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Analysis and profiling of the light element isotopes; a new approach using non-elastic recoil following heavy ion nuclear reactions

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March 1980

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Analysis and profiling of the light element isotopes;
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ion nuclear reactions

T W Conlon and D J Parker

ABSTRACT

A new method is proposed and described for the analysis and profiling of the light element isotopes ^1H through ^{19}F . The method is based on the detection of recoil products following nuclear reactions in which the product yield is constrained to lie in a forward cone of half angle θ_{crit} , whose value is defined by the reaction kinematics. The main features which set apart non-elastic recoil profiling from other methods are : it is non-destructive; while an accelerator must be used initially, by choosing to observe radioactive products analysis can be done off-line; the whole element profile is given by a single irradiation which possibly might be done in air; it has potential for multi-element analysis/profiling.

Suitable reactions are identified for the analysis and profiling of each of the main isotopes of the light elements and a number of them discussed in detail. Features affecting the profiling depth, depth resolution and sensitivity are discussed.

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1. Introduction

Light elements, such as hydrogen, oxygen, carbon, nitrogen, are common contaminants present in the near surface of materials and yet are notoriously difficult to detect and profile by conventional techniques e.g. Rutherford backscattering (RBS) Auger Spectrometry, SIMS, etc. Other methods, such as the use of ion beams and the observation of prompt nuclear reaction products (charged particles, gamma-rays, X-rays), are often employed ^(1,2). These methods involve the 'on-line' use of accelerators often for a considerable period of time, since in general the depth profile must be built up in steps thus requiring corresponding changes in the ion beam energy. Sputter profiling ⁽³⁾ in which a low energy ion beam is used to erode the substrate followed by mass spectrometry of the eroded products, is often used but considerable difficulty is experienced in establishing a depth scale.

Conventional ion beam activation, i.e. using light ions, is potentially an attractive alternative which allows a sample to be treated in a relatively short time and analysed off line (albeit crudely) by sectioning or sputter profiling. However in general light ions such as p, d, ^3He , ^4He do not produce suitable activation products in undergoing nuclear reactions with the interesting light elements, defined here as those elements from ^1H to ^{19}F . Activation by heavy ion beams, which has been proposed ⁽⁴⁾ for the analysis of H, D, is considered unsuitable for profiling owing to the kinematic recoil effects which destroy the correlation between the initial profile and final activated product distribution. While this is certainly true in general, in what follows we consider these recoil effects and show that under certain conditions this correlation can be preserved, leading to a new method of profiling described here for the first time. These conditions are quantified and shown to be realisable for all light elements considered; specific reactions for the profiling of each of the main isotopes of the light elements of interest are identified. This new method has the particular merit that it is non-destructive, sectioning of the target being unnecessary since the profile information is contained in the energy spectrum of recoiling product

nuclei. Moreover the whole profile is available in a single bombardment. The method lends itself to simultaneous multi-element (more accurately, multi-isotope) analysis and can provide absolute bulk concentrations as well as profile information. It may be possible to use it in air or poor vacuum conditions thus avoiding the complexity of vacuum chamber operation, where sample integrity allows.

It is seen that, while we emphasise here long-lived radioactive products (for off-line analysis), the new method, which we call recoil activation profiling or recoil implantation profiling (depending on whether or not a radioactive product is produced), can also be used and may find more general use for on-line (prompt) analysis. Nor is the method limited to light element profiling (although this may be the most interesting application); intermediate and heavy elements can also in principle be profiled.

2. Method

The method is closely related to indirect recoil implantation which has recently been described by Conlon ⁽⁵⁾. Figure 1 (a) illustrates the method in its simplest form. A suitably thin target containing the element(s) to be profiled is bombarded with a high energy heavy ion beam chosen to yield a charged particle reaction product (which can be, but is not necessarily, a radioactive product). A suitable product in this sense is one for which

(1) A large production cross-section exists.

(2) A critical angle, θ_{crit} , exists.

It is shown in Section 3 below that at the critical angle

(a) A relatively large fraction of the total channel yield is concentrated.

(b) The recoil energy is uniquely related to the incoming primary beam energy and hence there is the possibility of obtaining depth information by observing the energy or range of the recoil reaction product.

The detection system is placed to accept the reaction product in a relatively narrow angular range close to the critical angle. For a radioactive product the system could be (Figure 1 (a)) an energy dispersive device with a catcher plate for later positional analysis which might be

preceded by a capacitance focussing device (as suggested by Asher et al ⁽⁶⁾); where only crude profiling is required a set of catcher foils (with the additional attendant energy straggling) could be sufficient. For a prompt reaction product a particle identifying system based on a ΔE -E counter telescope or magnet and focal plane detector would be required.

It is clear that for a thin target the energy, E , at which the reaction product emerges will be directly related to the original position, x , in the sample provided that, for the substrate of interest, either

$$(i) \quad (dE/dx)_{ion} \gg (dE/dx)_{recoil} \text{ or}$$

$$(ii) \quad (dE/dx)_{ion} \ll (dE/dx)_{recoil}$$

When (iii) $(dE/dx)_{ion} \sim (dE/dx)_{recoil}$ the emerging product has the same energy independent of its position of formation and no depth information can be obtained. Note that for optimum depth resolution, in case (i) the interesting surface should face the beam; in case (ii) the interesting surface should be remote from the beam.

Figure 1 (b) shows the more typical situation where the near surface of a solid object is to be examined. The primary ion beam is incident at an angle $\alpha < \theta_{crit}$ and the reaction products can be examined on-line by counter telescope or off-line as described above. For a thick target profiling information can also be obtained in case (iii) above. In this case depth resolution decreases with depth in sample due to straggling and multiple scattering of the recoil product and kinematic dispersion (see Section 4.3).

