ISBN: 978-3-95450-214-1 ISSN: 2673-5490 doi:10.18429/JACoW-IPAC2021-WEPAB012

PRELIMINARY INVESTIGATION INTO ACCELERATORS FOR IN-SITU CULTURAL HERITAGE RESEARCH

T. K. Charles*[1], University of Liverpool, Liverpool, UK
A. Castilla†[2], Lancaster University, Lancaster, UK
[1]also at Cockcroft Institute, Daresbury, UK
[2]also at CERN, Geneva, Switzerland

Abstract

Ion Beam Analysis (IBA) centres have provided researchers with powerful techniques to analyse objects of cultural significance in a non-destructive and non-invasive manner. However, in some cases it is not be feasible to remove an object from the field or museum and transport it to the laboratory. In this paper, we report the initial investigation into the feasibility of a compact accelerator that can be taken to sites of cultural significance for PIXE analysis. In particular, we consider the application of a compact, robust 2 MeV proton accelerator that can be taken into the field to perform PIXE measurements on rock art. We detail the main challenges and considerations for such a device.

INTRODUCTION

Proton Induced X-ray Emission (PIXE) is the most widely-used IBA technique. Being non-invasive and non-destructive makes PIXE popular for studying items of cultural heritage significance, to determine elemental compositions [1]. The basic principle of PIXE is as follows: protons (typically 2-5 MeV) are fired at a sample, ionising an electron. When an electron from a higher energy shell falls down to fill the vacancy, it emits a characteristic X-ray, which can be used to identify the elements in the sample.

Typically PIXE is performed at dedicated IBA centres, with large electro-static accelerators. However, in some circumstances items we wish to study cannot be moved from the field or museum. In this case, a portable accelerator would be beneficial.

Uncovering Hidden Rock Art

Ancient rock art is one example that could hugely benefit from PIXE analysis performed with a portable accelerator. Over time, rock art fades due to exfoliation of the paint pigments, or through the accumulation of dust, micro-vegetation such as lichen, or mineral accretion, all of which make the rock art appear faded [2]. PIXE analysis would allow us to identify the pigment elements that exist in trace amounts or hidden by micrometer layers of dust or graffiti, which could allow us to build up a 2D elemental map to reveal how the artwork may have looked thousands of years ago.

Why PIXE and not XRF?

Portable X-ray Fluorescence (pXRF) devices designed to be taken into the field, are popular among some archaeologists. Whilst these devices give an immediate indication of the elements present, limitations related to the low photon flux and large spot size (up to a few cm^2) [3] mean these devices and are often only used as a preliminary step in assessing rock art [4–6].

One key difference between PIXE and XRF is that PIXE demonstrates higher sensitivity to low-Z elements, whereas (lab-based) XRF demonstrates higher sensitivity for higher-Z elements. With an extracted beam, PIXE is highly sensitive to elements between Sodium (Z=11) and Ga (Z=32) [7]. Whereas pXRF has a more restricted elemental acquisition, limited to elements between $Z = 19$ and 41 (i.e. the elemental range bounded by K and Nb) [3, 7]. Additionally, when compared to XRF, PIXE generally exhibits a greater signal-to-noise ratio, due to lower bremsstrahlung interactions, as well as Rayleigh and Compton scattering [8].

One limitation of both PIXE and XRF is that they will only provide elemental information and no molecular information. Whilst the accuracy of hand-held pXRF devices has been acutely debated amongst archaeologists [5, 6, 9], the popularity of these portable devices clearly demonstrates the benefits of in situ elemental measurements [4].

Pigment Elements

Rock art paint chemical compositions, can vary widely across different countries and regions. Throughout this section, the bolded elements indicate elements that could be optimally detected by PIXE, and may be missed by pXRF.

