3 Activating radiation sources

3.1 Radionuclide sources

Radionuclide sources were the first to be applied for photon activation analysis[1,2,3]. Analyses of deuterium, beryllium and fissile material have been performed exploiting the photodisintegration process with direct observation of the promptly emitted neutron radiation. Strong ^{124}Sb gamma-ray sources have been used in almost all application cases (see e.g. Ref.[182]), but also others were reported[3,183].

The first photoexcitations of isomeric states through nuclide radiation were achieved with help of ^{226}Ra[184], ^{182}Ta and ^{60}Co sources[185]. In the laboratory practice, mostly strong ^{60}Co-sources (some 10^{13} up to some 10^{15} Bq) have been applied[185-192]. In a few cases others have been used[185-188,193]. In Tab.3-1 the applications of nuclide gamma-sources are summarised. However, using these sources, the achievable analytical sensitivity frequently suffers from unsuitable energy of the activating radiation; normally the gamma energy of the nuclide cannot excite the absorption resonance level of the target nuclide. Therefore, using radionuclide activating sources, radiation due to Compton scattering can only be exploited (see Ch.2).

Higher energies can be obtained through photon radiation promptly emitted during nuclear reaction of some elements. In this instance, several (p,γ)-reactions provide useful photon energies for photonuclear reactions, e.g.

$$^{11}B(p,\gamma)^{12}C \qquad E = 15.96\ MeV$$
$$^{7}Li(p,\gamma)^{8}Be \qquad E = 17.26\ MeV$$
$$^{3}H(p,\gamma)^{4}He \qquad E = 19.80\ MeV$$

However, for different reasons, these photon sources normally are of limited use for photon activation analysis[23,45,194].

Akbarow et al.[195] used the gamma emission of reactor-produced ^{16}N (by irradiation of fluorine compounds) for photon excitation of several isomeric states.

All in all, for several reasons (low achievable photon flux, monochromatic rad-

iation whose energy most probably does not coincide with the desired excitation level; see Ch.2) the use of isotopic sources for photon activation analysis is restricted to a few advantageous cases; see also Ch.6.2.

Tab. 3-1: Isotope γ-ray sources used for photoexcitation

Isotope	Half-life	Activity (Bq)[1]	Isomeres produced	Ref.
^{60}Co	5.27 a	$4 \cdot 10^{14}$	^{115m}In	186
		$6 \cdot 10^{12} - 7 \cdot 10^{13}$	^{115m}In	185
		$7 \cdot 10^{13}$	^{111m}Cd	185
		$3 \cdot 10^{15}$	^{77m}Se, ^{79m}Br, ^{87m}Sr, ^{107m}Ag, ^{109m}Ag, ^{111m}Cd, ^{115m}In	188
		$2 \cdot 10^{14} - 1 \cdot 10^{15}$	^{87m}Sr, ^{111m}Cd, ^{113m}In, ^{115m}In, ^{176m}Lu	189
		$2 \cdot 10^{14} - 4 \cdot 10^{14}$	^{77m}Se	190
		$7 \cdot 10^{14}$	^{77m}Se, ^{79m}Br, ^{87m}Sr, ^{107m}Ag, ^{109m}Ag, ^{111m}Cd, ^{115m}In, ^{179m}Hf, ^{191m}Ir, ^{195m}Pt, ^{197m}Au	187
^{182}Ta	115 d	$5 \cdot 10^{13}$	^{115m}In	185
^{116m}In	56 m	not given	^{77m}Br, ^{107m}Ag, ^{109m}Ag, ^{115m}In	188
^{24}Na	15 h	$7 \cdot 10^{12}$	^{167m}Er	193

[1] Values were transformed from integer Curie activities, and were rounded to integer Bequerel values.

3.2 Electron accelerators

The disadvantages of the isotopic sources mentioned above can be eliminated by using bremsstrahlung of accelerator-produced electrons for photoactivation. The achievable photon fluxes usually outrange those of radionuclide sources by orders of magnitude. Moreover, the effective cross section is significantly enlarged since the bremsstrahlung energy is continuous with the electron energy. Finally, photon energies can be produced which are much higher than obtainable with any isotope or nuclear reaction source. Therefore, activating with high energy bremsstrahlung, photonuclear rections can be induced in the target material, whereas - except very few cases - only isomeric state excitation can be achieved by gamma-rays from isotopes (see above).

An accelerator is a device to accelerate charged particles such as electrons, protons or heavier ions up to a kinetic energy through which they are enabled to induce reactions upon the electron shell or the nucleus of a target atom. One has to distinguish primarily between electrostatic and cyclic accelerators. The term "cyclic" exclusively relates to the operation mode of the machine. In some literature one finds this term used for circular path accelerators. This is not correct: "cyclic" in this context does not relate to the geometry of the particle trajectories.

One has also to distinguish between linear and circular machines, according to the accelerated particle path geometry. In this chapter only those machines are described which can be applied for the production of bremsstrahlung usable for photon activation. These are: Van de Graaff generator, linear accelerator, betatron and microtron. Others were used in comparably few cases and therefore are mentioned marginally if relevant.

In static accelerators electrons are accelerated by a constant high voltage potential. The maximum achievable particle energy is directly dependent upon the maximum high voltage of the individual machine. The most prominent examples are the Cockcroft-Walton generator[196-199] and the Van de Graaff accelerator. As far as it is known to the authors the former has never been used for photon activation analysis.

In cyclic accelerators electron energies are achieved by multiple application of comparatively low voltages upon the electrons. The maximum achievable energy is dependent on various parameters. Examples of these machines are: linear accelerator, betatron, microtron.

Regarding the requirements of photon activation analysis (see Ch.2) it is obvious that accelerators providing electron energies of more than say 50 MeV are unnecessary. Moreover, excessively high bremsstrahlung energies are unsuitable since they lead to undesirable competing photonuclear reactions (see Ch.2).

According to the practical experience of most analysts those machines meet their requirements best which provide around 30 MeV electron energy at average beam currents of at least 100 microamperes (see Ch.2).

For more general information about electron accelerators used in photon activation analysis the reader might refer to Refs.[14,200-204,508].

3.2.1 Van de Graaff generator

The Van de Graaff belt generator was first proposed in 1931[205]. The function principle is shown in Fig.3.1. An insulating belt is driven by a motor connected to one of the pulleys, at ground potential. Near the motor-driven pulley a row of points is located across the width of the belt and is kept at a potential of about 30 kV (charging supply). A corona discharge between these points and the moving belt ionises the atmosphere and electric charge is transferred to the belt. The other side of the belt support is inside the high voltage electrode. Here the belt-borne charge is removed and transferred to the outer surface of the high voltage electrode. Thus the voltage progressively increases. The electron source (electron gun) and the upper end of the accelerator tube are located inside the HV electrode. Normally the whole accelerator assembly is located in a pressure tank which is filled with highly pressurised insulating gas, e.g. sulfur hexafluoride. This gas inhibits uncontrolled discharge along the high-voltage bearing parts of the machine.

The first versions of this machine type could produce high voltages of close to one MV and thus electrons of about one MeV. Modified versions, equipped with special insulators were then quickly developed; they could reach up to about 3 MV[206-210].

Nowadays, however, Van de Graaff generators which can produce up to several tens of MeV electron energy are commercially available at comparatively moderate purchase prices. At energies of say five MeV, electron beam currents of some milliamperes are not unusual. These machines are relatively small and compact; they have been appreciated for their simplicity and flexibility. The fields of application are X-ray diffractometry, research on the atomic struct-

ure, non-destructive materials investigation and - last not least - for activation analysis[121,194,211,212].

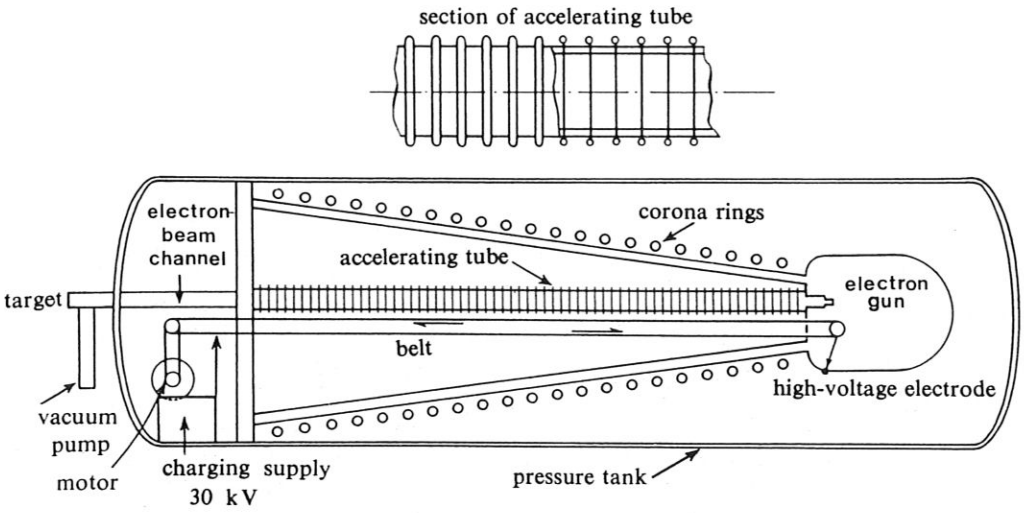

Fig. 3.1: Schematic representation of a van de Graaff-generator for acceleration of electrons

3.2.2 Linear accelerator

For photon activation analysis purpose, mostly linear accelerators have been utilised. Therefore, this machine type is here described in more detail than the other accelerators.