For either geometry the method can in principle yield absolute values for the total amount of element isotope as well as the depth profile provided the reaction cross-section is known. In a practical application a large area of the substrate may have to be profiled; this can be achieved by use of a diffuse beam (where angular definition allows, see Section 3.2.1) or by scanning the substrate in the directions indicated in Figures 1 (a), 1 (b). Where lateral resolution is required a microbeam might be used although there are relatively few such systems available at

present which provide the beams and energies required ⁽⁷⁾.

While we concentrate here on the general case of recoil following nuclear reactions, i.e. non-elastic recoil, the special case of elastic recoil has been discussed by Terrault et al ⁽²⁾ and independently and with different emphasis by Conlon ⁽⁸⁾. This special case also has considerable potential for multi-element profiling when used in conjunction with a mass analysis system ⁽⁸⁾ and a brief description is included below (Section 4.4) for completeness.

3. Theory

3.1 Basic Theory

Consider the reaction $A(a,B)b$ where A is the element of interest, a the primary beam species, B is the recoil of interest, b the associated residual product and A, B, a, b also denote the masses of those species.

The recoil velocity of B in the laboratory frame, $\underline{V}_{lab}$ can be decomposed as follows (see Figure 2 (a)):

$$\underline{V}_{lab} = \underline{V}_{c.m.} + \underline{V}_r$$

where $\underline{V}_{c.m.}$ is the velocity of the centre of mass frame and $\underline{V}_r$ the recoil velocity of the particle in the centre of mass. $|\underline{V}_{c.m.}|$ is determined by the primary beam energy and the masses of the target and ion beam; $|\underline{V}_r|$ is independent of the centre of mass angle and depends only on the initial and final masses, the beam energy E_{in} and reaction Q -value ⁽⁹⁾.

It is straightforward to show, for the most general case where $|\underline{V}_r| > |\underline{V}_{c.m.}|$ corresponding usually to a lighter mass ion beam and product and a heavier mass target material, that the reaction product B occurs at backward as well as forward angles and that the total cross-section is spread out over the whole reaction plane.

However it is possible to choose conditions such that $|\underline{V}_r| < |\underline{V}_{c.m.}|$; in this case the reaction product B is confined to a forward cone whose half angle, θ_{crit} , is given by

$$\sin^2 \theta_{crit} = \frac{Ab}{aB} \left\{ 1 + \frac{Q}{E_{c.m.}} \right\} \quad (1)$$

This is the case illustrated in Figure 2(a). Note that for each value of

θ_{lab} there are two values of $\phi_{c.m.}$, the centre of mass angle of B, and of $\underline{V}_r$ and hence $\underline{V}_{lab}$. It is this second case, usually corresponding to the observation of the heavier mass product ($B > b$) and often corresponding to a heavy mass ion beam incident on a lighter mass target ($a > A$), which is of interest for present purposes and which leads to the new method of profiling discussed here.

Figure 2 (b) shows the distribution of recoil particle energy as a function of laboratory angle, θ , for reactions which have a critical angle. For each value of $\theta < \theta_{crit}$ there are two recoil energies, E_1 and E_2 , and the function is single-valued (value E_{crit}) only at θ_{crit} . The plot of c.m. angle, ϕ , against θ_{lab} is similar to Figure 2 (b).

Consider now the yield in the laboratory frame. It can be shown (see Appendix and ref. 10) and indeed it is clear from Figure 2 (b) that the cross-section in laboratory coordinates will have a maximum close to θ_{crit} . Moreover the yield of implants against angle is weighted by a factor $\sin \theta_{lab}$ owing to the solid angle; thus the yield at zero degrees vanishes. Assuming for the moment an isotropic cross-section in the centre of mass, Figure 2 (d), the general shape of the yield in the laboratory frame will be as shown in Figure 2 (c). Indeed for an isotropic cross-section this curve is no more than the ratio of solid angles $d\Omega_{lab}/d\Omega_{c.m.}$ multiplied by $\sin \theta_{lab}$. It is therefore clear that no matter what the shape of the c.m. angular distribution, the yield in the laboratory frame will always reach a maximum close to θ_{crit} . (Indeed neglecting straggling and finite beam energy spread etc. it diverges at θ_{crit}). In order to focus more clearly here on the basic principles of the new method of profiling the detailed kinematical arguments, giving analytical expressions for laboratory energies, yields, etc., are described in a separate report (10). However a short list of the most useful expressions is given in the Appendix.

It is fortuitous that a large proportion of the yield occurs near the critical angle, since at θ_{crit} the recoil energy is single valued and uniquely related to the primary beam energy thus providing an ideal basis

for element profiling with high efficiency. Moreover a necessary condition for the existence of a critical angle ($aB > Ab$) guarantees a large recoil velocity at θ_{crit} . (However $dE/d\theta$ is maximised at θ_{crit} and this adversely influences depth resolution as discussed in Section 3.2.1).

At θ_{crit} :

$$E_{recoil} = \frac{aB}{(a+A)^2} (1 - \sin^2 \theta_{crit}) E_{in} \quad (2)$$

Thus by observing the yield of a suitable reaction close to θ_{crit} there are two principal possibilities for profiling as outlined in Section 2, viz.

$$(i) \quad (dE/dx)_{ion} \ll (dE/dx)_{recoil}$$

The energy of the recoil is directly related to the depth of the trace element in the substrate through the application of the stopping power equation. The energy of the emerging recoil ion can be measured directly in a particle identification system or indirectly by measuring the range or energy distribution of a radioactive product.

$$(ii) \quad (dE/dx)_{ion} \gg (dE/dx)_{recoil}$$

In this case the beam energy, E_{in} , decreases sharply with depth x of the element to be profiled; this leads to a reduced E_{recoil} through Equation (2). The energy loss of the emerging recoil can be measured as in (i).

