SPECIAL ISSUE

ACKNOWLEDGING FIFTY YEARS OF NEUTRON ACTIVATION ANALYSIS IN ARCHAEOLOGY

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Neutron activation analysis has been an important analytical technique for chemical characterization studies of archaeological materials since 1957, and remains one of the best analytical approaches for bulk chemical characterization of archaeological ceramics and other materials. This paper introduces a series of reports that document the history of NAA applied to archaeological materials.

KEYWORDS: NEUTRON ACTIVATION ANALYSIS, ARCHAEOLOGY LABORATORIES, HISTORY

INTRODUCTION

Archaeologists, historians, chemists and physicists have employed a variety of chemical and physical approaches to study artefacts and historical objects since at least the late 18th century (Caley 1949, 1951, 1962, 1967; Harbottle 1976). During the past 50 years, compositional analyses of archaeological materials have increasingly been used to address a broad spectrum of anthropological topics, including exchange, provenance and manufacturing technology. Today, chemistry-based studies of archaeological materials are considered routine, and many studies involving ceramic and lithic materials employ such approaches in one form or another.

Many of the analytical techniques employed by archaeometrists today are a direct outgrowth of research programmes developed by various governmental agencies during the Second World War and the period immediately following. In the nuclear sciences, for example, multiple research reactors were established at national laboratories and academic institutions around the world with the goal of developing nuclear applications for scientific research in disciplines unrelated to defence. One research area—neutron activation analysis (NAA)—was concerned with using neutrons to chemically characterize various materials. In late 1954, J. R. Oppenheimer contacted Richard Dodson to suggest the possibility of using NAA to determine the provenance of archaeological materials, a question that he had previously discussed with archaeologists. Following a meeting with Oppenheimer shortly afterwards, Dodson enlisted Edward Sayre to undertake the experimental work on samples of Old World pottery. The results of this experiment.
were reported at a meeting organized by Oppenheimer at the Princeton Center for Advanced Studies in March 1956; the results were subsequently published the following year (Sayre and Dodson 1957: for extended discussions, see also Harbottle 1976; Beaudry-Corbett 2003; Harbottle and Holmes, this issue).

The 1950s and 1960s were truly an exciting time in archaeological science. At about the same time that NAA was being developed, other parallel advances were being made in the areas of dating, remote sensing and materials characterization. Willard Libby and colleagues had in 1949 demonstrated the feasibility of the carbon-14 method of dating through the analysis of archaeological samples of known age (Libby 1952). The impact on archaeology was immediate, and within a few years several carbon-14 dating laboratories were established around the world. In 1948, the first X-ray fluorescence (XRF) spectrometer was constructed (Friedman and Birks 1948), and by 1960, Hall had reported that XRF was a well-established analytical method in the laboratory and in industry (Hall 1960). Proton magnetometers saw their first archaeological applications with the pioneering work of Martin Aitken (1958, 1959a,b). Other advances, such as archaeomagnetic and obsidian hydration dating, were developed and witnessed widespread application in the 1960s. In the late 1960s a major analytical breakthrough occurred when Ge-based and then later hyperpure or intrinsic Ge detectors were developed and NAA evolved into what is commonly referred to as INAA (instrumental neutron activation analysis). As the 1960s came to a close, approaches to dating, materials characterization and remote sensing were well developed and widely employed in archaeology—clearly the field of archaeometry had emerged, and NAA was near the forefront.

NEUTRON ACTIVATION ANALYSIS 1957–2007

The year 2007 marks the 50th anniversary of Sayre and Dodson’s seminal publication of the application of NAA to the study archaeological materials (Sayre and Dodson 1957). It seems appropriate to commemorate this golden anniversary with a special issue of *Archaeometry*. The journal *Archaeometry*, which was established in 1958, has grown from an annual bulletin into a world-renowned international quarterly publication. Today, the journal serves as a primary outlet for publication of the ever-growing body of interdisciplinary research that bridges the divide between the chemical and physical sciences and archaeology. Throughout its history, *Archaeometry* has published more than 100 papers concerning neutron activation analysis and/or the interpretation of the resulting data. In fact, the first two papers published in *Archaeometry* dealt with NAA of coins (Emeleus 1958; Kraay 1958).

