APPLICATIONS OF CYCLOTRONS IN TECHNICAL AND ANALYTICAL STUDIES

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Abstract

The following applications of cyclotrons are briefly discussed: simulation of radiation damage in the development of reactor materials; study of wear and monitoring of machine movements in mechanical engineering; several methods of elemental analysis using cyclotron beams. The emphasis is on methods which have passed the development stage and have been applied to a number of practical problems.

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During recent years cyclotrons have found some applications to technical problems. This talk discusses a few of these in the fields of reactor materials, mechanical engineering and elemental analysis. These examples have been selected because these applications have passed beyond the stage of development of a method and take a considerable fraction of machine time of at least one cyclotron.

1. Simulation of Radiation Damage

Structural materials in advanced nuclear reactors such as the liquid metal cooled fast breeder reactor are subject to intense bombardment by fast neutrons. Typical values of the neutron flux in question are above 10^{14} n/cm^2 sec. So the neutron flux integrated over the life time of a fuel element is of the order of 10^{22} n/cm^2. Interactions between fast neutrons and the atoms of the material may considerably alter the mechanical properties. The development of alloys resistible to radiation damage is therefore one of the key problems of the development of fast reactors. Similar difficulties, though even severer, are met by the fusion reactor with its higher neutron energies. One of the problems in this field of materials research is the time required to test a material in a reactor. Today no reactors are available with a fast flux appreciably higher than that of future commercial fast reactors. Therefore the test of a new fuel cladding material, e.g., will take as much time as the fuel element is expected to stay in the reactor, i.e. typically a year. Under such circumstances, progress would not be expected to be too rapid. It is at this point that cyclotrons can make an important contribution because charged particle beams from an accelerator can cause similar damage at a much higher rate. To understand why let us consider the main primary reactions of fast neutrons with the atoms in a solid:

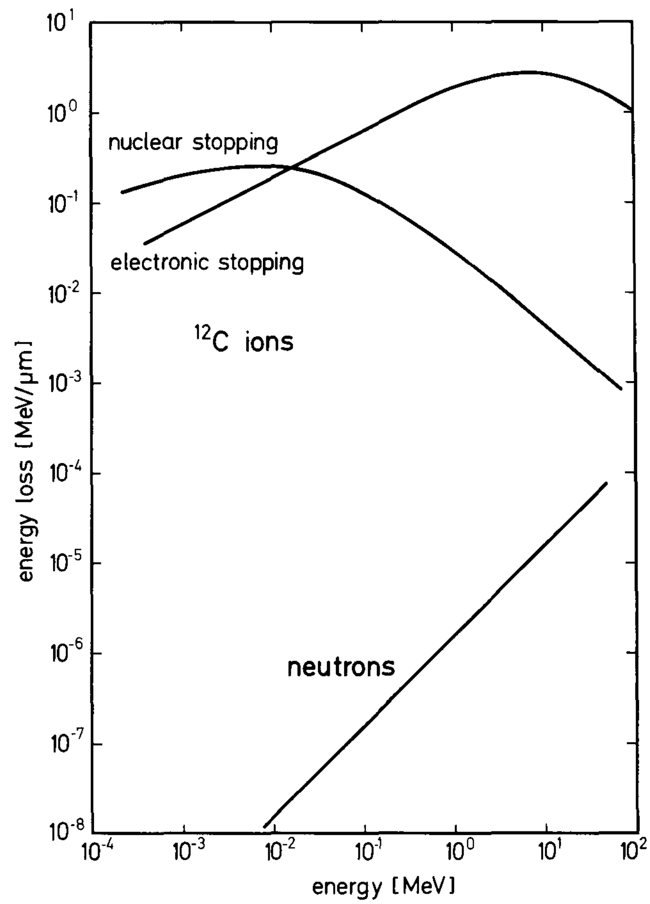

Fig. 1 Average energy loss of ^{12}C ions and neutrons in nickel. Nuclear stopping data are calculated according to Lindhard et al.[1]), electronic stopping data are from Northcliffe[2]). For neutrons, a total scattering cross section of 5 barns and isotropic scattering has been assumed.