In the general case it is always true that the energy of the emerging recoil decreases as the depth profiled increases, through a combination of the above effects; and so the method is still applicable. However in the discussion which follows we find it helpful to concentrate on the two extreme cases in which the depth information is respectively provided by (i) the energy loss of the emerging recoil or (ii) the energy loss of the ingoing ion.

3.2 Refinements to Basic Theory

The basic theory outlined above neglects the following important features which influence the sensitivity, depth resolution and profiling depth which can be achieved by this new method, viz.:

- (a) Variations in θ_{crit} due to changes in E_{in} as the beam penetrates the target.

- (b) Reactions leading to other states (different Q values).
- (c) Non-isotropic angular distributions.
- (d) Energy straggling in the target of the primary beam and outgoing observed product.
- (e) Multiple scattering of the primary beam and observed product.

The first three of these (a), (b), (c) are specific to a given ion beam/element combination and are considered in general terms here and in particular cases in Section 4.

The features (d) and (e) are important in all ion beam techniques and indeed determine the ultimate limits of sensitivity in this as in other ion beam profiling methods. (d) leads to a statistical spread in the energy of the emerging particles which smears out the depth resolution. (e) leads to uncertainty in the angle of emission which influences both the observed energy and the yield at a given angle. A comprehensive description of straggling and its influence on ion beam techniques is given by Deconninck ⁽¹¹⁾. The consequences of multiple scattering for recoil techniques are discussed by Conlon ⁽⁵⁾. No detailed description of these effects will be given here. It should, however, be pointed out that the magnitude of both effects increases as the M, Z of the projectile and observed product increase and decreases with increasing energy. So for a 'heavier' light element there is a considerable premium to be gained by operating at high energies.

Furthermore the consequences of multiple scattering for the technique described here are likely to be more severe than in other ion beam techniques since the energy of the observed product changes rapidly with angle near θ_{crit} . The practical consequences of this are being investigated currently ⁽¹²⁾.

(a) (b) and (c) are discussed separately below.

3.2.1 Variations in θ_{crit}

It can be seen from Equation (1) that θ_{crit} changes as E_{in} decreases with penetration of the beam into the substrate. The way in which it changes depends on the sign of the Q-value. This has two consequences as far as the proposed method is concerned. First, it leads to variations in yield to the

detector placed at a fixed angle; and second, it can be seen from Equation (2) that E_{recoil} is no longer strictly proportional to E_{in} if θ_{crit} changes. The effect of these changes can fortunately be minimised in a number of ways.

(1) It is minimised by choosing to observe states corresponding to $E_{\text{c.m.}}$

$\gg Q$ for the state to be observed, so that the term $Q/E_{\text{c.m.}}$ is always small compared to unity.

(2) The effect can be eliminated altogether for many of the cases discussed below in Table 1 by choosing to observe reaction products corresponding to $Q \approx 0$ (excited states of the product nucleus). Indeed many of the reactions listed in Table 1 and discussed below are expected to have much higher yields near $Q \approx 0$ than for the ground state for intermediate bombarding energies.

(3) Clearly ensuring that the beam energy loss in the target is small, i.e. $(dE/dx)_{\text{ion}} \ll (dE/dx)_{\text{recoil}}$, also minimises variations in θ_{crit} . This is usually the case for the reactions in Table 1.

The angular acceptance of the detector, $\Delta\theta$, (in the absence of straggling and multiple scattering) determines the depth resolution achieved. It can be seen from Figure 2 that, if the detector accepts recoils with a range of angles from θ_{crit} down to $\theta_{\text{crit}} - \Delta\theta$, the observed particles correspond to a range ΔE of recoiling energies. For small θ_{crit} ,

$$\frac{\Delta E}{E_{\text{crit}}} \sim \sqrt{32 \theta_{\text{crit}} \Delta\theta} \quad (3) \text{ (ref 10)}$$

(angles measured in radians)

For depth resolution in the measurement we require that this intrinsic energy spread, ΔE , be smaller than the variations in energy loss due to the differences in path length that we are trying to measure. This can only be achieved with a very small angular acceptance, $\Delta\theta$.

Moreover there is a direct trade off between resolution and yield, since for a given reaction the yield of recoils to the detector decreases as $\Delta\theta$ is reduced. In fact for the case of an isotropic angular distribution in the centre of mass the fraction of the total yield to angles

$$\theta > \theta_{\text{crit}} - \Delta\theta \text{ is : } \frac{\Delta\sigma}{\sigma} \sim \sqrt{\frac{2\Delta\theta}{\theta_{\text{crit}}}} \quad (4) \text{ (ref 10)}$$

(approximations valid for $\Delta\theta \ll \theta_{\text{crit}} \ll 40^\circ$)

Note from Equations (3) and (4) that both depth resolution and yield are optimized for a given $\Delta\theta$ by choosing reactions with small θ_{crit} . However, there are practical problems associated with the small grazing angles of incidence involved in using the geometry of Figure 1 (b) for a small critical angle.

The variation in θ_{crit} caused by changes in $E_{\text{c.m.}}$ effectively sets a limit to the range of depths which can be examined. When $\Delta\theta_{\text{crit}}$ corresponding to $\Delta E_{\text{c.m.}}$ exceeds the angular acceptance of the counter no events from the channel of interest are observed. The angular acceptance is effectively set by the depth resolution as discussed above; the depth resolution and profiling depth can be optimised by using the criteria outlined there. For all of the reactions in Table 1 one or more of these criteria can be used to minimise variations in θ_{crit} .