Much Australian rock art contains ochre, which is a mixture of minerals including iron oxide and clay [10]. Iron oxides, such as haematite (Fe2O3) and goethite (FeOOH), mixed with other minerals, create some of the characteristic reds, yellows and oranges in Australian rock art. White pigments can be comprised of Kaolinite ($\mathbf{Al}_2\mathbf{Si}_2O_5(OH)_4$), huntite ($Ca\mathbf{Mg}_3(CO_3)_4$), gypsum ($Ca\mathbf{S}O_4{\cdot}2H_2O$) and/or calcite ($\mathbf{Ca}CO_3$). And black pigments are often derived from charcoal, but also manganese dioxide ($\mathbf{Mn}O_2$) in certain parts of Australia [11]. Black pigments found in prehistoric art in France consist of three main oxides: $\mathbf{Mn}O_2$, Fe_2O_3 and BaO [12, 13].

Radio Frequency Quadrupoles

In very rough terms, a radio frequency quadrupole (RFQ) is a resonant structure, loaded with four capacitive elements or vanes, that operates in a tailored TE_{210} mode [14]. A modulation along those vanes produces an alternating gradient with varying period that provides both acceleration

---

[*] tessa.charles@liverpool.ac.uk

[†] a.castilla@cern.ch

and transverse focusing to the particles. The optimisation of the vane modulation responds to a complex problem that involves, not only the rf fields handling and the proper phase advance for the acceleration, but the transverse focusing and the space-charge effects accompanying the high intensity low energy particle beam as well.

RFQ technology, capable of accelerating protons to 2 MeV over 1 meter, could provide a compact source for PIXE analysis [15–17]. CERN has invested in developing RFQ technology for the Linac4 upgrade (350 MHz) [18], which also lead to a 750 MHz RFQ structure being developed for proton therapy and other industrial applications [19]. The MACHINA (Movable Accelerator for Cultural Heritage In-situ Non-destructive Analysis) project, which is a collaboration between CERN and INFN, will use 750 MHz RFQ structures for a dedicated PIXE accelerator that will be based in INFN, Italy [15, 16].

MAIN CHALLENGES

There are several challenges involved in designing a portable PIXE accelerator for rock art studies. Many of these challenges are already being address by the MACHINA project [15, 16]. However, there are some additional challenges posed by this study, in particular relating to beam scanning and energy variation for depth-profiling.

One of the main requirements for this accelerator is portability. In order to achieve a compact and relatively lightweight accelerator, the energy has been limited to 2 MeV. As the accelerator will operate in remote areas, powered by a generator, power consumption needs to be optimised.

Some of the main challenges include:

- Raster-scanning capability over large area.
- Uneven rock surfaces, or uneven ground.
- Ensuring operation below radiation damage limits.
- Minimising power consumption.
- Operation in remote, rural sites.
- Radiation safety considerations.
- Variable energy for depth profiling.

The following subsections expand on these challenges.

Scanning

One of the biggest challenges is to design a system that can scan rock art whilst not adding greatly to the overall size and weight of the accelerator. To begin with, we aim to develop a system capable of scanning a 30 cm x 30 cm area.

Raster-scanning could be envisioned through fast scanning magnets and moving the accelerator in stages - similar to the combined beam scanning and patient table movement used at some proton therapy centres. Mechanical maneuverability of the accelerator is a significant technical challenge, however the benefits of elemental mapping allowed by scanning the beam are predicted to be immense.

Uneven Rock Surface

Uneven rock surface morphologies could make it challenging to maintain constant close distance between detector and rock surface. A larger distance between the detector and the rock surface can result in lower energy X-rays being absorbed by the air and not detected. Therefore it is important to keep this distance as short and constant over the scan.

Some rock surface will be ineligible for this technique, having uneven rock surfaces. For other cases where the rock surface height varies by only a few centimeters, a mechanical stage that moves in x, y, and z could be used to ensure a constant distance is maintained. One proposed solution is to use a laser scanner to measure the distance from the accelerator and detector, to the rock surface, and with each scan step, adjust the accelerator/detector position in z.

Radiation Damage Potential

Ensuring the incident proton beam stays safely below radiation damage limits, is of utmost concern to ensure the rock art is protected. The large X-ray emission cross-sections, mean that low beam currents (typically in the order of tens of pA) can be used, which allows for fast measurements and limited risk of radiation damage. Another factor that reduces the risk of radiation damage is the use of an extracted beam, which allows for more efficient heat dissipation than what can be achieved under vacuum.