(i) By elastic and inelastic collisions, neutrons can transfer sufficient energy and momentum to displace the atoms from their lattice site. The integrated flux of 10^{22} n/cm^2 will result in every atom to be displaced between 10 and 100 times. It is obvious that this may have considerable influence on all material properties.

(ii) Nuclear transmutations occur and will lead to a change of the chemical composition. The most harmful of these is the production of helium via (n,α) reactions because helium is not dissolved in the metal and favours the formation of small voids which leads to swelling of the

Proc. 7th Int. Conf. on Cyclotrons and their Applications (Birkhäuser, Basel, 1975), p. 496-502

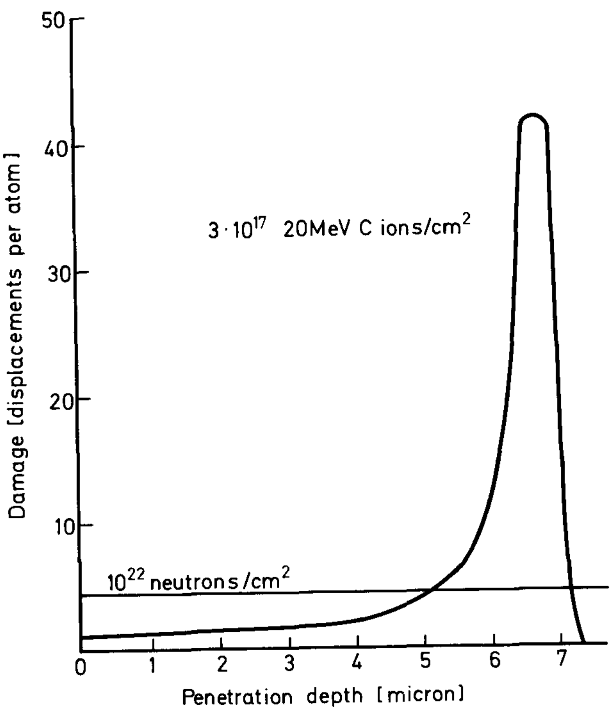

Fig. 2 Depth distribution of radiation damage by ^{12}C ions and fast neutrons in nickel. Note the difference in the number of incident particles. (Redrawn from ref. 3).

material.

Obviously atoms can also be displaced from their lattice sites by collisions with charged particles. Since charged particle cross sections are several orders of magnitude higher than those of neutrons the rate of displacement can be made much higher with charged particles and the time of experiment reduced. This is illustrated in fig. 1 which compares the average energy loss of neutrons and ^{12}C ions in Ni. While almost all of the energy lost by neutrons is transferred to atoms only the nuclear stopping part of the ^{12}C ion energy loss contributes to the displacement of atoms. Nevertheless, the difference in energy transfer is many orders of magnitude. From the same figure an important conclusion about the spatial distribution of the damage can be drawn. The fractional energy loss of neutrons is very small. Therefore most neutrons will be completely unaffected during the passage through the material and the damage will be very homogeneous. Charged particles, on the contrary, are slowed down, and due to the strong energy dependence of nuclear stopping a maximum of damage will occur near the end of the range. This is shown in detail in fig. 2. Here the depth distribution of damage for neutrons and ^{12}C ions in nickel is compared. Please note the more than 4 orders of magnitude difference in the number of incident particles for the two curves. In order to achieve a more homogeneous distribution of charged particle damage it is necessary to alter the energy of the particles incident on the probe. This is most easily done by a tapered absorber moved in front of the probe. An example of such an arrangement is shown in fig. 3. This apparatus also provides for a transverse movement of the samples for obtaining a homogeneous distribution in this direction. Under such conditions it is possible to simulate the damage corresponding to 10^{22} n/cm^2 in an irradiation time of approximately a day at a current of a few µA. The following requirements should be met for such a simulation:

(i) The range of the particles should be sufficiently high to produce a zone of damage of a few µm in depth beyond a surface zone of a few tenths of a µm.

