

INDUSTRIAL AND ENVIRONMENTAL APPLICATIONS OF CYCLOTRONS

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Abstract. - The application of cyclotrons in industrial development and in environmental analysis is reviewed. The simulation of fast neutron radiation damage, the activation of machine parts for wear studies, and analysis by proton induced X-ray emission (PIXE) are described in some detail. Finally some cost considerations are attempted.

1. Introduction. - This contribution deals with some industrial and analytical applications of cyclotrons. It is restricted - somewhat arbitrarily - to such techniques which have passed beyond the development stage and have found more than prototype use. Though for reasons to be discussed below the scope of this work will probably remain much more limited than the medical applications of cyclotrons the examples show that cyclotrons can be used to advantage in industrial research and development and in some cases open up unique possibilities. Similar reviews have been given at preceding cyclotron conferences (cf. refs. 1, 2, and 23).

2. Simulation of fast neutron radiation damage. - A somewhat specialized, but important industrial application of cyclotron beams is the simulation of radiation damage caused by fast neutrons in the structural material of fast breeder reactors^{3,5)}. Similar considerations hold for a prospective thermonuclear fusion reactor.

Fast neutrons interact with the atoms in a solid primarily in two ways:

- a) Atoms are displaced from their lattice sites by elastic and inelastic collisions. During the lifetime of a fuel element in a typical fast reactor this may occur up to a hundred times to every atom.
- b) Nuclear transmutations may occur which alter the chemical composition. The most important of these processes is the production of helium by (n,α) reactions.

The combined effect of these two processes is the formation of small voids. This is illustrated in figure 1. These voids lead to a volume increase of up to 30 % during the lifetime of a fuel element, and because of the inhomogeneous flux distribution this swelling is not uniform and hence may cause a considerable distortion of the reactor core.

The swelling behaviour has been found to depend on the composition and pretreatment of the material. It is therefore one of the major problems in the development of fast breeder reactors to identify materials for which swelling is reduced. Fuel elements are to stay inside a reactor for several years, therefore the radiation damage accumulated over such a period is of interest. The unique advantage of cyclotron beams in this field is the possibility to induce similar damage at a rate which is about 3 orders

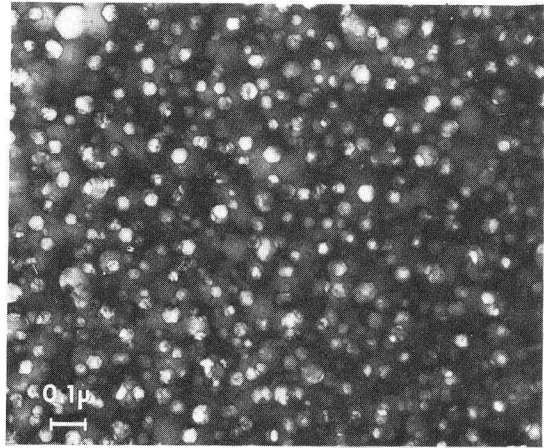


Fig. 1 : Electron micrograph of a Ni sample irradiated by 20 MeV C^{++} ions to a dose corresponding to 40 displacements of every atom (from ref. 3).

of magnitude higher than in a reactor^{3,4)}. This is demonstrated in more detail in figure 2 which compares the energy distribution of recoil atoms normalized to the number of incident particles for the elastic scattering of fast neutrons and energetic heavy charged particles. It shows that Ni ions are 3 to 4 orders of magnitude more effective in displacing atoms. Similarly alpha particles from cyclotrons have been used to implant helium in order to simulate the effects of (n,α) reactions.

In both cases homogeneous damage distributions over a depth of approximately 10 μm or more are required in order to reduce surface effects. In contrast to this charged particles show a maximum of energy loss and hence radiation damage close to the end of their range. It is therefore most effective to stop the particles in the specimen under study. This causes a change of the chemical composition which may influence the properties of the material. Therefore ions of one of the major constituents of the studied alloy are preferred. This had led to the development of Fe and Ni beams for the study of radiation damage in steel at the Harwell Variable Energy Cyclotron. The thicknesses of the specimens to be irradiated then require energies of 40 to 60 MeV. This is sufficiently low to avoid nuclear reactions, so the samples are