3.2.2 Reactions leading to other states

Equation (1) refers only to 'two-body' final state reactions (so that the energy of the recoil is uniquely defined at the critical angle) and indeed only for a specific final state of the particles B, b (unless $Q \approx 0$). In many cases of practical interest (see Table 1 below), b, in Equation (1) is a 'fundamental' particle, e.g. p, n, α , for which excited states lie at very high energy and are not populated and so can be neglected in the present discussion. B, however, is often a light or medium weight nucleus with excited states below 1 MeV and the reaction of interest will contain some contribution from events such as $A(a, B^*)b$ where the reaction proceeds via an excited state B^* other than the one of interest, which then decays by γ -emission $B^* \rightarrow B + \gamma$. In this case the appropriate Q-value must be used in Equation (1) and the additional recoil effects due to γ -ray emission need to be considered.

Both factors lead to variations in θ_{crit} . Fortunately the effect can be minimised by defining an angle window as in Figure 1 (a) and 1 (b) such that only products in a narrow cone, corresponding to a fixed Q or small range of Q , are examined; or as discussed in Section 3.2.1 above. (For elastic recoil discussed below it can always be considered negligible at energies of practical interest).

3.2.3 Non-isotropic angular distributions

The vast majority of angular distributions are not isotropic as considered above. A quite common angular distribution (c.m.) ⁽¹³⁾ can be approximated, particularly for particles emitted from reactions following compound nucleus formation, by $\frac{d\sigma}{d\Omega} = \frac{\gamma}{\sin \phi}$ where γ is a constant. Unfortunately since this diverges at 0 and 180° it is not a useful function for present purposes. The combination of an isotropic angular distribution and one of the form $\cos^2 \phi$, i.e. $\frac{d\sigma}{d\Omega} = \alpha + \beta \cos^2 \phi$ where α, β are constants and $\beta \gg \alpha$, is a good approximation to $\frac{\gamma}{\sin \phi}$ apart from very small and very large angles. In this section we consider the profiling possibilities with such a distribution. Having already dealt with the isotropic case we now consider only the second term: $d\sigma/d\Omega = \beta \cos^2 \phi$, as shown in Figure 3 (a).

Corresponding to Figure 2 (c) in the isotropic case, in the non-isotropic case the recoil yield versus laboratory angular distribution is shown qualitatively in Figure 3 (b). Note that $\phi_{\text{crit}} = \pi/2 + \theta_{\text{crit}}$.

The general shape of the yield curve shown in Figure 3 (b) is similar to Figure 2 (a) in that it has a maximum at θ_{crit} but exhibits a minimum at θ_{min} and another maximum at a smaller angle. The most important features for our present purposes are:

- (a) a unique energy is still associated with the critical angle,
- (b) there is still a peak in the yield curve at θ_{crit} although the usable yield will be smaller than for the isotropic case. Indeed most of the reactions in Table 1 are expected to be of this type (at least when averaged over a suitable number of excited states). However it should be feasible to use a number of excited states close to $Q \approx 0$ in order to alleviate the effect of the low yield in any given state at θ_{crit} .

4. General Discussion

4.1 Non-elastic recoil

We note that, leaving aside the term involving Q which can be either positive or negative, Equation (1) gives the required condition, $\sin \theta_{\text{crit}} \leq 1$, only for $Ab \leq aB$ and this is satisfied in general only for a heavy ion incident on a lighter mass element i.e. $a > A$, and/or where B is greater than b . However the Q -value also enters in Equation (1) and because any particular reaction has its own Q -value we need to consider each individual case separately. A number of particular examples for practical purposes are given in Table 1 and these best illustrate the general case. The important special case of $\theta_{\text{crit}} = 90^\circ$, as for elastic recoil, is considered separately below in Section 4.4.

Table 1, column 1 lists a number of reactions and radio-isotope products which can be readily produced and which are suitable, in the terms outlined in Section 2, for recoil activation profiling. Column 2 gives the Q -value and energy threshold for the particular reaction. The primary beam energy quoted first in column 3 is arbitrarily chosen as the maximum energy for a given beam from the Harwell 6 MV Tandem accelerator; it can of course be lower. It can also be much higher, e.g. by employing the Harwell 70" variable energy cyclotron (VEC). For the heavier recoil species there are considerable advantages in terms of depth resolution, profiling depth and yield to be gained at the higher energies available from a variable energy cyclotron. There are, however, drawbacks associated with activation of the substrate at higher energies. In order to give a more complete picture of the usefulness of the new technique, Table 1 includes the pertinent information for the higher energy beams available, for example, from the Harwell VEC. Also included in column 3 are the critical angle, the recoil energy at the critical angle and the recoil range in an iron substrate. Note that the recoil range is defined along the path of the recoiling particle and is not the same as depth in the sample. The two are related by a factor depending on θ_{crit} and α in Figure 1. In transmission geometry, Figure 1 (a), the maximum depth of profiling is $R_{\text{crit}} \cos \theta_{\text{crit}}$.

In each case in Table 1 the reaction channel defined is expected to be (one of) the dominant channel(s) at the energies considered; these reactions have been selected to illustrate a number of features of the process: viz.