(ii) A facility for producing damage homogeneous in three dimensions, such as shown in fig. 3, should be available.

(iii) Ions of one of the major constituents of the material under investigation should be used. Since maximum damage is obtained near the end of the

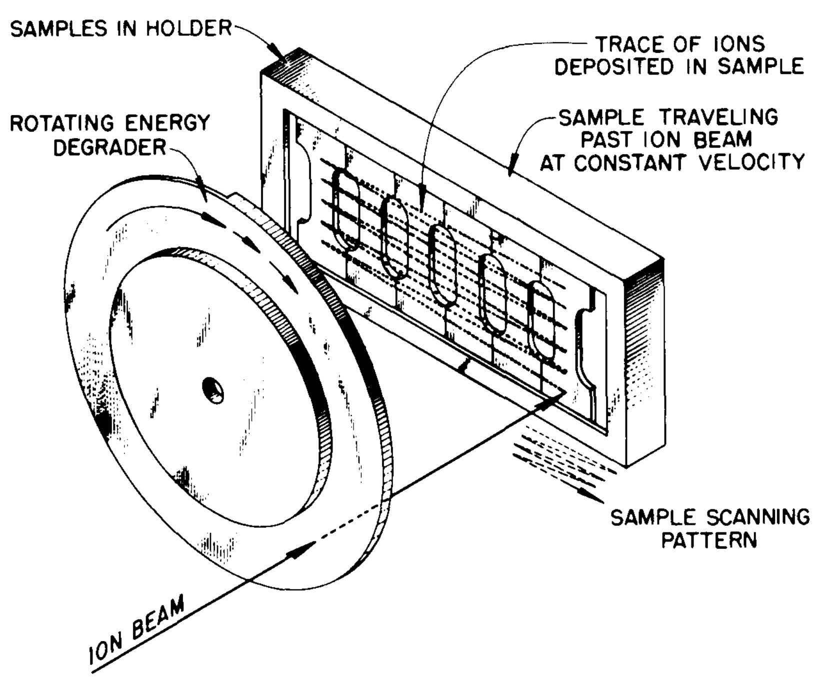

Fig. 3 Irradiation facility for obtaining a homogeneous damage distribution in three dimensions (From ref. 4).

range incident particles should be stopped in the sample and the number of particles thus implanted may be high enough to cause a disturbing change of chemical composition in that part of the sample which is subject to high damage. This has led to the development of metal ion beams at the cyclotrons at Harwell[5] and Oak Ridge[6].

Cyclotrons can also contribute to the study of the influence of helium created by (n,α) reactions on material properties. For this, helium is implanted into the material by an α particle beam either prior to a heavy ion bombardment or simultaneously. The same technique for obtaining a homogeneous distribution of helium in the sample is applied.

The importance of these simulation techniques is borne out by the fact that simulation of radiation damage takes a considerable fraction of machine time at some cyclotrons such as Harwell, Oak Ridge, and Karlsruhe, e.g.

2. Applications in Mechanical Engineering

A number of applications of cyclotron beams in mechanical engineering has been demonstrated during the last years[7]. I here want to discuss two of these, investigation of wear and measurement of the movements of such machine parts which are unaccessible during machine operation.

2.1 Wear Studies

One of the characteristic features of most radioactive material is that very small amounts can be detected easily. Since in most machines the amount of material removed by wear per unit time is very low it has been recognized very early that the use of activated machine parts could be of considerable advantage in wear studies. The basic procedure consists of activating a part, reassembling the engine and measuring the activity of the wear particles collected e.g. on an oil filter. This activity is directly proportional to the amount of material removed. In most cases γ-ray detection is applied. As we will see a few μg of wear particles can clearly be measured. This is a tremendous increase in sensitivity as compared to non-radioactive methods. A further important advantage lies in the fact that wear can be monitored continuously and no disassembly is required.

