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Title / Titre: THE APPLICATION OF NONDESTRUCTIVE TESTING TECHNIQUES TO THE ANALYSIS OF ARCHEOLOGICAL MATERIALS

L'APPLICATION DES MÉTHODES D'ESSAIS NON DESTRUCTIFS À L'ANALYSE DES MATÉRIAUX ARCHÉOLOGIQUES

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SUMMARY: The applications of x-ray fluorescence analysis, radiography, and other techniques to archeological materials are described.

RESUME: On décrit les applications de l'analyse par fluorescence X, radiographie et autres méthodes aux matériaux archéologiques.

I. INTRODUCTION

For several years, the Los Alamos Scientific Laboratory, operated by the University of California for the U. S. Energy Research and Development Administration, and the Laboratory of Anthropology of the Museum of New Mexico have engaged in a cooperative effort investigating the applications of nondestructive examination techniques to problems in the fields of archeology and museology. This cross-fertilization between the analytical disciplines and the archeological community has proved beneficial for both parties. As a direct result of these efforts, an archeological materials analysis center, hopefully, will soon be established as a joint effort between the two laboratories.

Several different nondestructive examination techniques have been applied to a wide variety of archeological materials. Samples that have been examined include ceramic wares, lithic artifacts, turquoises, metallic samples, teeth, and bones. X-ray fluorescence analysis, beta-backscattering measurements, scanning electron microscopy, and radiography have been utilized. Several of the preliminary studies are reported here. The results of these investigations and their importance to archeological research are presented.

II. RADIOGRAPHY OF METALLIC SAMPLES

Several heavy sections of metal were excavated from the ruins of an early Spanish mission in Northern New Mexico. It was known from historical documents that the mission had been destroyed by Indians during the Pueblo Uprising in the year 1680. One of the first acts the Indians perpetrated when razing the mission was to remove and destroy the mission bell. The archeologists felt the metal samples could possibly be from the Spanish bell. We undertook the examination of two of those samples.

A radiograph of the first sample is shown in Figure 1. This radiograph, taken at 1 MeV, shows the metal casting to be very sound, with one low density stringer present. The second sample is shown in Figure 2. You can see the very different appearance of the casting in this radiograph. The large number of voids indicates very poor casting techniques. Obviously, the two samples are not from the same casting. More information on these two samples was desired.

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III. X-RAY FLUORESCENCE ANALYSIS OF METALLIC SAMPLES

In an effort to determine the metal(s) in these two samples, x-ray fluorescence analysis was applied. Energy dispersive x-ray fluorescence analysis has become more common with the advent of the high-resolution, solid-state lithium-drifted silicon detectors. Using such detectors, with radioactive sources or x-ray generators as the excitation source, the simultaneous measurement of many elements can be performed. A typical source/collimator/detector assembly is illustrated in Figure 3. The sample is placed in front of such an assembly and is irradiated by the source, simultaneously emitting x-rays characteristic of the elements contained in the sample. The signal from the detector is amplified, digitized, and processed into the memory of a multichannel pulse height analyzer. A typical equipment arrangement is shown in Figure 4. Using such a system, with several different energy radiation sources, all of the elements above sodium can be detected. With radiation source strengths on the order of 10 to 25 mCi, sufficient data for most purposes can be collected in a few minutes. This method is very sensitive to the surface elemental content of the sample, penetrating no more than a fraction of a millimetre for most samples. Very little sample preparation is required, in most instances, other than cleaning of the surface to be measured.

Using such an analysis system, the two metallic artifacts were examined for elemental content. The surfaces were cleaned with distilled water and vigorous brushing. No attempt was made to remove the patina from the surface. Two radiation sources, ^{109}Cd and ^{145}Sm, were used to cover the energy ranges 1 to 20 keV and 1 to 40 keV respectively. To maximize the sensitivity for trace element content, counting times of 1200 s were used. The major constituent elements in each sample were copper and tin (bronze), with significant amounts of iron, antimony, and lead also present. The radiographically sound sample contained much less of these impurities than the poorly cast sample, indicating different smelting and refining techniques. The radiographically good sample, surprisingly, contained two additional elements - silver and gold!

There are legends that personal valuables were added to the melt pot when bells for new missions were being cast. The discovery of these elements, silver and gold, in this fragment lends credance to these legends and makes more credible the hypothesis that this fragment could be from the original Pecos Mission bell. We are attempting to obtain several additional metal fragments that were excavated at the same site fifty years ago for comparison purposes.