1. Suitable reactions can be found for all the light elements (although in Table 1 only long lived radioactive products are listed; the Table could be made much larger if non-radioactive products or short lived isotopes were to be included). Note that in general only two-body final state reactions are suitable. Three body reactions cause a smearing of the angular distribution and the expected maximum in the yield-angle curve at the critical angle, and on which the technique depends, no longer necessarily occurs.
2. The critical angle varies widely depending on the reaction; in the examples it ranges from 5.3° to 40.6° . (It is always 90° for elastic recoil).
3. In general the lighter the recoil product, the wider the range of depth for profiling.
4. The critical angle and the energy of recoil at the critical angle can be controlled by varying the primary beam energy only. Reference to Equation (2) shows that reducing the beam energy reduces the recoil energy. Reducing the beam energy can cause either an increase or decrease in the critical angle depending on the sign of the Q-value.
5. The angular acceptance of the detector determines the depth resolution and observed yield for the recoil species as discussed in Section 3.2.
6. Both isotropic and non-isotropic angular distributions are involved e.g. the $H(^7\text{Li}, ^7\text{Be})n$ reaction is known to be approximately isotropic, near $E_{\text{cm}} \sim 3 \text{ MeV}$ ⁽¹⁴⁾. Most of the others are expected to approximate to the form $\alpha + \beta \cos^2 \theta$. The angular distribution for $^{12}\text{C}(^{14}\text{N}, ^{22}\text{Na})\alpha$ is known to be of this form ⁽¹⁵⁾. Note that not all excitation functions for the reactions are readily available. Indeed the measurement of these would be a considerable research programme in itself if they cannot be gleaned from the literature. In the discussion below we concentrate on those for which

angular distribution information is readily available or predictable.

4.2 Examples of non-elastic recoil

It has already been pointed out that while Table 1 shows possible profiling reactions for the element isotopes from ^1H to ^{19}F , only reactions leading to long lived radioactive products are included, in order to keep this discussion to manageable proportions. It should also be noted that the beam energies quoted are arbitrarily chosen as the maximum energies available for the Harwell Tandem and VEC accelerators. They do not always correspond to the energy range over which a particular reaction can be used for profiling but are quoted mainly to illustrate the potential depth range (R_{crit}) which might be achieved by the method and to give representative values for parameters such as θ_{crit} , E_{crit} and their range of variation.

We now consider a number of reactions in some detail. In what follows we shall neglect the possibility of competing reactions, i.e. reactions from elements other than the one of interest which lead to the same final nucleus. This is currently under investigation ⁽¹²⁾ but outside the scope of the present report.

4.2.1 Hydrogen Profiling

The reaction $\text{H}(^7\text{Li}, ^7\text{Be})\text{n}$ is a good example which illustrates the potential of the new method in the particular case of hydrogen profiling. The requisite nuclear data information is available at least at low energies ⁽¹⁾. At a ^7Li beam energy of 24 MeV (3 MeV c.m.) the cross-section is known to be close to isotropic and the treatment of Section 3.1 applies. Table 1, line 1 indicates the potential of profiling to depths of up to 11.6 mg/cm^2 in an iron substrate (15 μm) at a critical angle of 5.6° . This reaction is currently being studied for recoil implantation and profiling potential ⁽¹²⁾ as well as for bulk activation ⁽⁴⁾.

At higher energies (e.g. 108 MeV or 13.5 MeV c.m. as from the Harwell VEC) the potential profiling depth is up to 120 mg/cm^2 (150 μm) in iron. Nuclear data at higher energies indicate approximate isotropy up to 15 MeV c.m.

The reaction $H(^{10}\text{B}, ^7\text{Be})\alpha$ is also suitable for hydrogen profiling. The potential range of profiling depths is similar to the $^7\text{Li} + \text{H}$ reaction at Tandem and VEC energies. However the reaction Q-value is positive (whereas $^7\text{Li} + \text{H}$ is negative) and the critical angle is larger.

4.2.2 Other light elements

For many of the reactions listed in Table 1, cross-section information is sparse or not available at all. No attempt has been made at this stage to catalogue references containing appropriate information. There is no doubt that if the new method described here for light element profiling is to be used, a great deal of cross-section information in the form of excitation functions and angular distributions for the particular reactions will have to be either measured or preferably gleaned from the literature.

In the particular case of $^{12}\text{C}(^{14}\text{N}, ^{22}\text{Na})\alpha$, detailed measurements of angular distributions and excitation functions have been made ⁽¹⁵⁾, so we use this to discuss the general case of the light elements other than hydrogen. The angular distribution (cm) of the recoil product is close to $\gamma/\sin \phi$ when averaged over a suitable energy interval (~ 1 MeV) and this is typical of the c.m. angular distributions expected for reactions in Table 1. The laboratory angular distribution is then similar to Figure 3 (b). By choosing to observe states close to $Q \simeq 0$:

(a) The yield from a relatively large number of final states can be used to determine the profile. (The yield near $Q \simeq 0$ is anyway much higher than near $Q_{\text{g.s.}}$ for the range of E_{in} of interest here, owing to the weighting described by the Statistical Model of nuclear reactions given to the higher spin states at higher excitation energies, corresponding to more negative Q values).

(b) θ_{crit} is independent of E_{in} .

4.2.3 Simultaneous profiling of the light elements

The reaction $^{12}\text{C} + ^{14}\text{N} \rightarrow ^{22}\text{Na} + \alpha$ was used above to illustrate the potential of the method described for carbon profiling. Clearly by reversing target and beam, the same reaction can be used for nitrogen profiling using

a carbon beam. This can be done in general without destroying the essential coning effect, whenever beam and target are comparable in mass. (It can also be done for most of the other reactions of Table 1 but in those cases where target and beam are not comparable in mass, the transfer of energy from projectile to observed product is inefficient and the observed product emerges at low energies - unsuitable for profiling).