Among the cyclic machines the linear accelerator, also briefly called "lineac" or "linac", was developed first. The term "linear accelerator" does not only imply the fact that particles are accelerated along straight trajectories, but also the application of a high frequency source to produce a wave with help of which the particles are carried to their final energy. The first theoretical considerations were published in 1924[213]; the first operating linac was reported in 1928[214]. In the early 1930's machines were built which could produce more than 1 MeV maximum energy for mercury ions[215] and up to 2.5 MeV for electrons (Refs.[216-219]).

In the analytical context the early experiments of Coates[220] are of some interest; radiation was analysed which originated from bombardment of several target elements with Hg-ions, not very successfully due to the lack of high performance radiation spectrometry equipment at that time.

In the middle of the 1930´s the further development of the linac intermediately stopped in a stadium of prematurity because of the lack of high frequency generators with high output power. Moreover, the cyclotron and - almost simultaneously - the betatron was developed at that time and thus the "linear" idea was abandoned in favour of the "circular". The linac principle reentered the scene when the desired high-power frequency source, namely the klystron, was invented. Its development was pushed foreward during World War II under the pressure of radar protection. The klystron was announced and developed by different groups[221].

Major development work on high frequency sources and linear accelerators was performed by the linac research group at Stanford, U.S.A.. The first linac at Stanford provided a maximum electron energy of 4.5 MeV. Later on, series of progressively larger linacs were built to finally achieve an electron energy of more than 20 GeV[222-224]. Barely any of these machines would have met the requirements of photon activation analysis; it is not known to the authors, if it was ever attempted to use any of the Stanford machines for analytical purpose. These machines were designed to serve physical research purpose exclusively.

Also other research groups have developed and constructed linear accelerators (Ref´s.[222-228]), but the most relevant work on electron linear accelerators was undoubtedly performed at Stanford.

3.2.1.1 General description

A linear accelerator is represented schematically in Fig.3.2. Electrons are accelerated along straight trajectories by alternating electromagnetic fields supplied by a radiofrequency (rf) system. These fields are made to propagate along the axis of a cylindrical structure. The rf system consists of an rf-generator which supplies microwaves of highly stable frequencies of several gigacycles per second. The power of the microwaves is amplified (up to several tens of megawatts peak power) by a high power klystron. Frequently a multiple-step power amplification system is used.

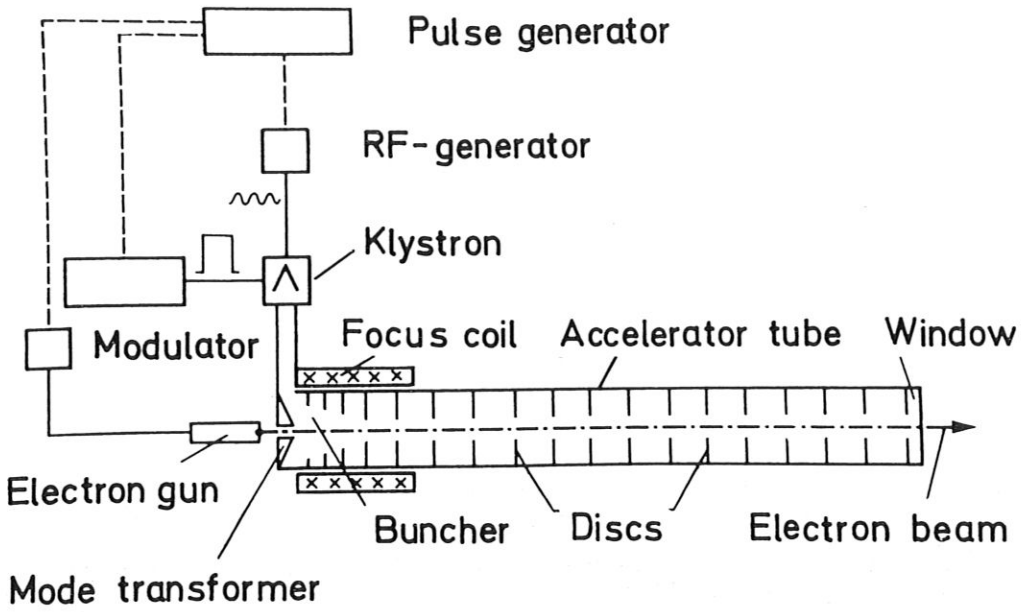

Fig. 3.2: Schematic representation of a travelling wave linear accelerator for electrons with a single accelerator tube section

The electron source (electron gun) basically consists of a heated metal or metal oxide from which the electrons are emitted. These are extracted from the source region and focussed by electrodes and then injected into the buncher region of the accelerator. The electron gun - as well as the rf-system - is designed for pulsed operation at repetition rates selectable from a few cycles per second to maximally several thousand. The pulses feeding the electron gun and the microwave amplifier are provided by a high power modulator driven by a master pulse generator. Pulsed operation is necessary because of the excessively large power required in electron accelerators. The microwave amplifier can provide the excessively large rf-power only in pulsed operation at relatively low duty cycle.

The electron beam, before being injected into the buncher section, has to be well collimated, and it must be homogeneous in energy to avoid too large spread during acceleration. Actually, the solid angle of injection must not exceed several thousandths of a steradian. The sharp focalisation of the electron beam is realised by focus coils which surround the accelerator tube in the buncher

region. In some linacs, focussing coils are provided over the entire length of the accelerating tube. The injection is performed at an energy selected so that the electrons are readily captured by the electric field within the accelerating section of the linac; usually it is some tens to several hundreds of keV. The wave mode of the accelerating microwave supplied by the klystron, before being transferred to the accelerating tube, is converted by a mode transformer. Hereby it is transformed so that its electric field vector coincides with the beam direction.

The accelerating structures are specially designed waveguides. These allow continuous energy transfer from the electromagnetic wave to the electrons up to the desired value.

A waveguide is a metal duct which is evacuated or filled with a dielectric. Under certain conditions electromagnetic waves can propagate through it, "guided" by the metal wall. In electron accelerators, waveguides are supplied with discs containing circular holes in the center ("irises") placed at certain distances within the accelerator tube. The phase velocity of the accelerating wave increases with the distance of the irises (if there were no irises provided the phase velocity would exceed light velocity and thus become unsuitable for particle acceleration). These "loaded" waveguides enable the formation of a travelling wave to carry electrons to their final energy, maximally about 20 GeV hitherto, for photon activation analysis optimally around 30 MeV (see above and in Ch.2).

Out of the electrons injected into the buncher section only those are captured by the travelling wave which "see" its proper phase whilst all residuals are discarded. The irises within the buncher area are arranged in increasing distances so as to obtain continuously increasing phase velocity of the travelling wave up to nearly light velocity. Thereafter the irises normally are equidistant until the electron window at the end of the accelerating tube. Any further energy increase after the buncher section serves only for relativistic mass increase of the electrons near light velocity.

At the end of the accelerating tube the electron beam is transmitted through a beam window which usually consists of a thin metal foil, e.g. titanium.

Bremsstrahlung is produced by absorption of the electron beam in a target of high Z material whereby a part of its energy is converted to X-radiation. The bremsstrahlung production mechanism is discussed in 3.5 below.

3.2.2.2 The linear accelerator used in the present work

The machine used for this work is discussed only briefly here, emphasising the data and facts which are relevant for photon activation analysis. A detailed description of the accelerator is given in the BAM Linac Report[55]. A schematic representation of the electron linear accelerator of the Bundesanstalt für Materialprüfung is given in Fig.3.3.

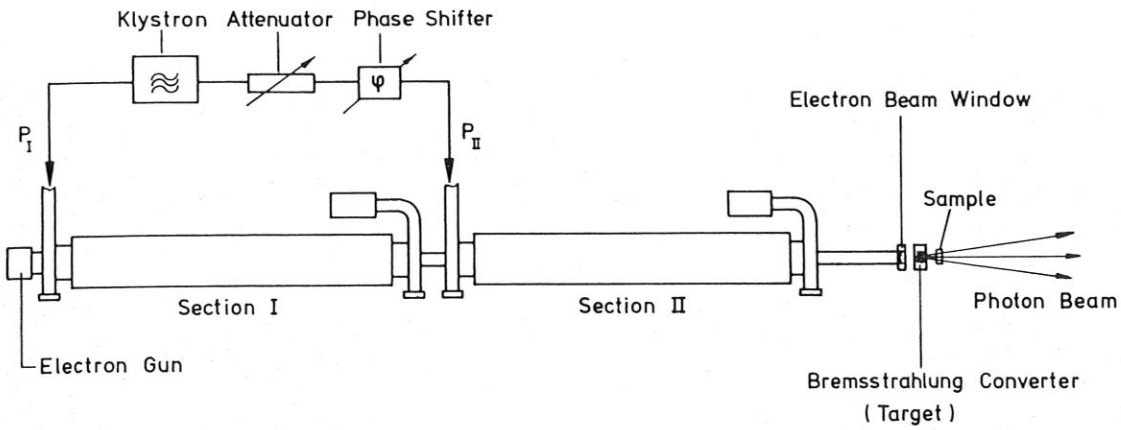

Fig. 3.3: Schematic representation of the BAM Linac

a) The accelerating waveguide

In this machine the accelerating waveguide is divided into two sections. This is done for the following reason: A continuous acceleration of electrons in a linear accelerator could only be obtained by a quasi-constant accelerating field strength along the entire waveguide. However, the field strength decreases rapidly along the accelerating path due to losses through resistance and continuous energy transfer to the electrons to be accelerated. Therefore, the continuous accelerating path length and thus the achievable output electron energy of this system is limited to about two meters or about 17 MeV, respectively. Further energy increase can be obtained only by a subsequent waveguide section with its own rf power supply. In the described machine one rf-system,

equipped with a power divider (or attenuator), supplies both waveguide sections with the required energy. The sections are coupled electrically with a phase shifter. The first section operates at constant power providing electrons of about 17 MeV. By proper setting of the phase shifter the electron energy increases continuously to a maximum value of about 35 MeV. The phase shifter can also be set so that the electrons "see" a reversed electric field and thus are decelerated. The actual value of acceleration or deceleration is determined by the input power of the second waveguide section which is selectable with help of the power attenuator. In Fig.3.4 the electron output energy as a function of the electron beam current is shown, whereby the graphs 1 to 9 represent the energy at different input powers of the second waveguide section. The dashed line is valid for maximum input power. Thus, the final electron energy is continously selectable from about 4 to about 35 MeV.