Other metal objects that have been examined with this technique include projectile points, lance tips, medallions, and religious artifacts. A certain group of copper artifacts, all from the same locality and dating from the eighteenth century, contain significant amounts of silver and small amounts of strontium and rubidium. There are known copper mines in the same region and these samples may represent crudely refined copper from these mines. We are presently obtaining ore samples from that area for reference purposes.

IV. CERAMIC ARTIFACTS

X-ray fluorescence analysis has proved valuable in the study of ceramic materials, glazes, and paints. Washed sherds of several different types of Spanish, Mexican, Indian, and contemporary pottery have been examined. In the clay paste, significant variations have been observed in the concentration of certain elements - rubidium, strontium, yttrium, and zirconium. Many other elements also appear in typical clay samples, but most do not appear diagnostic for our purposes.

Definite relationships have been established between cert[illegible]ttery types and their regions of manufacture. For example, x-ray fluorescence [illegible]is was used to characterize several series of "Apachean" pottery sherds recover[illegible]m three different archeological sites. [1] This pottery belongs to a class of [illegible]ceous culinary ware commonly found on 18th, 19th and, occasionally, on early 20th [illegible]ry ruins throughout the Upper Rio Grande culture province. Two of the sites, [illegible]rupa Ruin (LA 5050) and San Miguel del Vado (LA 2734), are located in the Pecos River drainage, while the third, Los Alamos Canyon (LA 5140), is in the Rio Grande Valley. On the basis of the Rb/Sr/Y/Zr ratios, the Pecos sherds are quite distinct from the Rio

Grande samples. The samples from the two different Pecos sites are virtually identical, as are the individual Rio Grande sherds. The two different elemental content patterns imply localized clay sources for these two areas where the Jicarilla Apaches were known to have been producing pottery. A typical x-ray spectrum of the Rio Grande Apache pottery is shown in Figure 5. The different elemental pattern for the Pecos sherds is shown in Figure 6.

Similar variations in elemental content patterns have been noted in other ceramic assemblages. Preliminary studies have been completed on Mayan Fine Orange and Thin Slate wares excavated in Mesoamerica. Spanish and Mexican majolica wares - a class of soft paste, lead/tin glazed pottery - have been examined to determine the lead/tin ratios in the various types. The elements used for the colored paints in the glazed wares have been determined. Thus, x-ray analysis has offered information on many facets of ceramic manufacture and decorative techniques.

V. RADIOGRAPHY OF CERAMICS

Two principal methods were used to construct pottery in the prehistoric American Southwest. The technique used by the Puebloan groups was primarily the coil-form method. A long thin strand of clay would be wound helically to form the desired shape, and then the surfaces smoothed by scraping. The method used primarily by the Apaches was paddle-and-anvil construction. A lump of clay would be squeezed between the fingers and thumb or other suitable paddle and anvil until it was the desired shape. A third technique was the combination of the first two in the same vessel. When fragments of pottery are excavated, it is difficult to know which method of construction was used. Low voltage radiography has been used to examine this particular problem. A radiograph taken at about 75 keV of an Apache sherd is shown in Figure 7. The high density inclusions in the paste are particles of temper. Tempering materials are added to the clay to prevent shrinkage and cracking during the firing of the pottery. As can be seen in this illustration, the orientation of the temper is totally random. This has proved true in all of the paddle-and-anvil pottery examined to date. In coil-formed pottery, the temper particles have been seen to assume a preferred orientation as a result of the formation of the individual strips of clay used to manufacture the vessel walls. Unfortunately, this is almost impossible to see in reproductions of the original radiographs. Here we have seen how routine radiography can aid the archeologist in the solution of a difficult problem.

VI. EXAMINATION OF LITHIC MATERIALS

The most durable and the oldest traces of man's early activities are the wide variety of stone tools and implements found throughout the world. The study of these lithic artifacts can provide much information on methods of manufacture and the spread of this technology through trade or travel. A recent study from Japan traces stone implements to the source of raw materials by x-ray fluorescence analysis [2]. We are presently involved in the identification and sourcing of a series of obsidian and chert implements using the same technique. The preliminary data are incomplete, but are encouraging.