In Table 2 we break down column 1 of Table 1 in order to show which beams are useful for profiling each light element. (Once again one must emphasise that we restrict the discussion to radioactive products; otherwise Table 2 would be much larger). The possibility of multi-element profiling is most clearly demonstrated in this table by the ${}^7\text{Li}$, ${}^{16}\text{O}$ and ${}^{19}\text{F}$ beams; viz. by use of an oxygen beam ${}^7\text{Li}$, ${}^9\text{Be}$ and ${}^{10}\text{B}$ could be simultaneously profiled by observation of the recoil product at the critical angles which are 22.7° , 28.8° and 40.6° respectively. When reactions other than those leading to radioactive final nuclei are considered, the multi-element profiling capability is vastly increased. Indeed the latter method of observing prompt reaction products would probably be the most suitable for multi-element profiling by this method since not only would θ_{crit} be different for each element but so would the final product, giving an extra degree of discrimination for different elements.

4.2.4 Light ion recoil product

In order to complete the discussion of these examples of non-elastic recoil it should be noted that for entries 1, 2 of Table 1 it is also possible to obtain depth profiling information by observation of the light reaction product. Table 3 gives the appropriate information for the light product, as Table 1 does for the heavy product. The light product (n and α respectively in 1, 2) must be observed under prompt reaction conditions (since they are not radioactive) but their energy distribution contains the same information as the heavier product.

In the case of 1, viz. $\text{H}({}^7\text{Li}, {}^7\text{Be})\text{n}$, the depth resolution is expected to be better since there is no inherent energy straggle for the neutron

exit channel and the depth information comes from the stopping power for the ${}^7\text{Li}$ beam.

In the case of 2, viz. $\text{H}({}^{10}\text{B}, {}^7\text{Be})\alpha$, the depth resolution and accessible depth range are determined by the incident beam and the improvement in resolution with respect to the heavier mass case is expected to be marginal.

4.3 Limits of sensitivity

Without a particular profiling problem in mind, it is difficult to evaluate the ultimate sensitivity of the proposed new method for light element profiling; however some general comments on sensitivity in the two different modes of detection by which the technique can be used are given.

4.3.1 Radio isotope (off-line) detection

In all of the cases listed in Table 1, the final product is the only long lived (half-life greater than a few days) radio isotope available in that part of the nuclide chart. Even if activation of the substrate cannot be avoided (indeed it is probably unavoidable at VEC energies) the long lived products of the substrate, which could also recoil into the catcher foils, should be sufficiently different from the light element recoil products for high selectivity to be achieved.

McGinley ⁽⁴⁾ has studied bulk activation of hydrogen and deuterium by reactions 1, 2 and 3 in Table 1 and has demonstrated the feasibility (at cyclotron energies) of detecting hydrogen in titanium at a level of 30 ppm with a precision of 10%; the detection limit is quoted as approximately 0.5 ppm. In the new method of depth profiling suggested here, there is a direct trade off between observed reaction yield and resolution as discussed in Section 3.2.

In addition to the other considerable potential advantages to profiling by recoil activation as discussed in Section 4.5, it is appropriate to mention here that with development it might lend itself to analysis in air or poor vacuum conditions. For bulky objects or those of complex geometry, irradiation by a beam of charged particles brought

through a thin window into air or a poor vacuum would offer considerable advantages to processing in a beam line/scatter chamber arrangement.

4.3.2 Prompt (on-line) detection

There is clearly an enormous increase in efficiency to be gained by the direct detection of individual recoil particles as distinct from waiting for their radioactive decay, so that while most of the discussion above is dedicated to profiling by observation of radioactive recoil products (with the associated advantages of off-line processing) the method lends itself to on-line processing either by direct observation of individual radioactive nuclei (by means of the reactions listed in Table 1) or direct observation of individual non-radioactive products of reactions analogous to those listed in Table 1. In both cases a mass sensitive particle identification system is required as discussed in Sections 2 and 4.

The sensitivity in this case is determined by the relative magnitude of the 'signal' to the 'noise' generated by other reactions. Identification of individual mass nuclei in the presence of large quantities of background from other masses is a phenomenon commonly encountered in nuclear reaction studies. Conlon ⁽⁵⁾ has pointed out the considerable selectivity afforded by observing a particular reaction product close to the critical angle for the two body channel of interest. Since each channel has its own particular θ_{crit} the effect of other channels, can be greatly reduced.

Experiments aimed at quantifying and comparing the relative usefulness of each method of detection are at present under way ⁽¹²⁾.

4.4. Elastic recoil

In contrast to the non-elastic process whose use in element profiling is described above for the first time, the related process of elastic recoil has also been proposed ^(2, 8) and used ^(2, 16, 17) for element profiling. First of all we consider the generic relationship between the two processes before commenting on the usefulness of the elastic recoil method.

In the case of elastic recoil instead of the general form $A(a,B)b$ the reaction has the form $A(a,A)a$ with Q equal to zero and so θ_{crit}

from Equation (1) is 90° . * Figure 4 illustrates the kinematics of this particular case as Figure 2 did for the general case. Figure 4a shows aspects of the centre of mass situation; in this case $\bar{V}_{cm} = \bar{V}_{lab}$. Figure 4b shows the energy spectrum of recoil atoms as a function of laboratory angle, θ .