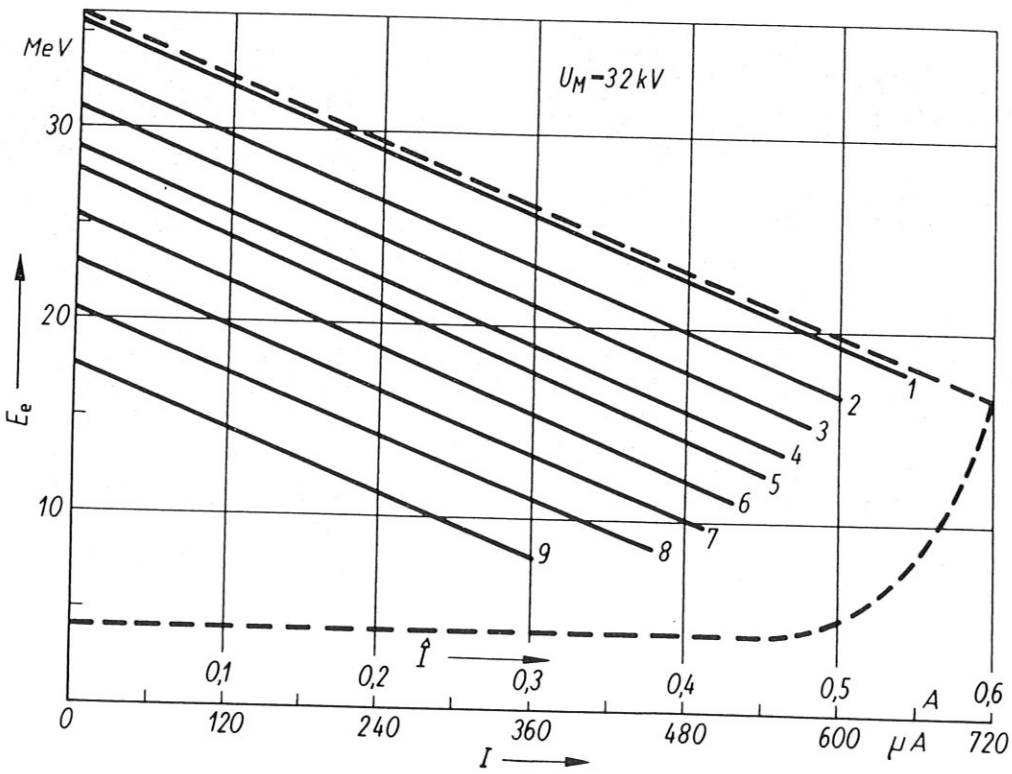

Fig. 3.4: Load curves of the BAM Linac; relationship between electron output current and energy ($\hat{I}$ = peak current; I = mean current); parameter = input power of the second waveguide section

Assuming 40 keV injection energy of the electrons, their velocity is 0,37 c. They are then accelereated to about 0,99 c. Any further energy increase appears as relativistic mass increase. To avoid undesirable spread of the electrons the entire waveguide is surrounded by focussing coils.

The accelerator is equipped with a beam steering system containing special steering coils and a magnetic quadrupole lens at the end of the waveguide.

b) the rf - system

The rf energy is produced by a commercially available five-chamber radar klystron with a peak output power of 24 MW (mean value: 24 kW; actually, a multiple step system containing a highfrequency oscillator, a driver klystron and the mentioned main klystron is used). The klystron is equipped with two power outputs of 12 MW peak power each. The rf-system (as well as the electron gun; see below) is pulsed at repetition rates selectable in steps from 12.5 up to 300 s^{-1}. The pulse length of the microwave is 4 µs.

The output power of the klystron is transferred to both accelerator sections through waveguides with rectangular cross section; the first accelerator section operates at a constant input power (PI) of 12 MW (peak value). As shortly mentioned above, at the entrance of each accelerator section the electric field vector of the microwave is rotated by 90° by a mode transformer so that its direction is parallel to the electron beam axis. The second accelerating waveguide section operates at a power selectable with the attenuator up to a maximum of 12 MW peak value (PII).

c) the electron source

In the electron gun electrons are produced by a heated tantalum disc. The disc is heated through bombardment with electrons which are emitted from a hot tungsten wire and directed to the tantalum disc by a static voltage of 6 kV.

The electrons emitted by the tantalum disc are extracted from the electron gun pulsed (see above) high voltage (-40 kV), the drift-tube having zero potential. The electron source is sealed in a glass tube which is directly attached to the mode transformer at the first accelerator section.

Further recommended literature about electron linear accelerators can be found in Ref's. 55,69,76,116,122,229-232,682.

3.2.3 Betatron

The betatron, a circular accelerator designed to accelerate electrons, is a development logically following the cyclotron invented by Lawrence in 1930[233]. Theoretical considerations of Wideröe[214] inspired Lawrence to the idea of a circular particle path. Until the end of World War II cyclotrons were the only devices to produce high ion energies up to about 40 MeV. Since cyclotrons cannot be used for photon activation analysis they are just mentioned here and not discussed further.

Going out from the cyclotron principle the development of the betatron was straightforward. The basic principles were established - independently from one another - by Wideröe and by Slepian. The latter, in his U.S. patent[234], suggested the operation principle of the betatron. The first successfully operating betatron was built in 1935; electron energies up to 1.8 MeV were achievable by this machine.

In the beginning of the 1940´s it was D. W. Kerst who was the first to construct a betatron which provided a beam current output in the microampere region at comparatively high maximum electron energies, namely up to 2.3 MeV[99, 100, 235, 236]. He also, together with Serber, evaluated the theory of orbit stability[237].

Excellent detailed descriptions of the historical development of the betatron were given by Kopfermann[238] and Wideröe[239].

The schematic design of a betatron is presented in Fig.3.5. The accelerator consists of a magnet fed by alternating current of a frequency normally between 50 and 200 s^{-1}. A vacuum chamber is placed in the magnet gap (because of its shape the vacuum chamber usually is called "doughnut"). In this chamber the electrons are made to circulate.

The magnetic field has a two-fold purpose:

1 - The magnetic field present in the doughnut exerts a Lorentz force upon the electrons, which points towards the center of the doughnut and so forces the electrons on a circular orbit.

2 - The magnetic flux linked to the doughnut changes in time, and induces an electric field whose force lines form concentric circles orthogonal to the

axis of symmetry.

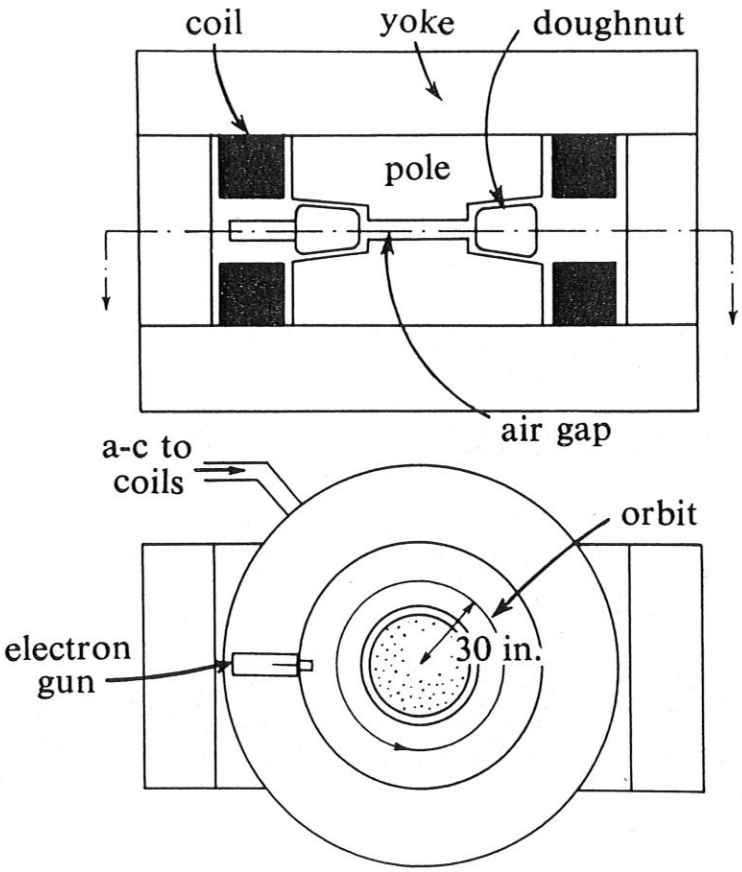

Fig. 3.5: Schematic representation of a betatron

One of these circular force lines is the "central orbit" of the electrons. This electric field accelerates the electrons.

The betatron basically can be regarded as the analog of a transformer whose primary current is the alternating current which excites the magnet and whose secondary current is the electron current circulating in the vacuum chamber.

Since the induced electric field is alternating electrons can only be accelerated in the half-cycle of increasing field strength. Therefore the electrons circulate only for a half-period of the alternating current or less. Thus the

machine runs at pulsed operation whose repetition rate coincides with the current frequency; the electrons are injected when the magnetic field is zero (or nearly zero). The beam is extracted (or used in any other desired manner) when the field has reached its maximum value (otherwise, in the case of further presence in the electric field the electrons would be decelerated). Bremsstrahlung is produced by absorption of the electron beam by a heavy metal target (see 3.2.2) either internally (within the vacuum chamber) or externally after extraction through an electron beam window.