Early investigations used activation by reactor neutrons. This results in very high total activities because the part under study is activated all through. This requires heavy shielding of the test facilities and in some cases even remote handling during machine assembly. Activation by accelerator beams, in the contrary, offers the following advantages[8]:

(i) Activity is confined to the surface region and may be concentrated on those surfaces which are subject to wear. This reduces the total activity by many orders of magnitude.

(ii) Due to the low amount of activity to be handled no intricate precautions have to be taken during assembly. In many cases test facilities available in machine factories can be used without shielding.

(iii) Sufficient accuracy is obtained when measuring the residual activity on the activated part instead of the activity removed. This method, called thin layer difference method, allows the study of problems where the material worn off cannot be collected.

(iv) Different machine parts may be activated by different projectiles or at different energies. In this way different radioactive nuclides are produced which may be distinguished by the methods of γ ray spectroscopy. This allows the study of the wear of different parts of an engine in the same experiment.

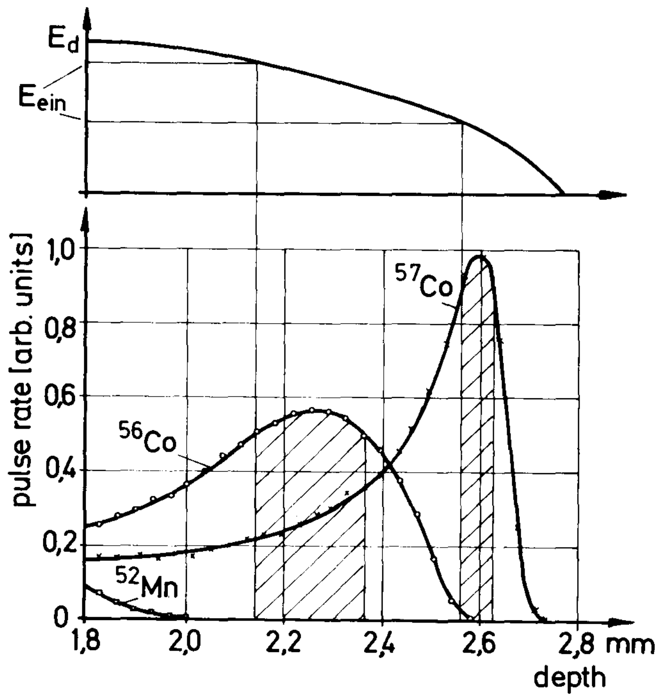

Fig. 4 Upper part: Energy versus penetration depth for a charged particle beam. Lower part: depth distribution of activity produced by 52 MeV deuterons in iron. The hatched area indicates the part of the range most useful for wear studies. The upper part indicates how the corresponding particle energy is determined.

Table 1 shows a number of nuclear reactions suitable for activating different materials. Since the reaction cross sections depend on the energy and incident particles are slowed down, a very inhomogeneous depth distribution of activity is produced. An example of this is shown in fig. 4. It is convenient to work at an energy at which the cross section does not vary too much with energy because then the relation between activity and removed material is linear. Preferably, an energy slightly higher than the maximum cross section is chosen.