Figure 4c shows the yield as a function of laboratory angle; in this case the yield is more strongly concentrated near the critical angle than in the general case because of the strong dependence of the cross-section. For elastic recoil the reaction cross-section in the centre of mass cannot be taken to be isotropic or of the form $\gamma/\sin \phi$, but follows the well known Rutherford formula at sufficiently low energies, i.e. it is proportional to $1/\sin^4 (180-\phi)/2$ where ϕ is the centre of mass angle of the recoiling nucleus (Figure 4d). In this case no depth information would be gained by observing the recoil product at θ_{crit} since $\theta_{crit} = 90^\circ$ and E_{crit} from Equation (2) is zero. However, contrary to non-elastic recoil, in the elastic recoil case the recoil energy and c.m. angle are single valued so that there is no intrinsic energy spread at angles other than θ_{crit} . At very forward angles, detection is complicated by primary beam effects (ref. Conlon (5) and Section 3.2) hence an intermediate angle $0^\circ \ll \theta \ll 90^\circ$ is appropriate.

A most interesting feature of elastic recoil for element profiling is that it can in principle permit the simultaneous profiling of all elements in a substrate (8). This is possible since the information in Figure 4 applies to each and every recoil species; i.e. for elastic recoil Equation (1) does not contain the recoil mass explicitly. Not only has the yield curve the same shape (apart from a scaling factor, see Appendix) for all recoils but so has the energy distribution. At θ_{lab} :

$$E_{recoil} = E_o \cos^2 \theta_{lab} \text{ where } E_o = \frac{4Aa E_{in}}{(A+a)^2} \quad (5)$$

Profiling information can be obtained in the same way as for non-elastic recoils - namely by utilising the observed or apparent energy loss of the

* Inelastic scattering $A(a,A')a$ is a non-elastic process where A,A' differ only in excitation energy. Since Q is negative, A' has a critical angle $\theta_{crit} < 90^\circ$.

recoil species owing to primary beam energy loss or recoil product energy loss respectively. In order that a common depth scale be applicable to all recoil products, it is preferable to use a primary beam whose stopping power is much greater than that of any of the species to be profiled.

In practice the recoil product is observed in a particle detector with small angular acceptance $\Delta\theta$ at an arbitrary angle θ chosen to obtain suitable range and resolution. The general considerations of Sections 2 and 3.1 apply in this case.

Terrault et al⁽²⁾ have shown the potential for hydrogen profiling and for the simultaneous detection of a number of light elements using an incident ³⁵Cl beam. Ref. 2 also contains useful source references. Conlon⁽⁸⁾ has suggested the use of elastic recoil in conjunction with a Buechner magnet/PSD system as a mass identifier to provide multi-element profiling information. Parker et al⁽¹⁶⁾ have investigated the profiling of C,H in plastic foils using a number of heavy ion beams in conjunction with the Buechner magnet/PSD system⁽¹⁶⁾; Sofield et al⁽¹⁷⁾ have used this system for profiling hydrogen in solid electrolytes.

It should be noted that in common with the non-elastic recoil method, the elastic recoil method is likely to be limited in depth resolution by multiple scattering effects.

4.5 Comparison with other ion beam methods

In comparing the recoil methods which are non destructive with conventional techniques, in order to compare like with like we shall exclude techniques such as SIMS which involve erosion of the material, and microbeam techniques which involve sectioning. Currently used NDT ion beam profiling techniques (in what is a rapidly growing area) have been summarised by Bøttiger⁽¹⁾ for hydrogen and helium isotopes and by Terrault et al⁽²⁾ for light elements up to $A \lesssim 10$. Most of these (reactions with light ions, resonant reactions, forward scattering coincidences) are specific to a particular element and all require the use of an accelerator on-line. The latter is also true of elastic recoil which is described in those reviews

and covered here in a different way owing to its close relationship to the proposed new method of non-elastic recoil.

The main features which set apart non-elastic recoil profiling from the other methods are

- (a) No sectioning of the target is required.
- (b) Like elastic recoil the whole profile is given in a single bombardment by energy analysis of the recoil product(s).
- (c) It can be used off-line from an accelerator (radioactive products) although its major use could well be on-line.
- (d) It has potential for use in air or poor vacuum conditions and hence on real objects as distinct from small samples.
- (e) It may be applied to single elements as well as to multi-element profiling (off-line radioactive products).
- (f) As an on-line method it could have multi-element analysis potential.
- (g) Used in conjunction with a microbeam there is the prospect of lateral resolution (from microbeam scanning) as well as depth resolution from recoil profiling.

5. Conclusion

A new method is proposed and described for the analysis and profiling of the light element isotopes ^1H through ^{19}F . The method is based on the detection of recoil products following nuclear reactions in which the product yield is constrained to lie in a forward cone of half angle θ_{crit} , whose value is defined by the reaction kinematics. The main features which set apart non elastic recoil profiling from other methods are: it is non-destructive; while an accelerator must be used initially, by choosing to observe radioactive products analysis can be done off-line; the whole element profile is given by a single irradiation which possibly might be done in air; it has potential for multi-element analysis/profiling.

Suitable reactions are identified for the analysis and profiling of each of the main isotopes of the light elements and a number of them discussed

in detail. The depths accessible for profiling depend upon the choice of accelerator used and in an iron substrate range up to 15 μm for H, 1 μm for ^{19}F with a 6 MV Tandem accelerator and to 150 μm for H, 5 μm for ^{19}F with a 70" sector focussed cyclotron. In the case of the proposed technique there is a trade off between depth resolution and sensitivity. In general resolution is limited by multiple scattering and deteriorates with penetration depth and as the mass of element to be profiled increases - for a given accelerator facility.

