EXAMINATION OF FLINTS BY INELASTIC SCATTERING OF PROTONS

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Inelastic scattering of protons is a recent technique which uses a proton beam from an accelerator to measure the amounts of various light clements, being mainly useful as far as silicon (Sippel and Glover 1960).

This study is an attempt to compare flints from the proposed Chowilla dam area with those found in various areas associated with Mt. Gambier.

Principle

When a proton strikes a nucleus and is reemitted with low energy, the nucleus remains in an excited state for less than 10^{-12} s, and on its return to the ground state emits gamma rays, the energy of which is only a function of the isotope involved. Thus two isotopes of the same element will emit different gamma rays. These gamma rays can be detected with suitable instrumentation and used to give information about the chemistry of the sample.

The different isotopes have different energy levels, and some, which are low, are well suited for analytical work, since it requires little energy to excite a nucleus sufficiently for them to be occupied. Another factor requiring consideration is the means of excitation. Gamma rays, atomic particles, and in some cases even energetic X-rays can be used for excitation, but only atomic particles, like protons, deuterons and alpha particles give a radiation background sufficiently low to be analytically useful if trace analysis is being considered. Protons are best for this work, because other particles tend to react with the nucleus in different ways, giving other gamma ray peaks which confuse the analysis.

Protons have a charge of +1, and considerable energy is required to make them strike the nucleus, which is also charged. This problem becomes greater the heavier the nucleus, and it will be seen that this type of analysis, for a fixed proton energy, will be largely limited to the light elements.

Other features of the analysis include its relatively non-destructive nature (since the nuclei all fall back to the ground state they originated from) and the fact that the technique is essentially limited to surfaces. This arises because the protons are very easily stopped by a thin layer of atoms. In the case of the 3 MeV beam of protons used in this work, they have a range of approximately 40 microns in the average siliccous material. This places stringent demands on homogeneity of specimens or demands a number of replicate analyses on different parts of the sample.

Procedure

Specimens were mounted on aluminium backing plates and subjected to a beam of about 0.5 microamp of protons for eight minutes. The energy of the beam was 2.95MeV and its diameter (controlled by magnetic lenses) was kept large (0.5 cm) to cut down the influence of sample variability. The samples were irradiated in a vacuum of 10^{-4} torr.

Gamma rays from the specimen emerged from the sample chamber through a mylar window and were detected by a lithium-drifted germanium detector of volume 5cc. An Ortec preamplifier and 410 amplifer were used to shape the pulse that resulted and the gamma ray spectrum was stored in a 4096 channel analyser, divided into eight 512-channel segments. All elements were hence recorded simultaneously. A pulser was set to produce pulses which were fed into the detector system and back into the spectrum. The peak that resulted was used for electronic stabilisation of the system, and simultaneously afforded a method of allowing for deadtime in the electronics.

The current of protons reaching the electrically isolated target chamber was converted to pulses (at the rate of 10^{-8} coulombs per pulse) by an Ortec current digitiser and the number of pulses recorded in a scaler. The output from this, the time taken, and the number of pulses led into the detector system from the pulser were automatically punched out onto paper tape at the end of each run.

The derived spectra were dumped electronically from the memory of the multichannel analyser to a PDP-8 computer and the areas under the observed peaks calculated via programmes written for the purpose. The paper tape was also fed in, and the spectra were all normalised to constant current reaching the target. This procedure has better than 0.1%accuracy.

U.S. Geological Survey Standard rocks were used as standards for the irradiation. The supplied powders were compressed under 25 tons to a self-supporting slab, and from the known analyses it was possible to derive, by the plotting of calibration curves, absolute analysis figures for the flints. The individual points on the graphs were shown reproducible to a few percent.

Silicon did not yield a meaningful calibration eurve, showing a vastly increased yield of gamma rays with content of silicon. It is in any case a major element and would presumably not show very great changes from one sample to another.

Samples Analysed

The Mount Gambier Limestone in SE. South Australia has a large quantity of good quality flint. This was utilized by the Aboriginals, who traded it over hundreds of miles. It is not uncommon in N. Victoria, but its distribution suddenly ceases at the Murray River valley tract. The tribes associated with this valley between Mildura in Victoria and Renmark in S. Australia did not use this flint, but preferred the local common opal. Thus, during the Chowilla Project of the National Museum of Victoria (Melbourne), only one flint artefact was found among the thousands that were examined. The present test was applied to see if it could be proved that the flint of this artefact came from Mt. Gambier area. Marine fossils in the flint showed that this could be so.

- Reg. No. M14458A Location: Port Macdonnell, 26 km S. of Mt. Gambier, S. Australia.
- Reg. No. M14458B Location: Port Macdonnell, 26 km S. of Mt. Gambier, S. Australia.
- Reg. No. M14458C Location: Port Macdonnell, 26 km S. of Mt. Gambier, S. Australia.
- Reg. No. M25360 Location: Port Macdonnell, 26 km S. of Mt. Gambier, S. Australia.

These flints samples were from the mineralogical collection of the National Museum of Victoria, and were kindly supplied by Dr. A. W. Beasley.

Reg. No. X34381 Portland (collected 1927).