Betatrons are used for energies between five and several tens of MeV (the largest betatron constructed hitherto has a maximum energy output of 300 MeV). For lower energies, electrostatic machines (e.g. van de Graaff accelerators) are more convenient. Higher energies would require excessively large magnets and thereby render the machine too costly. The theoretical energy limit is reached when the energy increase during acceleration equals the losses by radiation damping. This is the case at approximately 500 MeV. For photon activation betatrons of more than say 30 MeV electron energy output are not necessary or even unsuitable for the already mentioned reasons (undesirable nuclear reactions; no appreciable electron beam currents; beam currents are lower than those achievable in comparable linear accelerators by about two orders of magnitude; see above).

Low energy betatrons are frequently applied in medicine (tumor treatment). Higher energy (around 20 MeV and more) machines are mainly used in the field of material research and investigation (X-ray diffraction, radiography) or photon activation.

For more detailed information about the use of betatrons in photon activation analysis the reader might refer to Ref's[25,57,108,110,144,194,240-262] and Ch.6. Unwanted photon activation during betatron irradiation was discussed by Kuttemperoor[245,981] and Tuchscheerer[263].

3.2.4 Microtron

The above mentioned limitations of the betatron, namely limited energy and beam current can be overcome - besides using a linac - by the microtron. The concept of the microtron was established 1945 by Veksler[264-267]. The first operating microtron was reported in 1948 in Ottawa, Canada[268-271]. Further development work was performed by Kaiser in the beginning of the 1950's[272-274]. These machines yielded maximum electron energies around 6 MeV. The first microtron with

comparably high electron energy (about 30 MeV) was constructed 1953 in London (Ref's.[275-279]).

The microtron is represented schematically in Fig.3.6. The electrons are accelerated by a fixed-frequency resonant cavity and are made to move in a constant magnetic field where they describe circular trajectories with increasing radius. The orbits are tangential to the axis of the cavity.

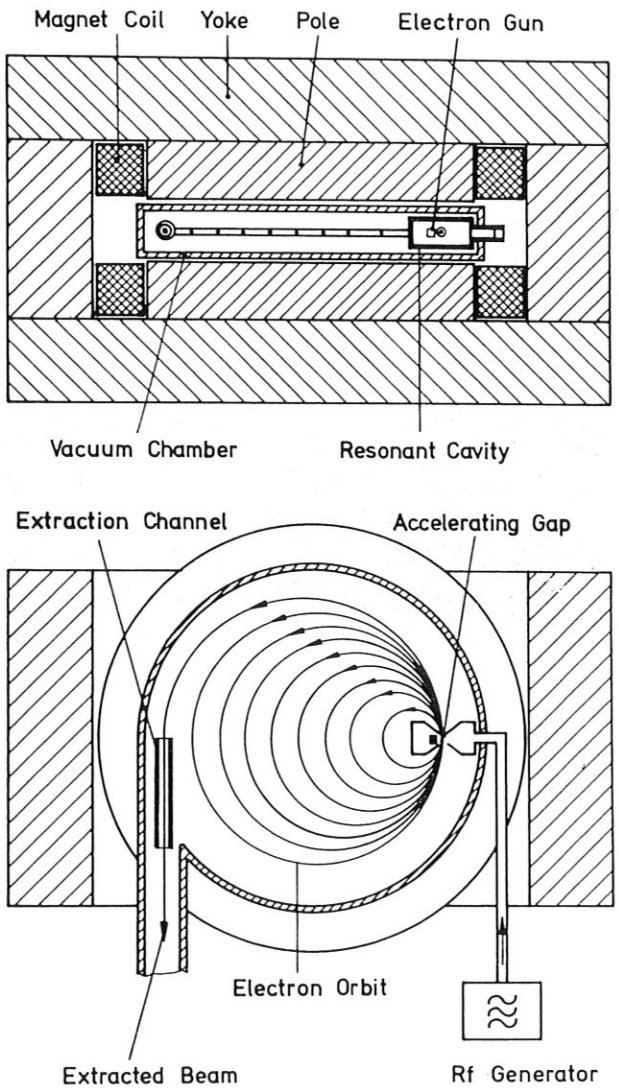

Fig. 3.6: Schematic representation of a microtron

The special advantages of the microtron are the relatively simple construction, the easy deflectability of the electron beam and its excellent energy homogen-

eity. Moreover, achievable energies and electron beam currents are in the region of medium energy linear accelerators, namely several tens of MeV at mean beam currents of about 100 μA. The values ideally meet the requirements of photon activation analysis.

A certain problem of the microtron is the vertical beam instability since the quasi-homogeneous magnetic field in the vacuum chamber does not enable any vertical focussing. Moreover, the focussing effect during transit of the electrons across the resonant cavity is limited. Thus the tolerance values in the construction of the magnet are extremely small and thus the machine might render costly, in particular if large energies are required as is the case in photon activation analysis.

Microtrons which most favourably meet the requirements of photon activation analysis were reported by Kapitsa[280,84] and Baciu[281,282]. Because of its brilliant energy homogeneity and easy-to-perform extractability of the beam the microtron can be used most advantageously as injection source for larger accelerators. For photon activation analysis, however, the microtron, in spite of the convincing advantages mentioned above, has not gained as much importance and widespread application as sometimes was predicted; see e.g. Ref.[50].

Because of some similarity to the cyclotron (acceleration by a constant high frequency in one resonant cavity in a constant magnetic field along circular trajectories) the microtron sometimes is called "electron cyclotron"[283]. A "true" electron cyclotron was built in 1952 by Salow[284]. However, this machine can produce only very low electron energies and so never has been used but for demonstration purpose.

For more information about the microtron and its use in photon activation analysis, the reader might refer to Ref.'s[25,194,280,281,283,285-300] and Ch.6. Recently, Kapitsa et al. published a survey article about the use of the microtron for photon activation analysis in the USSR (Kapitsa et al.[998] and the literature cited therein).

3.2.5 Other electron accelerators

There are two other kinds of electron accelerators which can be used for photon activation analysis, namely the synchromicroton and the electron synchrotron. Whilst the former[301-303] - as far as the authors know - has hitherto not been used for activation analysis purpose, the use of the latter has been reported

surprisingly often[25,194,304-307]. This machine was developed quasi-simultaneously by three groups[303,308,309] in 1945. It is designed to produce electrons of extremely high energies and therefore basically is not suitable for photon activation analysis. However, the workers cited above reported special applications at high (70 MeV and more) energies. This kind of accelerator is not recommendable for routine activation analysis.

3.3 Production and physical properties of bremsstrahlung

The primary radiation almost exclusively used for photon activation analysis is bremsstrahlung (X-radiation, X-rays) which is produced by stopping an electron beam from an accelerator in a heavy metal disc. A certain portion of the electron energy is converted into photons, the rest is dissipated in the converter as heat. A bremsstrahlung photon is produced when an electron incident on an atom of the converter material interacts with the electrostatic field of the nucleus. Under the influence of the attractive force acting upon the electron it is deflected from the straight path which corresponds to an acceleration in the direction towards the nucleus. From electrodynamics it is well known that an accelerated electric charge emits electromagnetic radiation. Quantum mechanically speaking, the interaction of the electron with the electric field results in the emission of a photon and a corresponding energy loss of the electron.

3.3.1 The spectrum of the bremsstrahlung photons (X-ray spectrum)

Since the radial acceleration of the electron depends on its orbit relative to the nucleus and the orbits randomly distributed we expect already in the framework of classical electrodynamics that the spectrum of emitted photons is continuous. If the electron is only slightly deflected a low energy photon is emitted but in the case of a nearly central collision with the nucleus the bremsstrahlung photon carries almost the total initial electron energy leaving the electron with a very low kinetic energy. From this simple argument we deduce that the energy of bremsstrahlung photons ranges from zero up to a maximum value which equals the energy of the incident electrons. In fact, quantum mechanical calculations yield the same result. We also expect that the probability of weak deflections is much higher than that of strong deflections because the number of electron passing the nucleus at a large distance is greater than the number of close encounters. The contribution of low energy photons to the total bremsstrahlung intensity is indeed much higher than that of high energy photons near the maximum energy. Fig.3.7 shows that the spectrum continuously declines with increasing photon energy down to zero at the maximum energy (electron energy). The spectrum also depends on the photon emission angle with respect to the direction of the incident electrons. With increasing angle the intensitiy rapidly drops for all photon energies and the spectrum becomes softer because the decrease with emission angle is more pronounced for high than for low energy photons. In Fig.3.8 the same plot is shown for 60 MeV electrons impinging on a thick tungsten target. We notice that at 60 MeV for all photon

energies the number of emitted photons is higher than at 30 MeV electron energy. Moreover, the bremsstrahlung intensity is considerably more peaked in the forward direction.