When NAA was first developed, the advantages of this analytical technique over other chemical characterization techniques were quickly recognized by researchers. These include: (1) ease of sample and standard preparation; (2) determination of the concentrations of multiple elements in a bulk sample; (3) many elemental determinations with high analytical precision; and (4) good inter-laboratory comparability. Prior to the development of NAA, most chemical studies of archaeological materials, particularly ceramics, centred around wet chemistry or emission spectrometry. NAA rapidly replaced these analytical techniques as the ‘true’ bulk sample technique of choice, and in the ensuing decades NAA emerged as one of the most powerful and widely applied analytical techniques for chemical characterization and provenance-based research of ceramics, obsidian,1 chert, flint, basalt, glass, metals and other archaeological and

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1 We recognize, however, that during the past two decades most chemical analyses of obsidian (and probably metals) have been conducted by XRF.
historical materials. It is difficult to provide exact numbers, but at least one dozen major laboratories and numerous smaller laboratories are or have been involved in the application of NAA to archaeological materials, in many cases involving the direct collaboration of physical scientists and archaeologists.

Despite the proliferation of NAA in the 1960s and 1970s and the fact that it remains today one of the best analytical approaches for bulk chemical characterization of archaeological ceramics and other materials, many of the major archaeometry-based NAA programmes are no longer in existence. These include programmes at the Brookhaven (BNL) and Lawrence Berkeley National Laboratories (LBL), which operated under the auspices of the US Department of Energy (DOE) until the mid-1980s, when DOE’s mission changed and funding for the laboratories was withdrawn. In the early 1990s, the NAA programmes at the Hebrew University of Jerusalem, the University of Manchester, and the British Museum ceased operation. More recently, the Ford reactor at the University of Michigan and the SLOWPOKE reactor at the University of Toronto were closed. The closure of these programmes in some cases corresponded to the retirement of key personnel, but in many cases resulted from the decommissioning of research reactors. Although funding and permission to build research reactors was easily attainable during the 1950s and 1960s, in today’s world reactors are expensive to maintain, upgrade and replace. Additionally, the increased need for security and a general misconception by the public about the hazards of small research reactors will in all likelihood preclude any new research reactors from being built—at least for the foreseeable future. There is some hope, however, given that Kuleff and Djinkova (this issue) indicate that, after being closed for more than a decade, the University of Sofia reactor is scheduled to again come on-line within the next year.

Although the closure of so many laboratories may at face value suggest an ominous future for NAA, this is not necessarily the case. When the University of Michigan reactor closed in 2003, the archaeometry programme was transferred to the Oregon State University reactor. In South America, there are several relatively new NAA-based archaeometry programmes, located in Peru, Chile, Argentina and Brazil. Although the South American programmes are smaller in scale, researchers at each facility have made important contributions to South American archaeology. In Europe, research at the Bonn (Germany), Demokritos (Greece), ITN (Portugal), University of Pavia (Italy) and Budapest reactors remains strong. In North America, the MURR and Smithsonian–NIST programmes continue to analyse large numbers of samples. Several smaller programmes also exist at Texas A&M, the University of California–Irvine, the University of Wisconsin, École Polytechnique (Montreal) and the Instituto Nacional de Nucleares in Mexico, among others. As a consequence of the research conducted at these laboratories (and many others not mentioned), archaeological applications and publications concerning NAA are at an all-time high.