Many tools are produced by flaking techniques. This can be accomplished by percussion, pressure, or by heating and sudden cooling. Each of the methods leaves flake scars with distinctive characteristics. Tools were also produced by grinding and polishing. The surfaces of implements are usually examined by optical microscopy, but this technique is limited by the depth of field. One of the advantages of scanning electron microscopy is the very large depth of field, even at high magnifications. An x-ray detector can also be incorporated into the scanning microscope, allowing x-ray fluorescence analysis on a microscopic scale.

A typical application of the scanning electron microscope is shown in Figure 6. This object is a very small, flaked chert drill excavated at the Pecos Pueblo (LA 625). Such drills were used to make the holes through beads and pendants and when other small diameter holes were needed. The flake scars are clearly seen in the figure and are typical of the appearance of pressure flakes. Also shown in this illustration are use or wear scars on both edges and at the tip. In cases such as this, the great depth of field of the scanning electron microscope is clearly an advantage. Mosaics comprising many micrographs can be laid up to cover the entire object.

VII. SOURCING OF TURQUOISES

Turquoise artifacts are found in archeological sites throughout the American Southwest and Mesoamerica. There are many sources of turquoise that are known to have been mined in prehistoric times. It is desirable to trace artifacts to their original source as a possible indicator of patterns of trade or travel. Sigleo[1] has successfully determined the origin of turquoise beads excavated at the Snaketown site in Arizona by thermal neutron activation analysis. We are applying x-ray fluorescence analysis to the study of turquoise as a complement to the neutron analysis. The x-ray technique offers better sensitivity for certain elements with low neutron cross sections. We have obtained samples from several known prehistoric mining sites and are in the process of building a reference library of turquoise x-ray spectra. A large number of these samples have already been analyzed using thermal neutron activation, providing the opportunity for a direct comparison of the two techniques.

During the excavation of a kiva, a ceremonial chamber, at the Tijeras Pueblo (LA 581) in New Mexico, an unusual mosaic was discovered. Bits of worked turquoise, coral, obsidian, and shell were inlaid directly into the floor in a very orderly pattern. No one knows the meaning or origin of this practice. We examined the eleven turquoise samples and they are apparently all from the same source. We do not have reference samples with the same elemental x-ray patterns in our files, so we cannot identify that source at present.

VIII. OTHER TECHNIQUES AND MATERIALS

Several other analytical techniques have been used in the examination of archeological materials. The beta particle backscatter inspection has been developed for the measurement of density and/or thickness of materials. The amount of beta backscatter from a sample is directly proportional to the product of the density and the square of the atomic number of the material, if the material is thicker than about one quarter of the range of the beta particle. If, for example, a high Z, high density material is deposited on a low Z, low density substrate, thickness measurements of the deposit can be made over a limited range. After the maximum range is reached, only density variations in the deposit can be determined. We have used this technique to measure the density of the lead/tin glazes on Spanish and Mexican majolica wares. These glazes are usually very thick, approaching 1 mm, making the density measurement possible. We have found a direct correlation between the lead/tin content of the glaze and the density of the glaze. For Indian lead-painted pottery, the paint is usually quite thin, allowing thickness measurements to be performed. We are presently measuring a series of Indian lead-decorated wares from different time periods to follow the development of the use of mineral paints.

Lead-glazed pottery, when improperly fired, will react with acidic foods with lead going into solution. Extensive use of low-fired lead-glazed ware will result in increased lead levels in the body. We have been investigating lead levels in teeth and bones using the x-ray fluorescence technique. Some evidence of increased lead levels in prehistoric Indian bones has been noted, but the results to date are inconclusive.

The x-ray fluorescence examination of teeth indicated a possible relationship between the strontium content of the teeth and the age of the individual. There is a wide variation of the strontium in teeth dependent on locale, strontium content of the water, and other undetermined factors. If we could assume that a given band of individuals lived their entire lives in one location, it may be possible to determine their ages through the strontium uptake. The usefulness of this measurement has not yet been successfully demonstrated.

In addition to the above techniques, we are investigating the applications of Mössbauer effect spectroscopy, prompt neutron activation analysis, electron spectroscopy for chemical analysis, and others. The organic chemistry group within the Laboratory has applied a wide variety of techniques to the study of organic residues, dye stuffs, and soil samples.