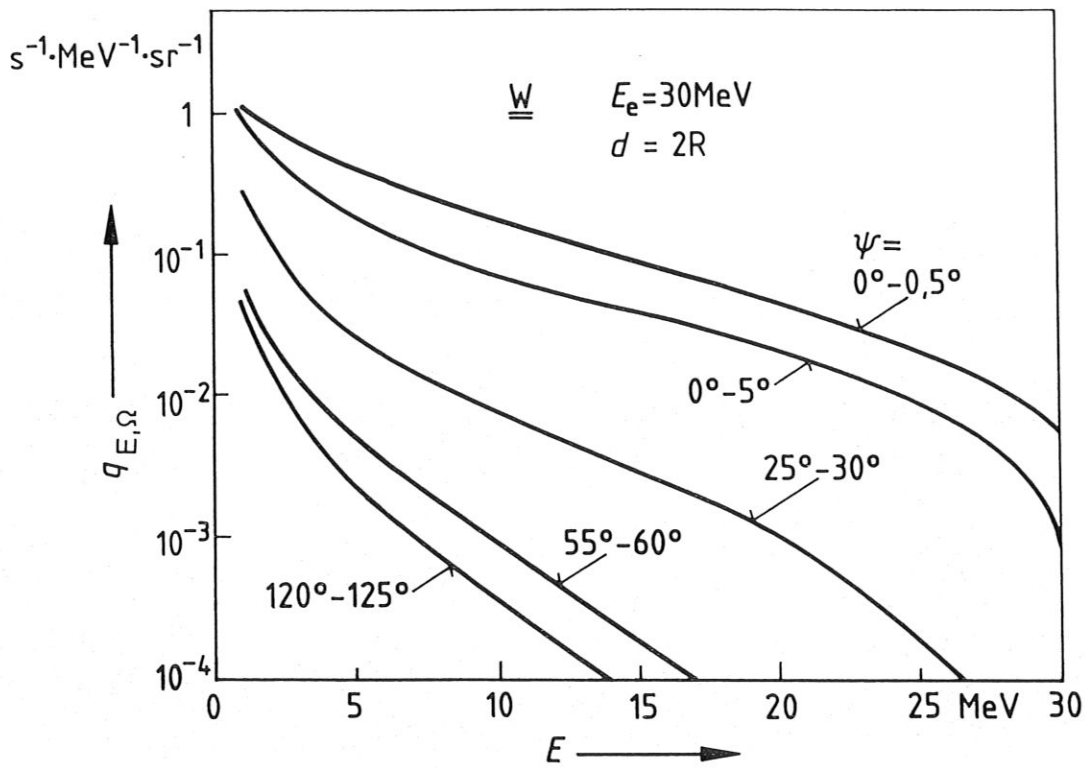

Fig. 3.7: Bremsstrahlung spectrum produced in a thick tungsten target by 30 MeV electrons at different emission angles

The angular distribution function is obtained by integrating the spectra for different emission angles over the photon energy[310,311].

Fig.3.9 shows that at small angles the total bremsstrahlung intensity rapidly falls off, then slowly decreases for medium angles and near 90° the intensity again sharply rises and than remains nearly constant throughout the major part of the backward hemisphere. The only marked change at higher electron energy is the increasing slope of the angular distribution near the forward direction (Fig.3.10). This means that the half width of the angular distribution decreases with increasing electron energy - or in other words - that the total photon intensity becomes more concentrated at small angles. In Fig.3.11 a measured

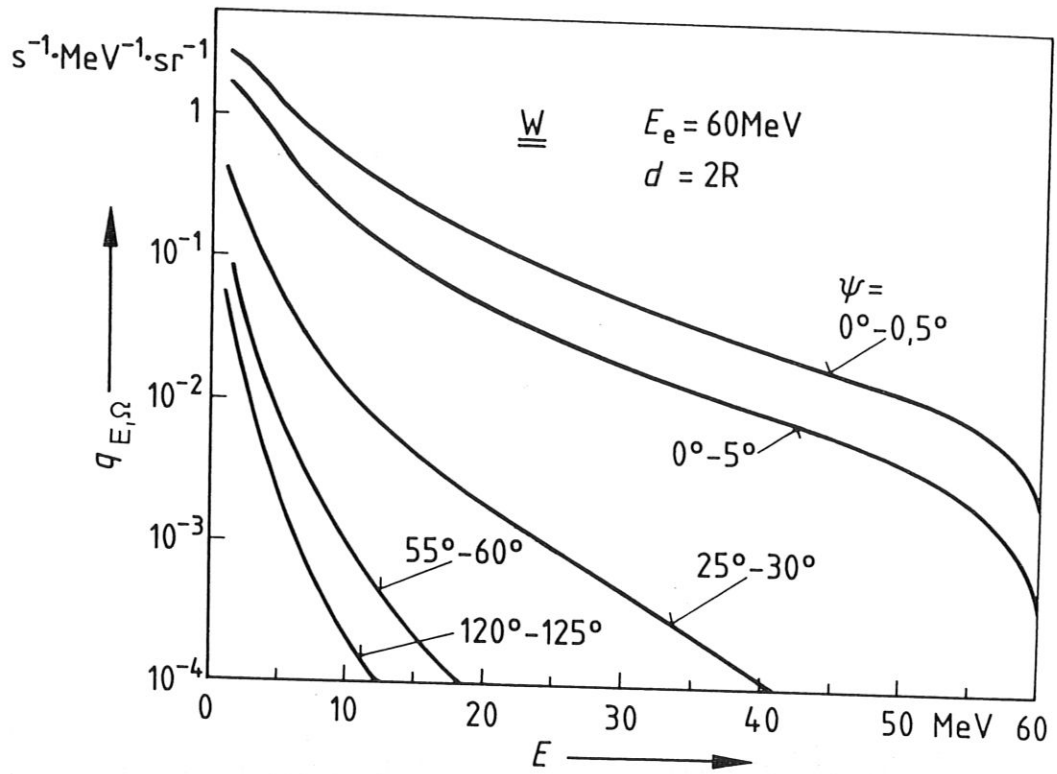

Fig. 3.8: Bremsstrahlung spectrum produced in a thick tungsten target by 60 MeV electrons at different emission angles

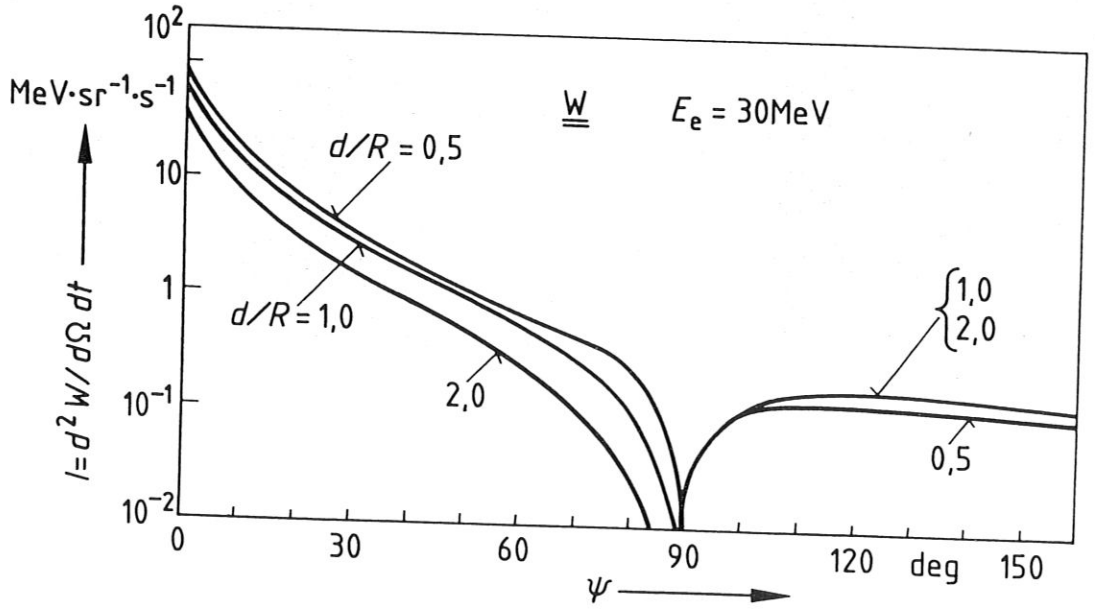

Fig. 3.9: Angular distribution of 30 MeV bremsstrahlung produced in tungsten targets with various thicknesses

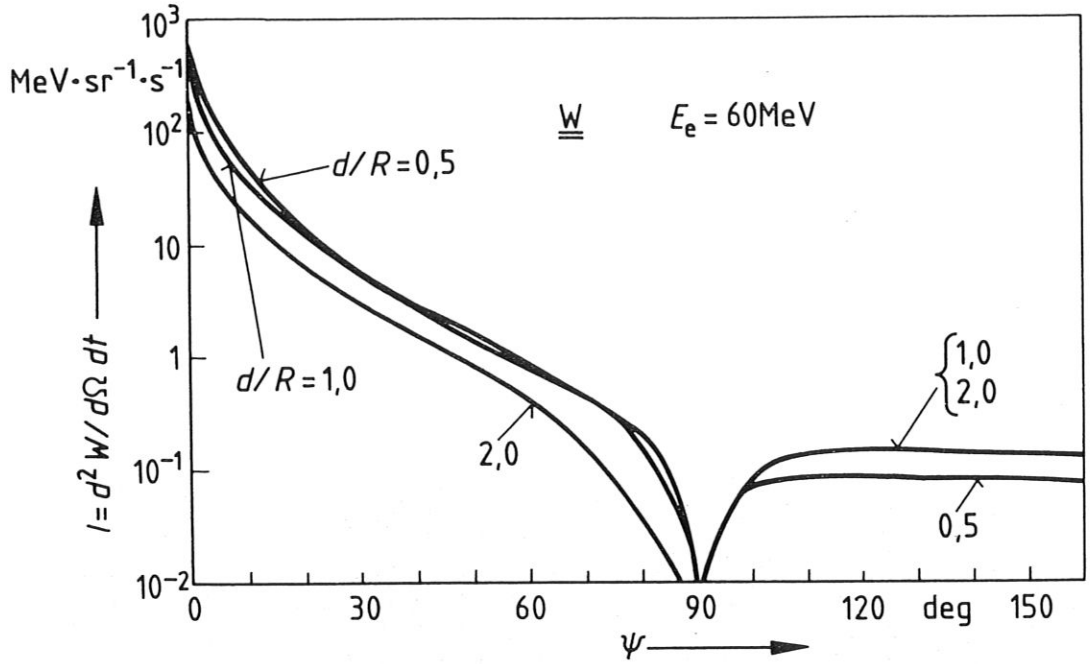

Fig. 3.10: Angular distribution of 60 MeV bremsstrahlung produced in tungsten targets with various thicknesses