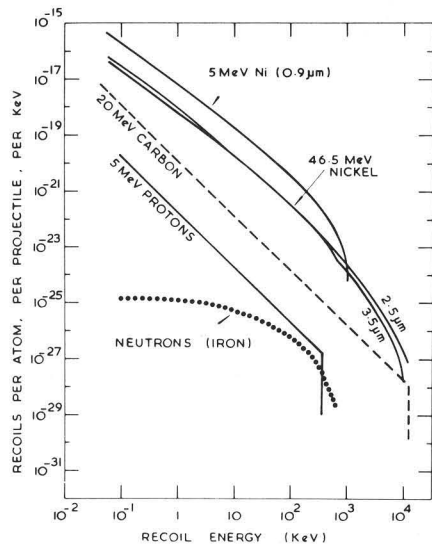


Fig. 2 : Energy distribution of recoil atoms in Ni subject to different types of irradiation. The curves are normalized to the number of incident atoms. The dotted neutron curve corresponds to a typical fast breeder neutron spectrum (from ref. 4).

hardly activated which facilitates post irradiation investigations.

A homogeneous damage distribution is accomplished by periodically changing the particle energy, usually by moving a wedge-shaped absorber in front of the irradiated sample. Ideally both types of damage should be induced simultaneously. Since this requires two different simultaneous particle beams and hence two separate accelerators most investigations have so far applied helium implantation before heavy ion irradiation.

It should be mentioned that radiation induced creep can be simulated and studied in a similar way⁵⁾.

The simulation of radiation damage by cyclotron beams and especially the unique heavy ion beams from the V.E.C. at Harwell have played an important role in the fast breeder development programs both in the United Kingdom and in the Federal Republic of Germany. The same will hold for a prospective thermonuclear fusion reactor for which the radiation damage problem is much more severe than for a fast breeder reactor.

3. Activation of machine parts for wear studies. - Charged particle beams in the energy range of 10 to 50 MeV are used very successfully in a special tracer method to study wear in mechanical systems. Before entering into a more detailed description of the method some more general comments on the problem of wear seem to be appropriate: Wear is one of the main causes that terminate the lifetime of all machines and other mechanical devices. In view of the annual production value of all sorts of engines (many billions of dollars in the Federal Republic of Germany alone) the economic importance of wear can hardly be overestimated. The minimization of wear is therefore one of the main problems when a new machine is de-

signed, and cyclotrons can make an important contribution at this stage.

The method proceeds by activating a critical surface of a machine part and measuring the removal of material via its gamma-activity. It has been described in more detail in two recent reviews^{6,8)} and more technical details can be found in a more specialized contribution to this conference⁹⁾. The machine part whose wear is to be studied is irradiated at a critical part of its surface. Reactions between the projectiles and nuclei in the material produce radioactivity in the part. For a quantitative relationship between radioactivity and the amount of material removed from the part during a test this distribution should be homogeneous in a sufficiently thick layer below the surface (typically 0.1 to 0.2 mm). This is achieved by choosing a bombarding energy close to the maximum of the production cross section. For the study of iron and its alloys, e.g., the nuclide ⁵⁶Co is usually chosen. It can be produced from iron with protons, and the production cross section attains a maximum near 10 MeV. Often the area to be irradiated is much larger than the beam diameter. Then a homogeneous activation is achieved by moving the irradiated part periodically. A typical irradiation set-up is shown in figure 3. In view of the size and complexity of many machine parts irradiations are usually performed in air.

Some time after irradiation the machine is assembled with the activated part and operated. During operation the removal of material from the activated surface area can be monitored by either of two ways:

- a) The activity of the removed material is measured. This is usually done if the wear particles are suspended in a lubricant which is circulated in a closed loop which is then led past a gamma-ray detector. Typical examples for this are most internal combustion engines. The registered signal is in this case proportionate to the total wear accumulated since the start of the measurement (cf. figure 4).