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TABLE 1
Examples of non-elastic recoil profiling

(1) REACTION	Q	(2) E _{th} (MeV)	E _{in} (MeV)		θ _{crit} (degrees)	(3) E _{crit} (MeV)		R _{crit} mg/cm ² Fe	
			Tan	VEC					
1. H(⁷ Li, ⁷ Be)n	-1.64	13.1	24	108	5.6	18.2	81.2	11.6	120
2. H(¹⁰ B, ⁷ Be)α	1.15	~15.4	36	138	16.1	19.2	74.9	13	100
3. D(⁶ Li, ⁷ Be)n	3.38	~ 3.5	24	126	15.8	14.6	78.3	8.7	110
4. T(²⁰ Ne, ²² Na)n	5.02	~16.6		155	5.3	127.8			22
5. ⁴ He(¹⁹ F, ²² Na)n	-1.95	11.2	42	163	4.8	33.0	127.6	5.3	22
6. ⁷ Li(¹⁶ O, ²² Na)n	7.28	~16.0	42	193	10.2	27.1	125.6	4.6	21
7. ⁹ Be(¹⁶ O, ²⁴ Na)p	7.74	~17.4	42	193	10.9	24.9	115.5	4.2	19
8. ¹⁰ B(¹⁶ O, ²² Na)α	10.07	~20.0	42	193	25.4	17.8	87.5	3.2	14
9. ¹¹ B(¹² C, ²² Na)n	5.78	~12.5	42	106	13.4	19.8	50.4	3.5	7.8
10. ¹¹ B(¹⁴ N, ²⁴ Na)p	12.66	~15.4	42	150	13.6	21.3	77.5	3.7	12
11. ¹² C(¹⁴ N, ²² Na)α	5.62	~17.4	42	150	26.7	15.3	56.8	2.8	8.7
12. ¹² C(¹¹ B, ²² Na)n	5.78	~11.5	36	125	14.8	15.4	54.1	2.8	8.3
13. ¹⁴ N(¹¹ B, ²⁴ Na)p	12.66	~12.1	36	125	17.1	13.9	49.5	2.6	7.6
14. ¹⁴ N(¹² C, ²² Na)α	5.62	~14.9	42	106	31.0	12.1	31.0	2.5	5.3
15. ¹⁶ O(⁷ Li, ²² Na)n	7.28	~ 7.0	24	108	22.7	5.9	27.9	1.6	4.7
16. ¹⁶ O(⁹ Be, ²⁴ Na)p	7.74	~ 9.8	30	149	18.8	9.3	47.4	2.2	7.4
17. ¹⁶ O(¹⁰ B, ²² Na)α	10.07	~12.5	36	138	40.6	6.8	30.3	1.7	5.1
18. ¹⁹ F(⁶ Li, ²⁴ Na)p	13.73		24	126	33.6	3.8	24.7	1.0	4.1

TABLE 2

Examples showing the potential for simultaneous profiling
of some light elements (based on the entries of Table 1)

Ion beam	Elements profiled
${}^6\text{Li}$ ${}^7\text{Li}$ ${}^9\text{Be}$ ${}^{10}\text{B}$ ${}^{11}\text{B}$ ${}^{12}\text{C}$ ${}^{14}\text{N}$ ${}^{16}\text{O}$ ${}^{19}\text{F}$ ${}^{20}\text{Ne}$	$\text{D}, {}^{19}\text{F}$ $\text{H}, {}^{16}\text{O}, {}^{19}\text{F}$ ${}^{16}\text{O}$ $\text{H}, {}^{16}\text{O}$ ${}^{12}\text{C}, {}^{14}\text{N}$ ${}^{11}\text{B}, {}^{14}\text{N}$ ${}^{11}\text{B}, {}^{12}\text{C}$ ${}^7\text{Li}, {}^9\text{Be}, {}^{10}\text{B}$ ${}^4\text{He}, {}^6\text{Li}, {}^7\text{Li}$ T

TABLE 3

Hydrogen profiling by the non-elastic
method using the 'light' ion recoil product

(1) REACTION	(2)		(3)			
	Q (MeV)	E_{th}	E_{in} (MeV) Tan VEC	θ_{crit} (degrees)	E_{crit} (MeV)	R_{crit} mg/cm ² Fe
H ($^7Li, n$) 7Be	-1.64	13.1	24 108	42.3 69.6	1.44 1.43	
H ($^{10}B, \alpha$) 7Be	1.15	~15.4	36 138	29.1 25.9	9.1 36.9	18.4 175

APPENDIX

Kinematic results of use in recoil profiling

	Non-elastic	Elastic
c.m. Angular Distribution, $d\sigma/d\Omega$ c.m.	constant	$1.296 \left(\frac{Z_1 Z_2}{E_{in}} \right)^2 \left(\frac{M_1 + M_2}{M_2} \right)^2 \sin^{-4} \psi \frac{\psi}{2} \text{ c.m.}$ $= A \sin^{-4} \psi \text{ c.m.} / 2$
Angular distribution of yield in laboratory frame $d\sigma/d\theta$ ($K = 1/\sin \theta_{crit}$)	$\frac{1 + K^2 \cos 2\theta}{\sqrt{1 - K^2 \sin^2 \theta}} \sin \theta$	$8 \pi A \frac{\sin \theta}{\cos^3 \theta}$
Energy distribution of yield in laboratory frame, $d\sigma/dE_{recoil}$ ($E_o = \text{c.m. energy of recoil}$)	$- \frac{\pi}{KE_o} \frac{d\sigma}{d\Omega} \text{ c.m.}$	$- \frac{\pi}{E_o} \frac{d\sigma}{d\Omega} \text{ c.m.} = \frac{-16\pi A E_o}{E_{recoil}^2}$

AERE - R 9724 Fig. 1
Non-elastic recoil profiling. (a) Thin target of profile material; (b) Thick target of profile material

AERE - R 9724 Fig. 2
Non-elastic recoil profiling with isotropic ang. distribution

(a)

(b)

AERE - R 9724 Fig. 4
Elastic recoil profiling