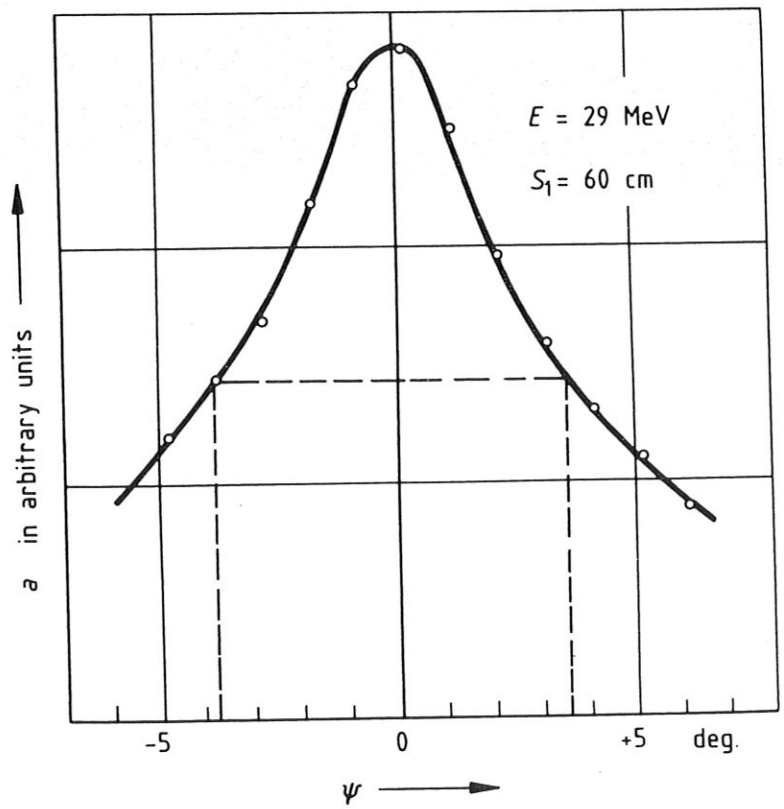

Fig 3.11: Bremsstrahlung beam profile for 29 MeV electrons measured by activation of copper

beam profile is plotted for 29 MeV electrons absorbed in a thick tantalum converter. The photon flux was determined from the activation of small copper discs arranged in a plane perpendicular to the forward direction at a distance of 60 cm from the bremsstrahlung converter. From the measured profile we conclude that the half-width of the angular distribution of photons contributing to the activation of copper is only about 3.5°. This means that in practical photon activation analysis work the large photons flux gradient of the bremsstrahlung beam near the converter must be taken into account. The sample and the reference material must be aligned as carefully as possible on the axis of the bremsstrahlung cone in order to ensure identical irradiation conditions for the sample and the reference material and a homogeneous activation of the material. The flux gradient perpendicular to the beam axis is smaller at larger distances from the converter but there the intensity is poor because the photon flux density decreases inversely proportional to the square of the distance from the converter[312]. Therefore usually a maximum distance of a few centimeters between the converter and the irradiation position is chosen. The difficulties due to the high transversal flux gradient can only be overcome by controlling the position of the electron beam - and thereby of the photon beam - during irradiation. Using the steering elements of the accelerator the beam axis can be adjusted in order to coincide with the center of the sample. The beam position is monitored by optical beam viewers or by induction monitors.

3.3.2 Bremsstrahlung efficiency

The conversion efficiency between electron beam power and the power radiated as bremsstrahlung photons depends on the electron energy and the material as well as the thickness of the converter. In Fig.3.12 the bremsstrahlung efficiency for a tungsten converter is plotted as a function of the ratio between the converter thickness determined by the competition between production and reabsorption of bremsstrahlung photons. If the converter is thin only a small number of incident electrons contributes to bremsstrahlung production whereas the major part of the electron beam transverses the material. With increasing converter thickness the number of photon producing interaction increases but also the attenuation of the photon flux by the converter material rises. In the case of a very thick converter the electron beam is completely absorbed thus producing the maximum achievable bremsstrahlung flux. But the photons are heavily attenuated in the subsequent layers so that the net photon flux emerging from the converter is low. Therefore, an optimum converter thickness exists from which the bremsstrahlung efficiency reaches maximum. The optimum converter thickness corresponds approximately to half the electron range.

Fig.3.12 shows that for 30 MeV electrons - which is a favourable energy for photon activation analysis (see Ch.2) - the maximum efficiency is about 0,4.

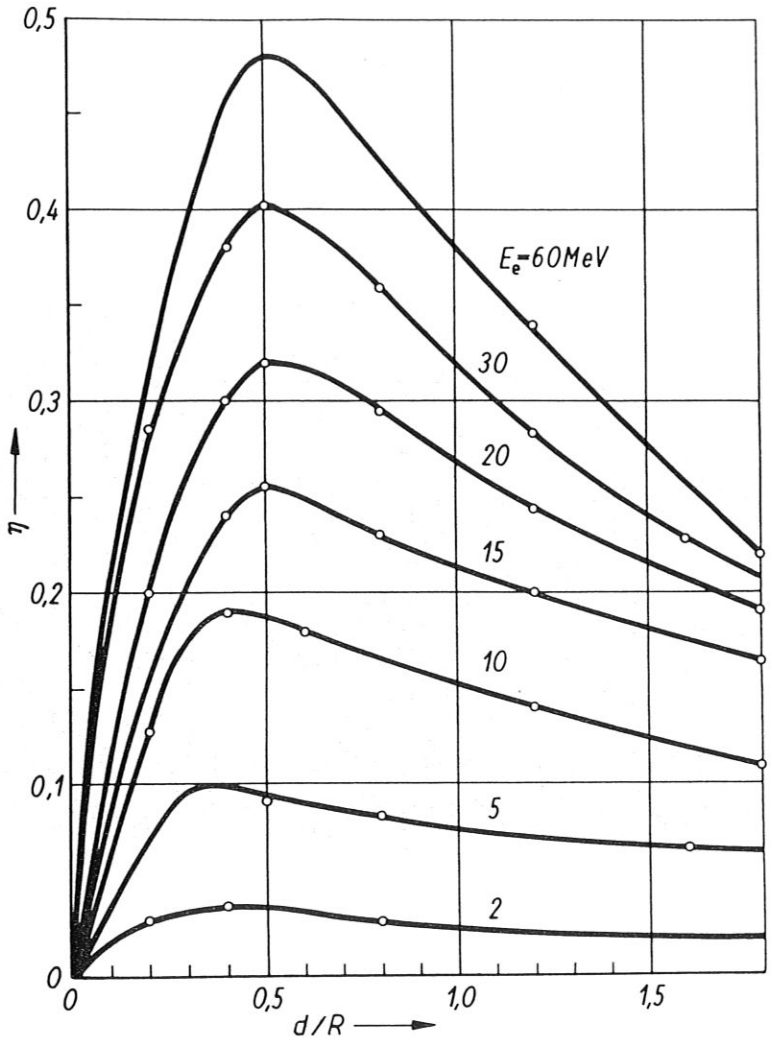

Fig. 3.12: Efficiency of bremsstrahlung production in tungsten; ratio between the total bremsstrahlung photon energy and the electron energy as a function of the target thickness and the electron energy

Therefore 40 % of the incident electron beam power is converted into electromagnetic radiation whereas 60 % is dissipated in the material as heat. From these figures it is clear that a bremsstrahlung converter must be well cooled. Since the beam power of electron accelerators used in photon activation analysis may be as high as 10 kW cooling may become a difficult problem because a thermal

power of several kW must be removed from a small volume. Therefore, the total bremsstrahlung converter is often divided into several metal discs cooled by an intense water or air flow. The most commonly used converter materials are the heavy elements tantalum, tungsten, platinum and gold. Heavy metals are preferred because the bremsstrahlung efficiency increases with increasing atomic number of the converter material. In Fig.3.13 a comparison between three converter materials is given for 35 MeV electrons.

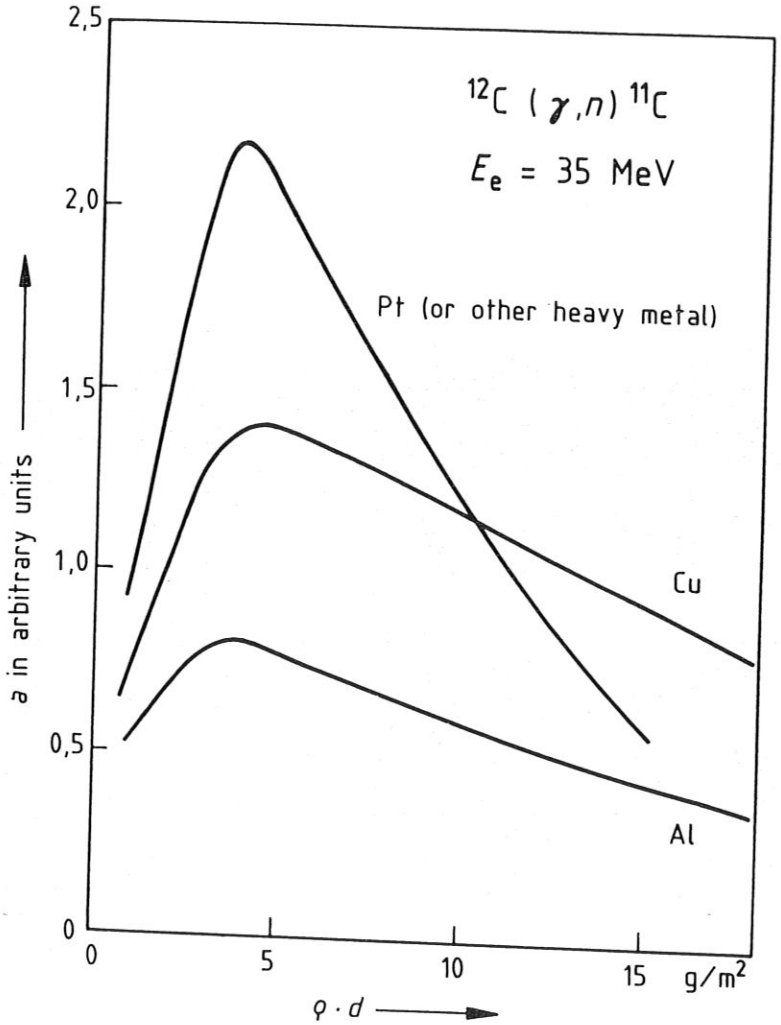

Fig. 3.13: Specific activity induced in carbon by 35 MeV bremsstrahlung produced through electron absorption in various metals as a function of the bremsstrahlung converter thickness

The flux density of bremsstrahlung photons in the forward direction is measured using the activation of carbon via the reaction $^{12}C(\gamma,n)^{11}C$. The resulting curve shows that there is indeed an optimum converter thickness yielding the maximum induced activity.