Short title
Titre court

NONDESTRUCTIVE TESTING IN ARCHEOLOGY

IX. SUMMARY

In the above discussions, we have demonstrated how many existing nondestructive examination techniques can be applied to a variety of archeological investigations. One of the main difficulties encountered in such a program is communication. Each discipline has its own jargon, therefore the analyst must learn the language of the archeologist and vice versa. The archeologist also must learn what questions can properly be asked of a particular inspection method. When this is accomplished and a cooperative and collaborative atmosphere is established, the merger of the two disciplines into the one of archeometry results. We, in the field of nondestructive testing, have the techniques to answer many problems in other fields, but, first, we need to know what these problems are. We must take the initiative in seeking them out.

X. ACKNOWLEDGEMENTS

This work was possible only with the cooperation and encouragement of the staffs of both the Los Alamos Scientific Laboratory and the Museum of New Mexico. David H. Snow, Curator of Archeology at the Museum, has earned special recognition through his patience in dealing with the non-archeologist and for his suggestions of areas of investigation. Most of the samples reported here were supplied by the Museum. Megan A. Cassidy of the Nondestructive Testing Group performed many of the analyses of these samples. This work was done under the auspices of the U. S. Energy Research and Development Administration.

XI. REFERENCES

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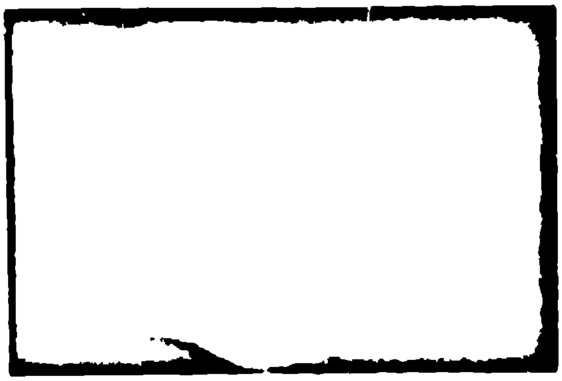

Fig. 1. Radiograph of sound casting.

Fig. 2. Radiograph of poor casting.

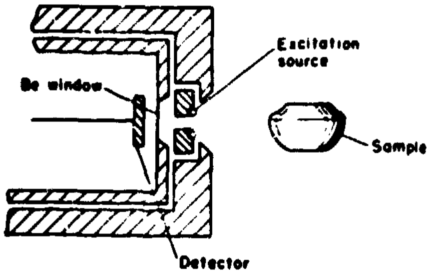

Fig. 3. Source/collimator/detector assembly.

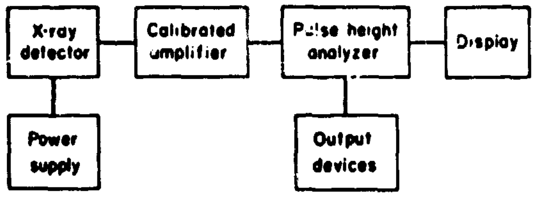

Fig. 4. X-ray analysis system.

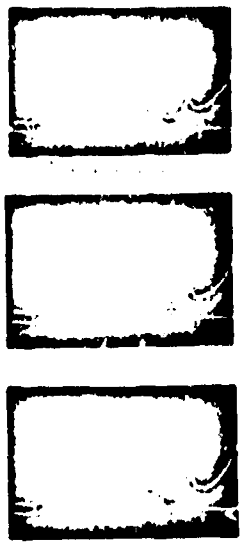

Fig. 5. X-ray spectra of Rio Grande Apache pottery.

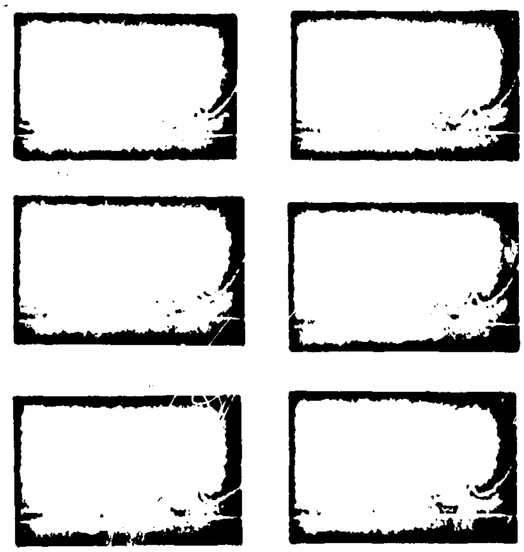

Fig. 6. X-ray spectra of Pecos Apache pottery.

Fig. 7. Radiograph of Apache sherd.

Fig. 8. SEM of pecos drill.