Table 1 Some nuclear reactions used for wear studies of different materials

Material	Reaction	Threshold (MeV)	Maximum of cross section[a] energy (MeV)	height[b] (mb)	Properties of radioactive nuclide half life (d)	energies of main γ rays (keV)
Mg	$^{26}Mg(d,\alpha)\,^{24}Na$	0	8.3	11	0.63	2754,1369
Al	$^{27}Al(d,p\alpha)^{24}Na$	5.8	24	55	0.63	2754,1369
Ti	$^{48}Ti(p,n)\,^{48}V$	4.9	12	380	16.0	511,983,1312
Cr	$^{52}Cr(p,n)\,^{52}Mn$	5.6	12.6	500	5.7	511,1434,936,744
	$^{52}Cr(p,pn)^{51}Cr$	12.2	∿27[c]	∿500[c]	27.7	320
Fe	$^{56}Fe(p,n)\,^{56}Co$	5.4	12	410	77.3	847,1238
	$^{56}Fe(d,n)\,^{57}Co$	0	∿8[c]	∿400[c]	270	122,136
Cu	$^{65}Cu(d,2n)^{65}Zn$	4.5	15	270	244	511,1115
Zn	$^{68}Zn(p,2n)^{67}Ga$	12.2	∿12[c]	∿130[c]	3.3	93,185,299
Pb	$^{206}Pb(p,2n)^{205}Bi$	11.6	21	250	15.3	1764,703,988

a: data from Keller et al.[9] ; b: referred to natural element; c: estimated according to Keller et al.[10]

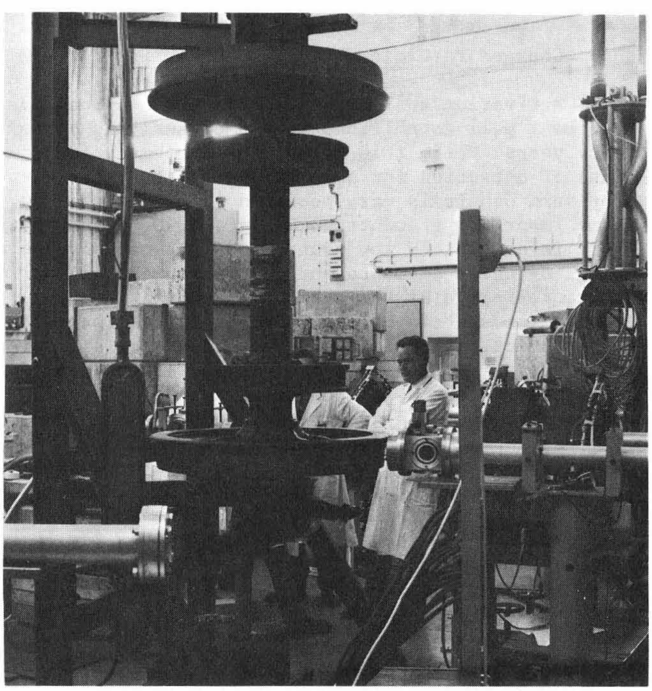

Fig. 5 A set of railway wheels being set up for irradiation at the Karlsruhe Isochronous Cyclotron.

As can be seen from table 1 this requires energies in the range 10 to 25 MeV for most reactions. Depth sensitivity can be increased by up to a factor of 10 by irradiating at a small angle between beam and target surface.

Irradiations are usually carried out in air. The beam leaves the beam pipe through a thin havar window. Currents of a few μA and irradiation times between 1 and 10 h are usually required. Fig. 5 shows a set of railway wheels being set up for irradiation at the Karlsruhe Isochronous Cyclotron. Here 20 spot irradiations were placed along a spiral around the circumference of the wheel. The wheels were then brough back in operation on the German railway, and the activity monitored at regular intervals. The total activity produced was so low that no special licence was required. This experiment would not have been possible with neutron activation.

A large number of different engineering problems has been studied by this method at Karlsruhe during the last 10 years. These include the wear of ball bearings at different loads and speeds and during running-in[10], the influence of additives to lubricants[11], influence of the shape of piston rings on wear[12] and wear properties of different materials for journal bearings[13]. Similar work is apparently going on in the USSR[14]. As an example fig. 6 shows the influence of water added to the lubricating oil on the wear of a cage of a roller bearing.