Cost and Power Consumption

Wangler [20], gives a rough way to estimate the total cost C of a simple linac, in terms of the effective capital cost per meter C_L and the capital cost per Watt of RF power C_P. After some simple arguments, where the cost estimates are treated as function of both the required gradient E and linac length L, Wangler arrives to a minimum cost estimate of $C_{MIN} = C_P\,(2P_S + P_B)$, where: P_S is the total resistive losses in the accelerating structure and P_B the power delivered to the beam. The beam power is determined by the required energy gain ΔW, in our case ≈2 MeV. Then:

$$P_B = \frac{I\Delta W}{q} = I \times (2MV)\,, \tag{1}$$

with I as the total beam current, and q the particle's charge. Typical peak currents for PIXE are about 200 nA, hence $P_B \approx 400$ mW, which is negligible with respect to the resistive losses, which are defined as:

$$P_S = \frac{2\pi f U}{Q}, \tag{2}$$

where f is the RF frequency, for a given gradient, the stored energy is $U \propto f^{-1}$ and the 3D quality factor $Q \propto f^{-1/2}$. Then, the dissipated RF power $P_S \propto f^{1/2}$. If we consider that the RF power cost does not scale with frequency for the frequency range we are looking at (e.g. $C_P = 2$ USD/W, as proposed by Wangler), we can say that the machine cost scales as $f^{1/2}$. Therefore, moving from 750 MHz to 1.5 GHz, given the same beam specifications, we would expect roughly a 42% cost increase, without considering any extra cost derived from other constraints, such as tighter manufacturing tolerances, etc. Continuing with our simplistic exercise, and taking the power loss of 65 kW for a 750 MHz RFQ from Pommerenke, et al. [16], we can then expect, for a 1.5 GHz structure, a power dissipation in the order of ≈92 kW. It is interesting to note that recent advances in the kW-class solid state power amplifiers at 1.5 GHz [21], could be combined as power sources for a high frequency RFQ.

Another potential benefit to explore, by going to higher frequencies, is the fact that the Kilpatrick limit (E_K) increases with frequency (see Fig. 1), therefore allowing for higher peak surface fields, which could permit smaller apertures, up to beam dynamics constraints.

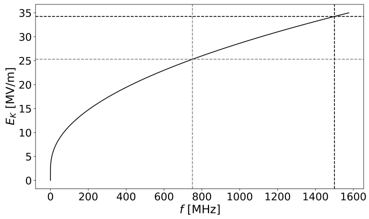

Figure 1: The Kilpatrick limit provides an empirical guide to how the tolerable peak surface fields scale with frequency.

Variable Energy Operation

One advantage offered by PIXE over XRF, is the ability to take measurements at various depths into an object by varying the proton energy. This is known as differential PIXE. Increasing the proton energy, we can probe further into the sample, and by comparing the X-ray spectra acquired from different proton energies, we can infer qualitatively how the elemental composition varies with depth (up to 100 µm - 200 µm) into the sample [22, 23].

One of the major limitations of pXRF analysis of rock art is that the irradiated volume includes the paint pigments and the rock substrate underneath. Much of the X-ray spectra detected is from the underlying rock. Attempts to subtract the substrate contribution from the pigment spectra have had limited success due to the non-homogeneity of the rock [24].

The use of close-coupled RFQ cavities has been implemented with up to 96% transmission [25, 26], where proton energy variation is obtained by adjusting the relative phase between the RFQ sections. Once the desired energy range is defined, it is possible to optimise the relative section's gain, to allow for the variability at an optimal total length.

Other Considerations

Accessibility of sites: We plan to use this accelerator in remote areas, there are some sites that are not accessible by road and would therefore be inappropriate for this type of measurement. Nevertheless, there are numerous sites that are accessible, have suitable space in front of the rock art, and would benefit from this technique.

Radiation safety: Radiation safety is a major challenge, although not technical in nature, and will require significant diligence. Various RFQ accelerators shrewdly design the accelerator so that longitudinal acceptance causes particles to be lost only at low energies [27]. Nevertheless, the extracted beam creates a radiation safety concern when used in open (unshielded) environments. When in operation, exclusion zones will most likely be set up with laser tripwires interlocked with the proton source, such that if the exclusion zone were to be breached (by person or animal), the proton source would turn off.