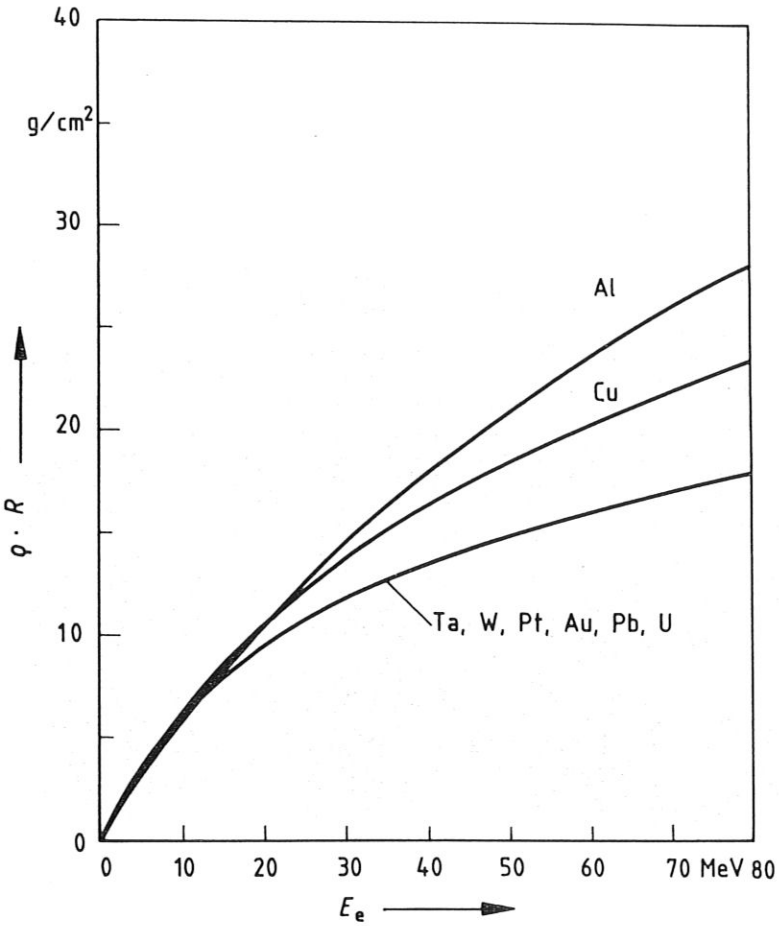

Fig. 3.14: Electron range in various heavy metals as a function of the electron energy (ρ = density of the target material)

For aluminium, copper and platinum converters this value is arround 4 g/cm^2 which is much less than the electron range in the material (see Fig.3.14) at 35 MeV. Qualitatively this is consistent with the theoretical curves in Fig.3.12. The difference between the theoretical and the measured optimum converter thickness can be explained by the fact that in the activation measurement only high energy photons above the reaction threshold contribute whereas the theoretical calculation takes into account all bremsstrahlung photons. Comparing the

yield curves in Fig.3.13 for Al, Cu and Pt we conclude that the heaviest converter material Pt is the best choice. Fig.3.15 shows the increase of the optimal thickness of a platinum target with the electron energy for (γ,n)-reactions.

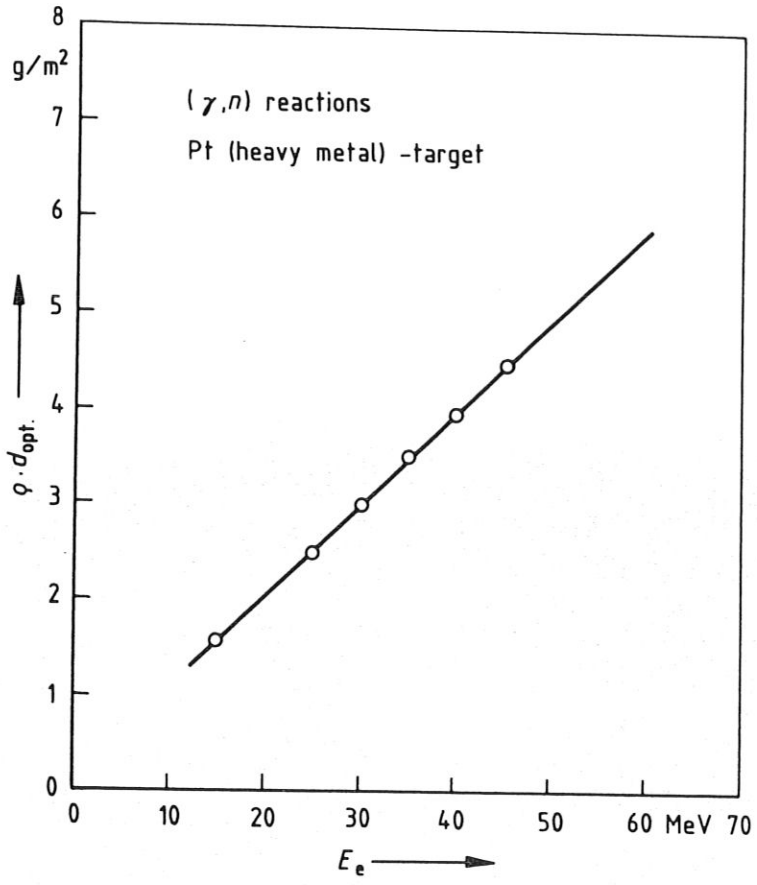

Fig. 3.15: Optimum thickness of a platinum bremsstrahlung converter as a function of the electron energy for (γ,n)-reactions

In the design of a suitable bremsstrahlung converter for photon activation analysis an additional aspect must be taken into ac-count. If the converter has the optimum thickness with respect to maximum activation of the sample a large portion of the electron beam will be transmitted through the converter because the optimum thickness is smaller than the electron range. The transmitted electrons can reach the irradiation position and dissipate their residual energy in the sample. Under unfavourable conditions the sample or even the irradiation

setup can thus be destroyed. To avoid any damage of the material the use of a cleaning magnet between the converter and the irradiation position is feasable which separates the electrons from the bremsstrahlung beam by deflecting them in the direction towards of a beam dump. This arrangement requires considerable space between the converter and the sample to be irradiated and then the activating bremsstrahlung flux density at the sample is poor. As far as the authors know, a cleaning magnet has hitherto not been used in photon activation analysis. Often the other alternative has been preferred. In order to absorb the total electron beam one selects the converter sufficiently thick. Usually the thickness is chosen to be slightly greater than the maximum electron range at the highest energy to be used for activation. Fig.3.13 shows that the resulting high energy photon flux contributing to the activation of the sample is only about a factor of two smaller than the maximum value. From the point of view of available activating flux at the sample position it is much better to place the sample at a distance of a few centimeters behind a thick converter than at several tens of centimeters (required for the deflection magnet) from an optimum converter. Yet another wayout is the use of an optimum converter and the removal of the residual electrons by a light material (e.g. aluminium) absorber. Several workers have applied this technique in photon activation analysis (see Ch.6).

Further recommended literature about the production and properties of bremsstrahlung can be found in Refs.[87,162,312-318].

3.4 The bremsstrahlung converter as a neutron source

A fraction of the bremsstrahlung photons produced in the converter interacts with the converter material by photonuclear reactions. Since the (γ,xn) reactions have a relatively high effective cross section the bremsstrahlung converter is always a neutron source with a considerable intensity. The neutron yield depends upon the electron energy and on the material and the thickness of the converter. Since both the bremsstrahlung efficiency and the effective photoneutron cross section increase as a function of the electron energy the neutron yield rapidly increase with increasing electron energy (Fig.3.16). The relationship between the neutron yield and the converter thickness has saturation character. For a thin converter most of the bremsstrahlung photons emerge from the material wihout nuclear interaction.