A potential application of the method is the

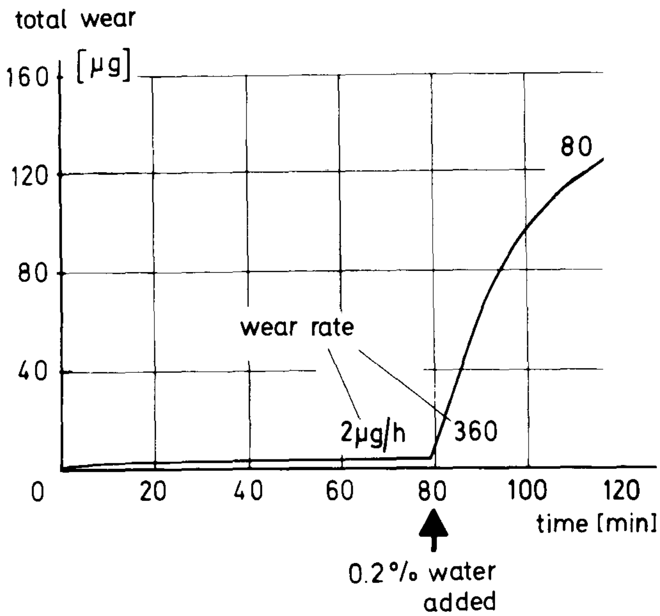

Fig. 6 Measurement of wear on the cage of a ball bearing. At the addition of 0.2 % of water to the lubricating oil the wear rate increases dramatically.

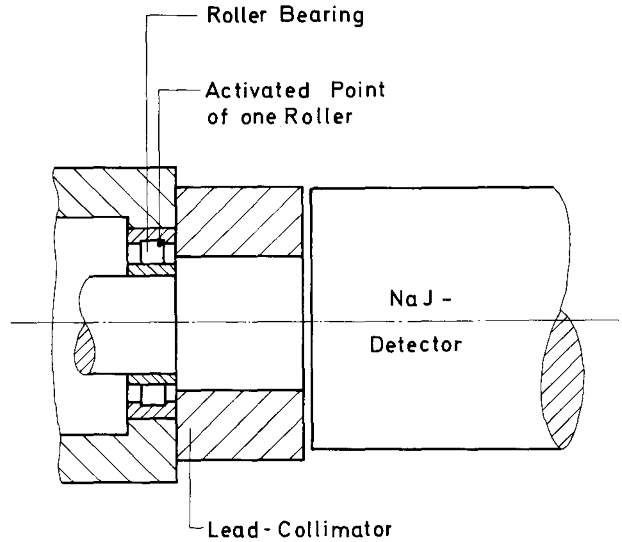

Fig. 7 Apparatus for measuring the speed of revolution of the roller of a roller bearing.

optimization of time intervals between maintenance of large machines.

This and similar work now takes about 15 % of all machine time at the Karlsruhe Isochronous Cyclotron, and this figure is still increasing. A considerable fraction of this is paid for by industry.

2.2 Monitoring of Machine Movements

Machine parts may be inaccessible or even invisible during operation. Information about details of their movements may therefore be difficult to obtain. Examples are balls or cages in ball bearings, inner parts of high speed turbines etc. One way of measuring movements of such a part is to mark a spot on it by activation and monitor the activity through an appropriately shaped collimator. An example[10] is shown in fig. 7. Here the roller of a roller bearing has been activated at the edge of a front face. The lead collimator in front of the NaJ γ-ray detector attenuates the radiation when the spot is at maximum distance from the bearing axis while there is no attenuation at minimum distance. This can give important information on slippage between rollers and cage. This apparatus has been used for speeds up to 10^5 rpm. The method can obviously be adapted to suit many similar problems.

3. Analytical Applications

In recent years a number of accelerator based methods has been proposed to determine the elemental composition of a sample. Some of these have been the subject of conferences of their own, therefore my remarks will be far from exhaustive. Before discussing some of the methods in more detail I should like to point out that some discrepancies exist in literature on the limits of sensitivity of most analytical methods. This is at least partly due to the fact that the sensitivity does not only depend on the method, but also on the composition of the sample to be analyzed, due to interference from other constituents. This should be borne in mind when considering published sensitivity data.