Detectors: PIXE is a multi-elemental technique - meaning we can detect the presence of elements within the ranges specified earlier, at the same time. One or more energy-dispersive detectors, typically Silicon Drift Detectors (SSD), are often optimised for a specified X-ray energy range.

CONCLUSION

This paper lays out the motivation and reasons why the development of a portable PIXE system is of potential interest to both the accelerators community (due to the inherent challenges, such as the complex optimisation of a high frequency RFQ to provide the proton beam requirements with a compact powering system) and to archeologist and Indigenous communities (due to the advantages offered by a powerful non-destructive in-situ analysis tool). We have included a non-exhaustive list of challenges and some simplistic arguments that are meant as conducting lines to address these challenges, rather than to solve them.

The aim of this exercise, far from providing definitive answers, is to probe the idea amongst the community, identify potential issues, as well as potential collaborators.

REFERENCES

[1] T. Johansson, R. Akselsson, and S. Johansson, "X-ray analysis: elemental trace analysis at the 10–12 g level", *Nucl. Instr. Meth.*, vol. 84, pp. 141–143, 1970. `doi:10.1016/0029-554X(70)90751-2`

[2] L. M. Brady and R. G. Gunn, "Digital Enhancement of Deteriorated and Superimposed Pigment Art:Methods and Case Studies", in *A companion to rock art*, Oxford, UK: Blackwell Publishing Ltd., 2012, pp. 625-643. `doi:10.1002/9781118253892.ch35`

[3] K. Janssens *et al.*, "Use of microscopic xrf for non-destructive analysis in art and archaeometry", *X-Ray Spectrometry*, vol. 29, pp. 73–91, 2000. `doi:10.1002/(SICI)1097-4539(200001/02)29:13.3.CO;2-D`

[4] G. Mauran *et al.*, "First in situ pxrf analyses of rock paintings in erongo, namibia: results, current limits, and prospects", *Archaeological and Anthropological Sciences*, vol. 11, pp. 4123–4145, 2019. `doi:10.1007/s12520-019-00787-7`

[5] J. Huntley and C. Galamban, "The Material Scientific Investigation of Rock Art", in *Palaeoart and materiality: the scientific study of rock art*, Oxford, UK: Oxuniprint, 2016, pp. 41-57.

[6] C. W. Koenig *et al.*, "Portable x-ray fluorescence spectroscopy of pictographs: A case study from the lower pecos canyonlands, texas", *Archaeometry*, vol. 56, pp. 168–186, 2014. `doi:10.1111/arcm.12060`

[7] M. S. Shackley, *Encyclopedia of global archaeology*, New York, USA: Springer, 2014.

[8] R. Cesareo *et al.*, "Portable Systems for Energy-Dispersive X-Ray Fluorescence Analysis of Works of Art", in *Portable X-ray Fluorescence Spectrometry: Capabilities for In Situ Analysis*, London, UK: The Royal Society of Chemistry, 2008.

[9] I. Liritzis and N. Zacharias, "Portable XRF of archaeological artifacts: Current research, potentials and limitations", in *X-Ray Fluorescence Spectrometry (XRF) in Geoarchaeology*, New York, USA: Springer, 2010, pp. 109–142.

[10] M. Elias *et al.*, "The colour of ochres explained by their composition", *Materials Science and Engineering B: Solid-State Materials for Advanced Technology*, vol. 127, no. 1, pp. 70–80, 2006. `doi:101016/jmseb200509061`

[11] B. H. Stuart and P. S. Thomas, "Pigment characterisation in australian rock art: A review of modern instrumental methods of analysis", *Herit. Sci.*, vol. 5, p. 10, 2017. `doi:10.1186/s40494-017-0123-8`

[12] L. Beck *et al.*, "Pixe characterisation of prehistoric pigments from abri pataud (dordogne, france)", *X-Ray Spectrometry*, vol. 40, no. 3, pp. 219–223, 2011. `doi:10.1002/xrs.1321`