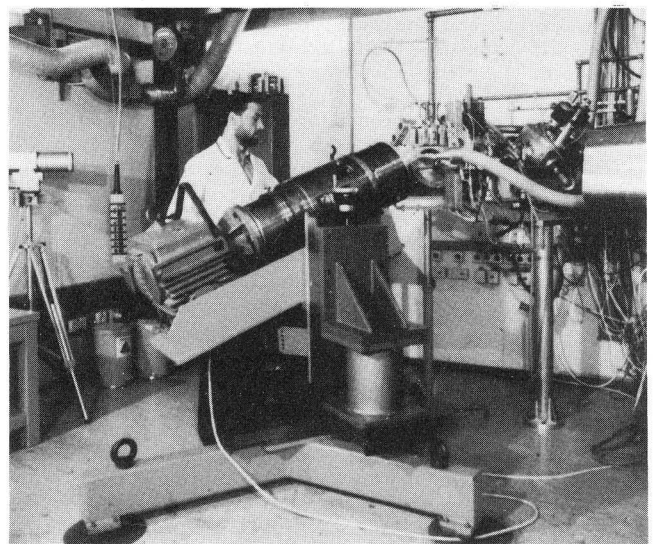


Fig. 3 : A piston liner of a marine engine set up for irradiation at a beam line of the Karlsruhe Isochronous Cyclotron.

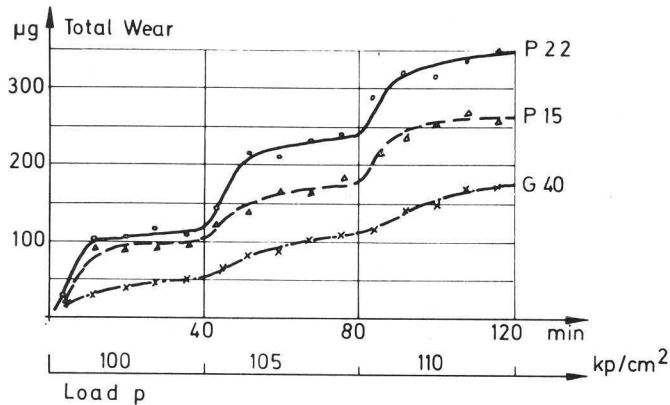


Fig. 4 : Wear of a journal bearing measured by the activation technique. Different curves correspond to different materials. It is seen that the rate of wear increases steeply after a stepwise increase of the load, but then levels off to very small values (from ref. 8).

b) The residual activity of the activated machine part is measured by a gamma-ray detector placed close to it. This procedure has to be chosen if the wear particles are dispersed as, e.g., those from an operating railway wheel.

Fig. 4 shows the result of an investigation with the first of the two procedures. It demonstrates the advantages of the method:

- High sensitivity and hence short measurement time (for iron, 100 µg removed from an area of 1 cm² corresponds to a layer thickness of 0.1 µm).
- It is possible to study many different operating conditions with one irradiated part.
- It is not necessary to disassemble the machine for the wear measurement. This represents a considerable time and cost advantage.

The technique described here briefly has been developed in Europe over the last 20 years and is now applied by an increasing number of companies as a tool in the development of new machines. There are now two laboratories in Western Europe which offer an irradiation service of the described type, Harwell (which uses a tandem Van de Graaff accelerator, though) and Karlsruhe. The wear measurement is usually performed by the engineering companies. At Karlsruhe the number of irradiated parts has increased by a factor of two over the last few years indicating the increasing interest of our industrial partners. The technique has been applied to a variety of problems, though only a small fraction of the results has been published for obvious reasons.

Most of the irradiations are performed with proton and deuteron beams between 10 and 20 MeV at currents of a few µA. Therefore cyclotrons and tandem Van de Graaff accelerators can be, and in fact are, used. For some applications, the activation of aluminum is an important example, higher energies are required which at present are only available from cyclotrons.

4. Analysis of environmental samples by proton induced X-rays. - A number of analytical methods has been developed making use of charged particle beams

from accelerators¹⁰⁻¹². Of these, the measurement of proton induced characteristic X-rays (PIXE) for elemental analysis has found very wide application during the last 10 years. The method has been reviewed in detail^{13,14} and is the subject of specialized conferences^{15,16}. It will therefore only be briefly described here:

The sample to be analyzed is irradiated by a charged particle beam, usually protons in the energy range of 2 to 4 MeV. The characteristic X-rays which are excited by the beam, are registered by a Si(Li) detector. The resulting spectrum (cf. figure 5) is then analyzed by a computer program giving the amount of each element present in the sample. Almost every piece of solid material can be used as a sample, though for highest sensitivity thin layers or foils are to be preferred.