3.1 Charged Particle Activation Analysis[16]

Activation analysis using reactor neutrons has become a well established method of analysis since many years. While this method has extremely low limits of detection for some elements its sensitivity for some others is very poor. Among the latter are some important elements such as C, N, O, S and Pb. This is due to the low neutron capture cross sections of these elements and/or unfavourable decay characteristics of the nuclides produced. Most of these elements can be activated by charged particle bombardment, and this can be exploited for analysis in a very similar way as neutron activation. The procedure thus consists of irradiating a piece of material at an accelerator and measuring the induced activity some time after the end of irradiation. A variety of nuclear reactions has been investigated for this purpose. For a large number of elements limits of detection well below the ppm level, in some cases even below the ppb level, are quoted for this method. A very important example is the determination of trace amounts of O in several metals or semiconductor materials via the reaction $^{16}O(^{3}He,p)^{18}F$ at energies between 10 and 40 MeV. Here limits of detection between 1 and 10 ppb have been demonstrated[17].

It should be mentioned, though, that some important differences exist between neutron and charged particl activation analysis. As was discussed in section 2 the distribution of induced activity is extremely inhomogeneous for charged particles. One has therefore to rely on a homogeneous distribution of the measured element in the sample under investigation. Furthermore, the range of the particles, i.e. the part of the sample which is activated is determined by the stopping power of the material, i.e. by the main constituents. For the measurement of one trace element in different matrices therefore different calibration factors apply and have to be determined.

3.2 Fast Neutron Activation Analysis

Activation analysis using 14 MeV neutrons from D-T generators is a well established technique[18], especially for the determination of O using the reaction $^{16}O(n,p)^{16}N$. A considerable number of other elements may also be determined by this method with high sensitivity[19]. Although cyclotrons can provide a higher flux of fast neutrons from the reaction of deuterons with beryllium and, in addition, higher neutron energies these possibilities do not seem to have been exploited on a broad scale. Estimates show, however, that considerable improvements can be achieved in a number of cases[20].

3.3 Prompt Nuclear Analysis[21]

Activation analysis infers the amount of an element present from the radioactive end product of a nuclear reaction. Instead, some prompt radiation emitted during the nuclear reaction - either charged particles, neutrons, or γ rays - may be measured. Since different primary particles can be used to induce the nuclear reaction a variety of possibilities results. A large number of these has been studied in detail during the last five to ten years. If charged secondary particles are to be measured the reaction has to take place somewhere near the surface of the sample to allow the particle to escape. This method therefore is well suited to study the surface layers of a solid. Due to the energy loss of incident and secondary particles during their passage through the sample, the energy of the reaction product may indicate the depth below the surface where the reaction has taken place. This depth resolution increases with decreasing energy because of the higher specific energy loss of low energy particles. A high spatial resolution perpendicular to the beam direction may also be obtained by focusing the beam to a small diameter[22]. Here values below 5 μm have been achieved. These techniques offer quite new possibilities of measuring the spatial distribution of minor constituents on a microscopic level.

3.4 Ion Induced X-Ray Analysis[23]

Fast ions colliding with atoms excite characteristic X-rays at a cross section which is several orders of magnitude higher than that of nuclear reactions. Measurement of these X-rays can therefore be used to determine the elemental composition of a sample. This method has a very high sensitivity as compared to excitation by electrons because the continuous background of bremsstrahlung is much lower. X-rays are usually detected by solid state counters. This allows the measurement of a large number of elements simultaneously. Sensitivities are often near 1 ppm, and in favourable cases quantities below 1 ng have been detected. The inherent limitation of the method lies in bremsstrahlung produced by the exciting ions or by recoil electrons from ion-electron collisions. Due to the large cross sections only a few minutes of beam time are required at currents of the order of 0.1 to 1 μA for a complete analysis. Small diameter beams for obtaining high spatial resolution may also be used.

