sheet and its scale is equal to that of the background sheet so that peaks on the data sheet will be identified by values on the interpretive sheet.

DETAILED DESCRIPTION OF THE INVENTION

This invention in effect provides a qualifying instrument for use in X-ray fluorescence and diffraction analysis. Since the interpretive sheet is derived from the X-ray scan in conjunction with the Bragg equation, it can be made applicable to any instrument crystal used or to be used. Troublesome instrument misalignment and the accompanying shift of peaks is immediately correctable by aligning the interpretive sheet on the tube peak. The apparatus can be made to cover any scanning distance, for example 4°to 88°2θ, 10°to 40°2θ, 20° to 30°θ, 30° to 146°2θ, and the like. In addition the apparatus can be made for use with any scanning speed, for instance 4° per inch, 2° per inch, 1° per inch, 1/4 ° per inch, etc. The apparatus can also be used with any X-ray tube, KV or MA setting, attenuation, slit, purge or mounting apparatus. These can affect intensity and the location of the peaks.

The interpretive sheet desirably is made of clear plastic, preferably one of the semi-rigid transparent plastic materials, to allow the analyst to easily extract intensities, which in fluorescence are directly proportional to the amount of the element present. The interpretive sheets can also be made of other transparent materials or other colors, but their use may be impaired by the inherent problems of such materials, i.e., loss of the starting point, misalignment due to nonvisibility, enhanced breakage, wear, and the like. Only the major alpha order lines and secondary beta order lines of the K and L series are normally used on the interpretive sheet except where confusion and interference occur. It is well known in the field that several lines characterize each element. As many of these lines as are needed for positive identification of the element are included on the interpretive sheet. The apparatus thus

provides a rapid and inexpensive method of interpreting fluorescence and diffraction scans.

For a more complete understanding of the interpretation apparatus of the invention it will now be considered in conjunction with the accompanying drawings.

FIG. 1 is a plan view showing the apparatus in use in X-ray fluorescence analysis.

FIG. 2 is a plan view showing the apparatus in use in X-ray diffraction analysis.

Referring first to FIG. 1, 2 indicates the background sheet containing an X-ray fluorescence curve 4. This can be a fluorescence spectrogram made by a direct recording X-ray fluorescence instrument. As can be seen curve 4 contains peaks 6 recorded at various crystal angles as the goniometer rotated. The height of the peaks is dependent upon their intensity since the intensity has been plotted as the ordinate versus the reflection angle as the abscissa.

Since various elements are identified by the characteristic wave lengths of their radiation, at this point it is necessary to determine the wave lengths at peak angles from the Bragg equation and then check published values to see which element is characterized by the determined value. By this invention, however, it is necessary only to superimpose interpretive sheet 10 over curve 4 as shown in FIG. 1 and to align interpretive sheet 10 with background sheet 2 using a starting point 12 which is optional with the user, so that degrees on interpretive sheet 10 coincide with reflection angles recorded on background sheet 2, taking into account previously ascertained crystal or instrument shifts. It can be seen that the elements whose peaks are seen that can be found on the interpretive sheet.

As an illustration a sample is scanned at reflection angles of 8° to 88°2θ and recorded at 4° per inch using a chrome tube X-ray source. The completed scan is 20 inches in length. In using the interpretive sheet with this can the starting point, i.e., 8°, is aligned with the 8° mark on the interpretive sheet. Peaks are noted the entire length of the scan being non-equidistant from the starting point or from other peaks in the scan. It is noted that the chrome line marked on the interpretive sheet does not correspond exactly with the peak which is known to be the tube peak. This small difference is known to be the shift of the instrument which can be attributed to malalignment and/or other sources of error known in the field. The interpretive sheet can be aligned to compensate for or to correct the instrument shift. Each peak then need not be separately corrected as is usually the case. After the interpretive sheet is realigned, the peaks can then be positively identified as elements from the lines identified on the interpretive sheet. The peaks which are now left on the recording paper which have not been identified as an element by the interpretive sheet can be assumed to be lesser lines of elements already identified. They are of little interest unless an interference modifies the major lines of an element.