The method offers the following advantages most of which are of direct relevance to the analysis of environmental samples:

- A large number of elements from sodium to uranium can be analyzed simultaneously, in principle.
- For most of these elements the detection limit lies between 1 and 10 ppm.
- Only very small amounts of material are required, typically between 10⁻³ and 10⁻⁶ g.
- Typically only several minutes of beam time are required, so very large amounts of samples can be investigated.
- The procedure lends itself to automatic sample change and data acquisition, again facilitating the analysis of large numbers of specimens.
- A high spatial resolution can be achieved by focussing the beam down to µm dimensions^{17,18} (proton microprobes).

While the PIXE method has been applied to many fields the analysis of airborne dust and of water samples has been among the major ones. An example of such an analysis is shown in figure 5.

PIXE systems are now installed at about a dozen cyclotrons, and at least one machine offers PIXE analyses on a commercial basis¹⁹ (at 2 to 25 \$/sample depending on sensitivity). This shows clearly that cyclotrons have to play a part in this field. Never-

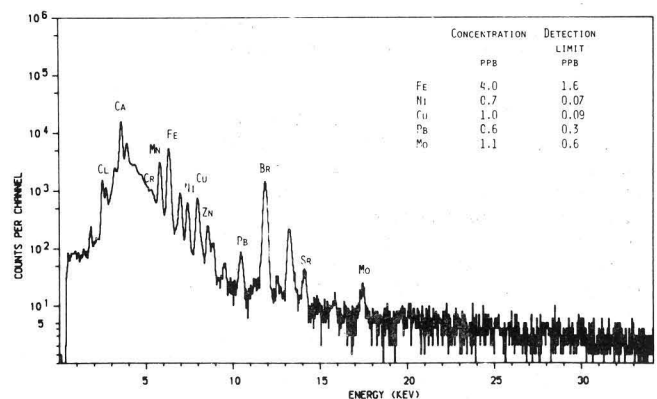


Fig. 5 : X-ray spectrum obtained from a sample of coastal water. All metals except alcalis and alkaline earths have been preconcentrated chemically (from ref. 22).

theless the great majority of work is done using small Van de Graaff accelerators in the 2 to 4 MV range which are quite appropriate for most of the problems and are available in large number all over the world.

There are some divergent views in literature²⁰⁾ as to whether the higher particle energies which most cyclotrons can provide offer a substantial advantage for PIXE. Since the merits of an analytical method for a specific application depend very much on the properties of the sample to be analyzed it is not easy to put forward arguments of universal validity. Nevertheless some comments seem to be appropriate in view of the importance of this question for the possible use of cyclotrons.

When working with protons around 3 MeV the elements with atomic number below ~ 45 are detected via their K X-rays, heavier elements via L X-rays. For most elements the K X-ray production cross section increases steeply when increasing the proton energy beyond 3 MeV. Unfortunately, so does the background in a way depending, among others, on the composition and thickness of the sample. An unavoidable contribution to the background stems from the onset of nuclear reactions. These result in the production of nuclear gamma-ray lines which in extreme cases may lead to severe misinterpretation of the results. There is experimental evidence, though, that at least in some cases the optimum energy may be somewhat higher (5 to

6 MeV) than previously believed²¹⁾. There is nevertheless at least one situation where the use of higher energies may be a decisive advantage: If the L X-rays of a trace element interfere with the K X-rays of a main constituent the detection limit may be very low at low proton energies, and the detection of the trace element via its K X-rays at higher proton energies may be the solution of the problem, in spite of the higher cost of beam time.

5. Concluding Remarks. - The examples described in sections 2 and 3 above clearly show that cyclotrons can be very useful in the solution of problems in industry. This short list is of course not exhaustive. Attention should, e.g., be drawn to the technique of proton radiography developed at Harwell and applied to the study of porosity in castings²⁴⁾. In all these cases cyclotrons are used as tools in industrial research and development rather than in production. In my opinion, the scope of industrial applications will therefore be much more limited than in medical isotope production.

The number of laboratories all over the world engaged in materials development for fast breeder and fusion reactors will probably remain rather limited. In contrast to that a much larger industry is confronted with problems of wear in design work. The potential of the procedures described in section 3 is therefore, in my opinion, far from being exhausted. In fact, the method is now used only in a few (mostly European) countries. The difficulties that hamper or at least slow down a rapid extension of this work are connected to the knowledge gap between mechanical engineers and nuclear physicists and to the widespread hesitance towards 'atomic' radiation.

The use of cyclotrons to PIXE analysis can, in my opinion, be a valuable additional application, and has been shown to be cost competitive in such a scheme. If a dedicated accelerator for PIXE were to be considered cyclotrons would have to face strong com-

petition from low energy Van de Graaff accelerators which are very inexpensive.