- Reg. No. X76430 Lake Bonney, S.A. (Collected 1969).
- Reg. No. X76431 Blackfellows Caves, S.A. (Collected 1969).
- Reg. No. X76433 Cape Banks, S. A. (Collected 1969).
- Reg. No. X76434 L. Bonney, S.A. (Collected 1969).

| Sample No. | F(ppm) | Na(ppm) | Li(ppm) | A1 (as % A1 ₂ 0 ₈) |
|--|---|---|--|---|
| Chowilla M14458A M14458B M14458C X34381 X76430(i) X76430(ii) X76434 X76431 X76433 | $210 \pm 100 \\ 16 \cdot 3 \pm 2 \cdot 5 \\ 413 \pm 16 \\ 23 \cdot 0 \pm 4 \cdot 1 \\ 59 \cdot 1 \pm 2 \cdot 6 \\ 17 \cdot 75 \pm 0 \cdot 05 \\ less than 7 \\ less than 7 \\ 14 \cdot 4 \pm 2 \cdot 6 \\ 18 \cdot 6 \pm 1 \cdot 6 \\ 16 \\ 16 \\ 16 \\ 16 \\ 16 \\ 10 \\ 10 \\ $ | $\begin{array}{c} 630 \pm 40 \\ 542 \pm 29 \\ 4800 \pm 80 \\ 270 \pm 9 \\ 341 \pm 14 \\ 710 \pm 40 \\ 1270 \pm 40 \\ 3090 \pm 100 \\ 376 \pm 14 \\ 0.049 \pm 0.003 \end{array}$ | less than 1 less than 1 $0 \cdot 2 \pm 0 \cdot 6$ less than $0 \cdot 2$ $1 \cdot 0 \pm 0 \cdot 12$ $0 \cdot 2 \pm 0 \cdot 3$ $0 \cdot 19 \pm 0 \cdot 33$ less than $0 \cdot 9$ $0 \cdot 001 \pm 1 \cdot 10$ less than $0 \cdot 1$ | $\begin{array}{c} 0.23 \pm 7 \\ 0.239 \pm 0.058 \\ 0.89 \pm 0.11 \\ 0.084 \pm 0.016 \\ 0.135 \pm 0.023 \\ 0.066 \pm 0.027 \\ 0.187 \pm 0.06 \\ 0.38 \pm 0.17 \\ 0.059 \pm 0.023 \\ 0.081 \pm 0.008 \end{array}$ |

These flint samples were from Aboriginal implements in the National Museum of Vietoria, and were kindly supplied by Mr. A. L. West.

Results

The results are reported in general as the mean of a number of determinations on the sample. The error quoted is the standard deviation of the mean (σ/\sqrt{v}) where sigma is the standard deviation of distribution and v is the number of determinations. Only the presented elements gave measurable analysis figures.

For sample X76430(i) and (ii) the two results are from areas 1 cm apart and show, for the fluorine figures especially, the extreme variations that may be encountered. It is of interest that the next tabulated figures for X76434 are from the same area (Lake Bonney). The material is quite heterogeneous even on a maeroseopic scale. To demonstrate this more fully, the results below are those obtained 0.5 em apart on a traverse aeross specimen M14458C.

This is why a large number of results must be averaged to obtain a reliable figure.

Conclusions

It is elear that if the Chowilla flint is eompared with a selected flint from Mt. Gambier it could be considered as significantly different, but when the overall range is considered, the hypothesis that the Chowilla flint originated in that area is perfectly consistent with the data, although its fluorine content is on the high side of the distribution.

It is not possible from the above data to state definitely where the Chowilla flint originated, assuming it is indeed from the area represented by the other specimens, but it would seem that the Blackfellows cave area is most unlikely in view of the fact that that specimen alone (X34381) has a significant amount of lithium in it.

The severe heterogeneity of the flint material submitted is in marked eontrast to obsidian material from various places in New Zealand (Coote et al.). In the latter ease obsidian may remain homogeneous for a distance of several kilometres. This is probably because a voleanie melt is significantly more likely to be mixed than a sedimentary material.

References

- SIPPEL, R. F., and GLOVER, E. D. 1960. Sedimentary rock analysis by charged particle bombardment. *Nucl. Instr. Methods* 9: 37-48.
 COOTE, G. E., WHITEHEAD, N. E., and McCALLUM,
 - COOTE, G. E., WHITEHEAD, N. E., and MCCALLUM, G. J. A rapid method of characterising obsidian specimens by inelastic scattering of protons. In press. Jl. Radioanalyt. Chem.

| Position | F(ppm) | Na(ppm) | Li(ppm) | A1 (as $\% A1_20_3$) |
|----------------------------|---|---|---|---|
| 1 2 3 4 5 6 | $\begin{array}{c} 41 \cdot 6 \pm 7 \cdot 3 \\ 19 \cdot 8 \pm 7 \cdot 9 \\ 19 \cdot 3 \pm 7 \cdot 9 \\ 11 \cdot 2 \pm 7 \cdot 8 \\ 21 \cdot 2 \pm 7 \cdot 0 \\ 25 \cdot 3 \pm 8 \cdot 4 \end{array}$ | $275 \pm 23 260 \pm 30 270 \pm 30 250 \pm 30 250 \pm 20 310 \pm 30$ | less than 0.2 less than 0.23 0.1 ± 0.2 0.1 ± 0.2 0.17 ± 0.20 0.15 ± 0.28 | $\begin{array}{c} 0.096 \pm 0.039 \\ 0.119 \pm 0.043 \\ 0.011 \pm 0.043 \\ 0.063 \pm 0.042 \\ 0.102 \pm 0.039 \\ 0.111 \pm 0.055 \end{array}$ |