The method has been employed by Cahill et al.[24] at the Davis cyclotron to analyse a large number of environmental samples. This is probably one of the optimum applications of the method because the detection of a large number of elements is required in a small amount of substance, and the sample is given on a thin filter backing.

3.5 Merits of Cyclotrons for Analysis

Cyclotrons have a chance to be applied in chemical analysis if the advantages of cyclotron based methods outweigh the disadvantage of operating cost. When trying to estimate the potential of the described methods one should bear in mind that cyclotrons have to face competition in this field also from other accelerators, e.g. small Van de Graaff machines. Any general statement is made difficult by the fact that advantages and disadvantages of a method depend very much on the specific problem to which it is applied. Therefore, the following remarks should not be taken too literally.

Charged particle activation analysis typically requires energies between 10 and 40 MeV and currents of the order of μA. Therefore the use of cyclotrons is very appropriate. The method can achieve very high sensitivities and for a number of cases is not surpassed by any other, especially in the field of trace determination in high purity materials. For highest sensitivities it requires machine time of the order of hours, though.

Prompt nuclear analysis has been studied at a large number of low energy electrostatic accelerators. The following arguments indicate that these machines are well suited for this type of work:

(i) The energy of the incident particle should be kept as low as possible in order to reduce the number of different nuclear reactions. At higher energies it is difficult to identify the products of the one reaction under study unless the sample is a thin foil which might be difficult to prepare without contaminating the sample.

(ii) Depth resolution increases.

(iii) The low emittance and energy spread, especially of Van de Graaff accelerators, facilitates the production of low diameter beams.

Although it has been demonstrated that cyclotrons can yield excellent results in the field of ion excited X-ray analysis there is no considerable advantage as compared to low energy electrostatic accelerators which are probably cheaper to operate.

In my opinion, cyclotrons will therefore only find limited application in analysis. The best chances are probably given in the field of trace determination in high purity materials by charged particle activation.

4. Concluding Remarks

The examples discussed above have shown, I think, that cyclotrons find practical applications in technical and industrial development work, and I am sure further cases of such work will be found. I should like to point out, though, that any such application has to be competitive on the basis of a realistic cost evaluation. By realistic I mean including depreciation and interest. Such figures are not easily obtained. I should therefore like to quote the Karlsruhe figure of 300 $ per hour of useful machine time. This is probably not untypical for a machine of this size. My estimate of the operating cost of a compact cyclotron to be installed in Germany today is somewhere around 60 to 90 $/h. As for the examples I have discussed, the development of a new type of reactor is such an expensive project that cyclotron operating cost does not present a serious problem. For the applications in mechanical engineering work at Karlsruhe has shown that industry is willing to pay the price.

Other factors which may affect industrial applications of cyclotrons are the limited availability and reliability.

The most important point in establishing such unconventional applications of cyclotrons, however, is probably a psychological rather than a technical or economic one. It lies in identifying problems which cyclotrons can help to solve, in bringing potential users and cyclotron people together and demonstrating the merits of a proposed method. It then may happen that a non-physicist discovers that a cyclotron is not an obscure, expensive apparatus for incomprehensible high-brow research but rather an ingenious machine invaluable for solving his very problems.

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DISCUSSION

N.N. KRASNOV: Did you investigate the influence of irradiation dose on the resistance to wear?

G. SCHATZ: I do not think there have been special studies on that problem at Karlsruhe, but from the experience we have, we do not believe this will present any difficulty. This is due to the fact that radiation damage occurs mainly in a thin zone at the end of the range, well below the surface.

E.G. MICHAELIS: Can you say how charged particle or neutron activation analysis compares with elemental analysis by mu-mesic X-rays as concerns sensitivity, it being understood that muons are more expensive than protons or neutrons?

G. SCHATZ: I do not have experience with mesonic X-rays, so I do not think I can give you a definite answer. From what I recall of how mesonic X-ray spectra look like, I should not be too optimistic.