Let me finish by some comments on the cost of beam time. These are mainly governed by three factors: the scheme of depreciation, the amount of useful beam time achieved, and the cost of the operating personnel which depends very much on the complexity of the installation. It is not easy to obtain reliable figures. My best estimate is that for the type and size of cyclotrons considered here the cost will scatter between 100 and 500 \$ per useful hour of beam time. This is not cheap, even on standards of industrial development. But the examples I have presented have shown, I hope, that cyclotrons are worth their money for a number of uses also outside basic research and the medical field.

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References

1. A. GERVE, G. SCHATZ, Proc. 7th Int. Conf. on Cycl., Zürich, August 19-22, 1975, W. Joho, ed., p. 496; Birkhäuser Verlag, Basel and Stuttgart (1975).
2. J.L. NEED, Proc. 8th Int. Conf. on Cycl., Bloomington, Sept. 18-22, 1978, J.W. Hicks ed.; IEEE Trans. Nucl. Sc. NS-26, No. 2 (1979) 2236.
3. R.S. NELSON, D.J. MAZEY, J.A. HUDSON, J. Nucl. Mater. 37 (1970) 1.
4. A.D. MARWICK, J. Nucl. Mater. 55 (1975) 259.
5. R.S. NELSON, Proc. 6th Int. Conf. on Cycl., Vancouver, July 18-21, 1972, J.J. Burgerjon and A. Strathdee eds., p. 664 ff; American Inst. Phys., New York (1972).
6. T.W. CONLON, Atom No. 287 (1980) 223.
7. Nucl. Meth. in Mater. Research, Proc. EPS Conf., Darmstadt, Sept. 23-26, 1980, K. Bethge et al. eds.; Vieweg Verlag, Braunschweig/Wiesbaden (1980).
8. G. ESSIG and P. FEHSENFELD, ref. 7, p. 70 ff.
9. E. BOLLMANN et al., these proceedings.
10. J.L. DEBRUN, IEEE Trans. Nucl. Sci. NS-26, No. 2 (1979) 2229.
11. P. MÜLLER, ref. 7, p. 82 ff.
12. Ch. ENGELMANN, ref. 7, p. 129 ff.
13. S.A.E. JOHANSSON and T.B. JOHANSSON, Nucl. Instr. Meth. 137 (1976) 473.
14. T.A. CAHILL, Ann. Rev. Nucl. Part. Sci. 30 (1980) 211.
15. Proc. Int. Conf. PIXE, Nucl. Instr. Meth. 142 (1977).
16. Proc. 2nd Int. Conf. PIXE, Nucl. Instr. Meth. 181 (1981).
17. J.A. COOKSON, ref. 7, p. 145 ff. Nucl. Instr. Meth. 165 (1979) 477; ref. 16, p. 115 ff.
18. B. MARTIN and R. NOBILING, in Applied Charged Particle Optics, A. Septier ed.; Academic Press, New York 1979, p. 321 ff.
19. T.A. CAHILL, private communication.
20. M.S. AL-GHAZI et al., IEEE Trans. Nucl. Sci. NS-26, No. 2 (1979) 2262.

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September 1981, Caen, France

21. A. YAMADERA et al., Nucl. Instr. Meth. 181 (1981) 15.
22. E.M. JOHANSSON and K.R. AKSELSSON, Nucl. Instr. Meth. 181 (1981) 221.
23. Proc. Conf. Uses of Cyclotrons in Chemistry, Metallurgy and Biology, Oxford, Sept. 22-23, 1969, C.B. Amphlett ed.; Butterworths, London (1970).
24. D. WEST and A.C. SHERWOOD, Non-Destr. Test. 6 (1973) 249; P. STAFFORD, A.C. SHERWOOD, D. WEST, Non-Destr. Test. 8 (1975) 235.

" DISCUSSION "

Y. JONGEN : Could you comment on the possible use of heavier ions in machine parts irradiation, to get thinner layers ?

G. SCHATZ : We do not have specific experience with heavy ions at Karlsruhe. I think one should have to consider the question of radiation damage, which might change the wear behaviour, because the ratio of radiation damage to activation is probably higher for heavy ions. It is possible of course, to obtain thinner layers by tilting the irradiated piece with respect to the beam.