We estimate that during the past 50 years more than 150,000 NAA analyses of pottery, obsidian, chert, metals and other matrices have occurred at more than 25 laboratories around the world. Estimating the cost of an analysis (both short- and long-lived isotopes) at a minimum of US$100, the value of these extant NAA databases easily exceeds US$15 million. This figure includes only costs associated with generating the analytical data, and does not reflect the value of information on past exchange and interaction patterns that archaeologists of the future might glean from these data. Data generated from these earlier studies can, in most cases, be intercalibrated with data generated at other laboratories. Intercalibration of NAA data is possible, in large part, because most NAA data are traceable to international standards and widely available quality-control samples. At the MURR laboratory, we have initiated a
programme to archive and disseminate NAA data to interested researchers via our website (http://archaeometry.missouri.edu). In addition to data generated at MURR, ceramic data generated at the University of Manchester and the Brookhaven Limestone databases are also available for download. We also recently acquired the Berkeley archives and have begun the process of converting the data into a usable electronic format. In addition to the MURR laboratory, the archaeometry group at Bonn has also begun making NAA data available for download via their website (http://www.iskp.uni-bonn.de/gruppen/mommsen/top.html). The Bonn website also hosts the Perlman–Asaro data bank of Mycenaean samples.

Archiving and disseminating NAA data, however, is only one of many steps required to ensure that data generated today have an intrinsic value for the next generation of researchers. As indicated above, many of the first-generation NAA-based archaeometry laboratories are no longer in existence. In other cases, many scientists who have operated NAA laboratories are retired, rapidly approaching retirement age or deceased. Finally, some NAA users are fourth- and fifth-generation practitioners who do not necessarily have a full appreciation or understanding of the earlier research. It seems inescapable to us that for NAA and provenance-based research to have relevance in the future, that we must pause to document the history of NAA thus far, so that this knowledge is preserved and passed on to future generations of scientists. Given that many NAA laboratories no longer exist, and that the long-term outlook for some others is less than optimal, we believe that it is important to document in one place, the histories of individual laboratories, their major accomplishments and their major contributions to the field of archaeology, so that as provenance studies progress into the future, we have documented, to some extent, where we have already been. The 17 papers in this issue are but one step in this process and provide individual histories of laboratories no longer in operation and those currently operational—including a few nascent programmes.

LOOKING TO THE FUTURE

As with any analytical technique, the future of NAA is uncertain. It is a fact that technology continually changes (hopefully for the better), and that new and presumably better methods and equipment are constantly incorporated into the available repertoire of analytical techniques. Newer techniques such as inductively coupled plasma–mass spectrometry (ICP–MS) are proliferating in chemistry-based studies at an exponential rate. One such example is the substantial increase in the use of multi-collector ICP–MS for heavy isotope ratio studies (in the fields of archaeology, geology, chemistry etc.), when only a few years ago thermal ionization mass spectrometry (TIMS) was the ‘industry standard’. Elsewhere, we have witnessed the widespread application of laser ablation ICP–MS for studies of obsidian, metals, and painted and glazed surfaces, among others (e.g., Speakman and Neff 2005). However, despite the widespread application of ICP–MS, few studies have focused on ceramics (e.g., Kennett et al. 2004; Little et al. 2004), in part because the labour and time required to properly digest ceramics to obtain a ‘true’ bulk analysis inhibits its widespread application. On other fronts, portable instrumentation, such as portable-XRF (PXRF), is seeing increased use in archaeology (e.g., Craig et al. n.d.). Regardless of these relatively recent advances with other techniques, requests for NAA, at least at the MURR laboratory, have steadily increased during the past decade. Although we welcome new analytical approaches, we remain optimistic that NAA to continue to play a major role in material characterization studies well into the future. Additionally, we believe that NAA will continue its role in validating new analytical techniques as they become available.

ACKNOWLEDGEMENTS

We thank Mark Pollard and Jane Simcox for their assistance and support in putting this special issue of *Archaeometry* together. We also thank Ron Bishop, Jeff Ferguson and Ron Hancock for their insightful comments on earlier drafts of this paper. Finally, we acknowledge each of the authors and reviewers whose contributions made this special issue possible.

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