[13] L. Beck *et al.*, "First use of portable system coupling x-ray diffraction and x-ray fluorescence for in-situ analysis of prehistoric rock art", *Talanta*, vol. 129, pp. 459–464, 2014. `doi:10.1016/j.talanta.2014.04.043`

[14] A. M. Lombardi, "The Radio Frequency Quadrupole (RFQ)", *CAS - CERN Accelerator School : small accelerators*, pp. 201–207, 2006. `doi:10.5170/CERN-2006-012.201`

[15] S. Mathot *et al.*, "The CERN PIXE-RFQ, a transportable proton accelerator for the machina project", *Nucl. Instrum. Meth. B*, vol. 459, pp. 153–157, 2019. `doi:10.1016/j.nimb.2019.08.025`

[16] H. W. Pommerenke *et al.*, "RF design studies on the 750 MHz radio frequency quadrupole linac for proton-induced x-ray emission analysis", *Phys. Rev. Accel. and Beams*, vol. 22, p. 052003, 2019. `doi:10.1103/PhysRevAccelBeams.22.052003`

[17] H. W. Pommerenke *et al.*, "RF Design of a High-frequency RFQ Linac for PIXE Analysis", in *Proc. 29th Linear Accelerator Conf. (LINAC'18)*, Beijing, China, Sep. 2018, pp. 822–825. `doi:10.18429/JACoW-LINAC2018-THPO058`

[18] O. Piquet *et al.*, "The RF Design of the Linac4 RFQ", in *Proc. 1st Int. Particle Accelerator Conf. (IPAC'10)*, Kyoto, Japan, May 2010, paper MOPD027, pp. 738–740.

[19] M. Vretenar *et al.*, "A Compact High-Frequency RFQ for Medical Applications", in *Proc. 27th Linear Accelerator Conf. (LINAC'14)*, Geneva, Switzerland, Aug.-Sep. 2014, paper THPP040, pp. 935–938.

[20] T. P. Wangler, *RF Linear Accelerators 2nd ed*, United States: Wiley, 2008.

[21] A. V. Smirnov *et al.*, "Progress on 1.5 GHz Multi-kW CW Amplifier", in *Proc. 9th Int. Particle Accelerator Conf. (IPAC'18)*, Vancouver, Canada, 2018, pp. 3821–3823. `doi:10.18429/JACoW-IPAC2018-THPAL073`

[22] P. A. Mandò *et al.*, "Differential pixe for investigating the layer structure of paintings", *Nucl. Instrum. Methods*, vol. 239, pp. 71–76, 2005. `doi:10.1016/j.nimb.2005.06.181`

[23] I. Brissaud, G. Lagarde, and P. Midy, "Study of multilayers by pixe technique. application to paintings", *Nucl. Instrum. Methods*, vol. 117, pp. 179–185, 1996. `doi:10.1016/0168-583X(96)00272-8`

[24] M. Gay *et al.*, "Efficient quantification procedures for data evaluation of portable x-ray fluorescence – potential improvements for palaeolithic cave art knowledge", *J. Archaeol. Sci. Rep.*, vol. 10, pp. 878–886, 2016. `doi:10.1016/j.jasrep.2016.06.008`

[25] R. W. Hamm *et al.*, "Characterization of a Variable Energy Deuteron RFQ System for Neutron Production", in *Proc. 19th Int. Linac Conf. (LINAC'98)*, Chicago, IL, USA, 1998, TH4106, pp. 1010–1012.

[26] C. B. Franklyn, "Utilization of variable energy radio-frequency quadrupole linear accelerator systems," in *Intl. Top. Mtg. on Nc. Res. Apps. and Util.of Accs.*, Vienna, 2009, AT: IAEA.

[27] A. M. Lombardi *et al.*, "Beam Dynamics in a High Frequency RFQ", in *Proc. 6th Int. Particle Accelerator Conf. (IPAC'15)*, Richmond, VA, USA, May 2015, pp. 2408–2412. `doi:10.18429/JACoW-IPAC2015-WEYB2`