To interpret fluorescence scans without the interpretation apparatus, consider the same scan 8° to 88° under the same instrument conditions. First, the scan has to be indexed according to the scanning speed, i.e., 4° per inch, each successive inch has to be marked from the starting point, i.e., 8°, 12°, 16°, 20°, 24°, 28°, etc. to 88° or the full distance which was scanned. Short scans are sometimes employed over short distances where one is looking for specifics. Next, each peak has to individually be marked with the degrees at which it occurs. The instrument malalignment must be taken into consideration if an accurate interpretation is to be made. This can be done before or after indexing the peaks with degrees and is accomplished by determining where the tube peak is supposed to occur, and where it actually occurs. This difference must be added or subtracted from each peak on the scan. After the correction is complete, each peak, usually starting with the largest on the scan, then must be ascertained from a book of tables, usually supplied by the maker of the instrument used. Each element is identified in this manner. As can be seen, experience is required in addition to the knowledge on where to look. There are also possible interpretation problems due to interferences.

Considering now X-ray diffraction, again each peak has to be located in a book of tables. The peaks are identified as "d" values. Locating values for each peak takes time, and this time is sometimes wasted because the instrument is malaligned. The procedure has to be repeated when it is realized that there has been a shift in the peaks.

The X-ray diffraction interpretation apparatus is shown in FIG. 2. The diffraction pattern 20 is again recorded on a background spectrogram 22, and the starting point 26 again is optional with the user. In this instance the interpretive sheet 24 does not immediately identify the peaks 28 as compounds. The *d* values are, however, taken directly off of the interpretive sheet. This aids in the indexing of the scan, and shortens the time spent on interpretation once the *d* values are established. The same advantages apply to the apparatus of FIG. 2 as to the fluorescence apparatus of FIG. 1.

If will be obvious to those skilled in the art that many modifications of the apparatus will become apparent through use of this invention. Thus misalignment—and subsequent shift of the peaks—is immediately correctable by aligning the interpretive sheet with a known element or compound peak which is always present on the data sheet as is well known in the field. All elements, whether identified by alpha, beta, *l* or *m* lines, and regardless of orders of lines (*n* values in Bragg equation), which can be detected by X-ray fluorescence techniques can be identified by means of the interpretive sheet. However, all elements, lines, and orders of lines are not needed on every sheet due to their rareness or the fact that they are not present in samples in the field in which the analyses are made. In addition the radioactivity of some elements bars this method of analysis. It can be seen that the invention provides an accurate and reliable method of interpreting diffraction and fluorescence scans from X-ray units of any manufacturer, using any target tube, either foreign or domestic, regardless of the measuring system used in scanning or calculations, i.e., inches, feet, meters, centimeters, etc. They can also be used when scanning from high to low angles or visa-versa. It will be apparent also that the invention will apply when more than one crystal is used. Thus it would be possible to combine indicia of several crystals on one interpretive sheet where the user has several crystals. There will be,

of course, an increased chance of error. Space can also be provided on the interpretive sheets for an individual's own conversion factors and the like. These and other variations are within the scope of this invention.

What is claimed is:

1. A process for identifying unknown substances from data on X-ray diffraction and X-ray fluorescence spectrograms obtained by direct recording spectrometers, the data thereon being plots of spectral intensities versus reflection angles, comprising preparing a transparent interpretive sheet having indicia imprinted in scale horizontally thereacross, in the form of an arrangement of elements to correspond with the spectrogram when it is an X-ray fluorescence spectrogram, and in the form of an arrangement of interplanar spacings to correspond with the spectrogram when it is an X-ray diffraction spectrogram, imprinting on said transparent interpretive sheet a vertical index line, making the scale of the abscissa on the interpretive sheet equal to the scale of the abscissa on the spectrogram, positioning the spectrogram in a working position, superimposing the corresponding transparent interpretive sheet over the spectogram and aligning the index line on the interpretive sheet with an abscissa value on the spectrogram so that peaks on the data sheet will be identified by indicia on the interpretive sheet.

2. The process of claim 1 wherein the spectrogram has thereon a plot of an X-ray diffraction pattern and wherein the transparent interpretive sheet has values of interplanar spacings thereacross in alignment with reflection angles on the spectrogram sheet.

3. The process of claim 1 wherein the spectrogram has thereon a plot of an X-ray fluorescence pattern and wherein the transparent interpretive sheet has symbols of chemical elements thereacross in alignment with reflection angles on the spectrogram.

4. The process of claim 1 wherein the transparent interpretive sheet has thereupon a correction vertical index line compensating for the instrument tube peak and wherein the index line is aligned with an abscissa value on the spectrogram.

5. The process of claim 1 wherein the vertical index line on the interpretive sheet is based on the interplanar spacings of a known crystal.

6. The process of claim 1 wherein the vertical index line on the interpretive sheet is based on the known wave length of the ray emitted by the target element.

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