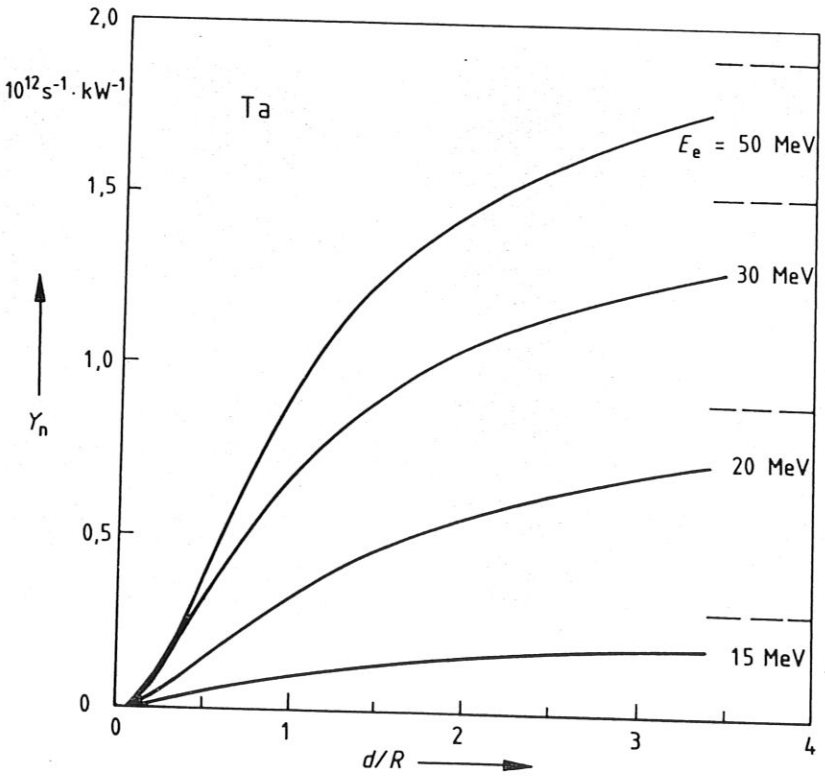

Fig. 3.16: Photoneutron yield of a tantalum target as a function of the target thickness for various electron energies; the dashed lines indicate the asymptotic values for infinite target thickness. Y_n (Yield) is the total neutron source strength per electron beam power unit.

The number of photoneutron reactions induced by the bremsstrahlung increases with increasing layer thickness. For thick converters however most of the photons are reabsorbed inside the material so that no significant increase of the photoneutron reaction rate is possible in the following layers. Fig.3.15 shows that 90 % of the saturation value of the neutron yield is reached at a converter thickness which is much larger than the electron range.

For typical working conditions of a photon activation analysis facility:

- electron energy : 30 MeV
- electron beam power : 3 kW
- converter material : tantalum
- converter thickness : 1 - 2 x electron range

the photoneutron yield will be about $3 \cdot 10^{12} s^{-1}$. This is a comparatively strong neutron source.

In Fig.3.17 calculated asymptotic neutron yields for infinitely thick converters are plotted as a function of the electron energy for various materials. Above 10 MeV the yield rapidly rises up to about 25 MeV. For electron energy above 40 MeV the neutron pro-duction rises very slowly. Since bremsstrahlung efficiency and effective photoneutron cross sections increase with the atomic number of the converter material the heavy metals yield is especially high due to photofission and neutron multiplication caused by neutron-induced fission (see Ref's.[311,319-324]).

The energy distribution of photoneutrons can be interpreted on the basis of the neutron evaporation model[313]. The high energy part of the spectrum is well described by a Maxwellian distribution with an effective nuclear temperature between 0,5 MeV and 1,5 MeV depending on the electron energy and the material. Around 1 MeV the spectrum has a maximum and rapidly decreases towards higher neutron energies. The spectral shape is similar to that of a fission spektrum. At higher neutron energies the spektrum deviates from the simple Maxwellian distribution due to the contribution of direct neutron emission. The average neutron energie is a few MeV (typically 2 MeV).

In photon activation analysis facilities the bremsstrahlung converter is often surrounded by shielding material in which the primary neutron emerging from the converter are scattered and moderated (see Fig's.3.18a and b). Therefore a large flux density of low energy neutrons can be observed at the irradiation

position giving rise to neutron capture reactions in the sample.

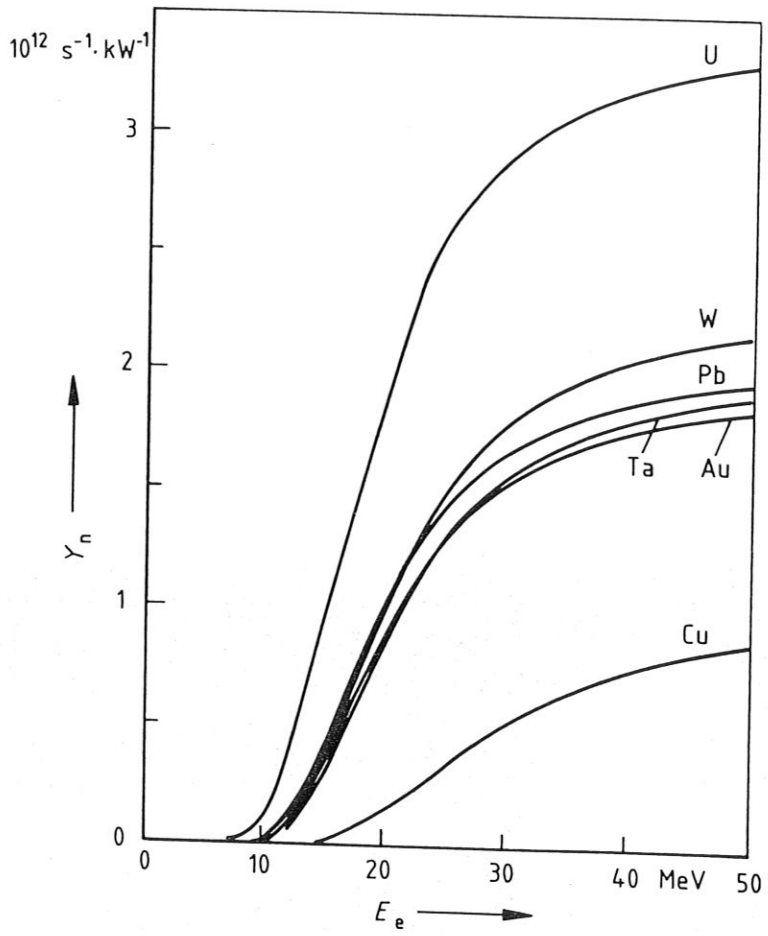

Fig. 3.17: Photoneutron yield of infinitely thick heavy metal converter targets as a function of the electron energy

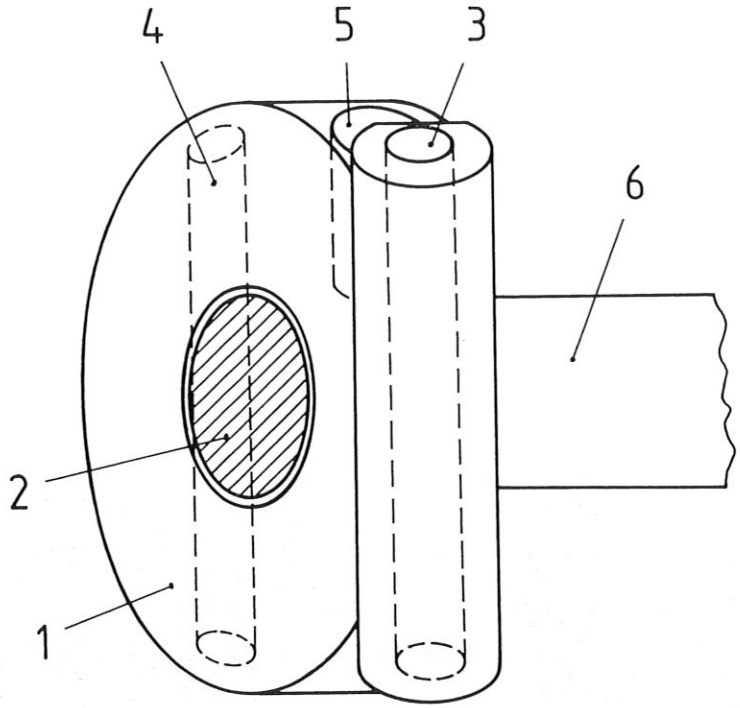

Fig. 3.18a

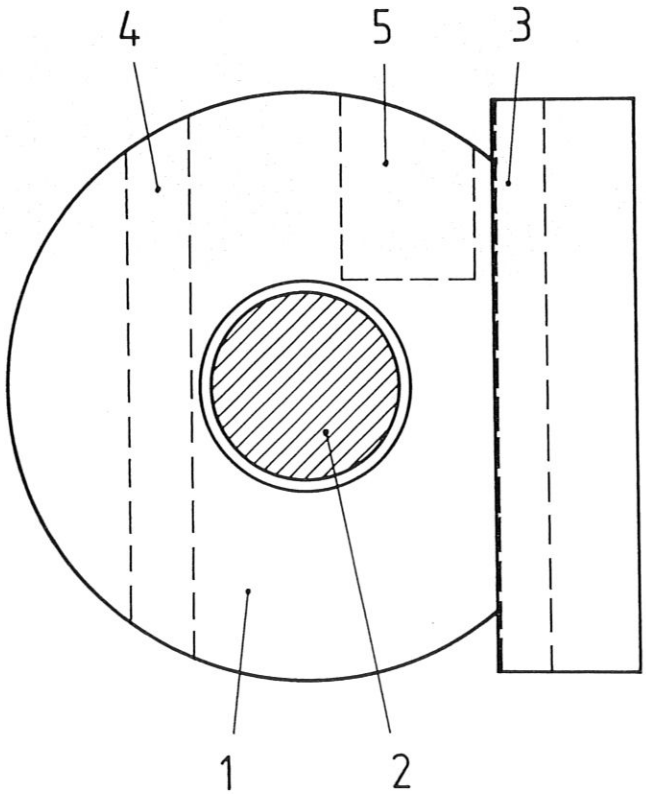

Fig. 3.18b: Photoneutron moderator (plastic material); 1 = moderator annulus, 2 = bremsstrahlung target (photoneutron source), 3 = sample rabbit channel for thermal neutrons, 4 = dto. for fast neutrons, 5 = position for large sample volumes, 6 = electron beam tube

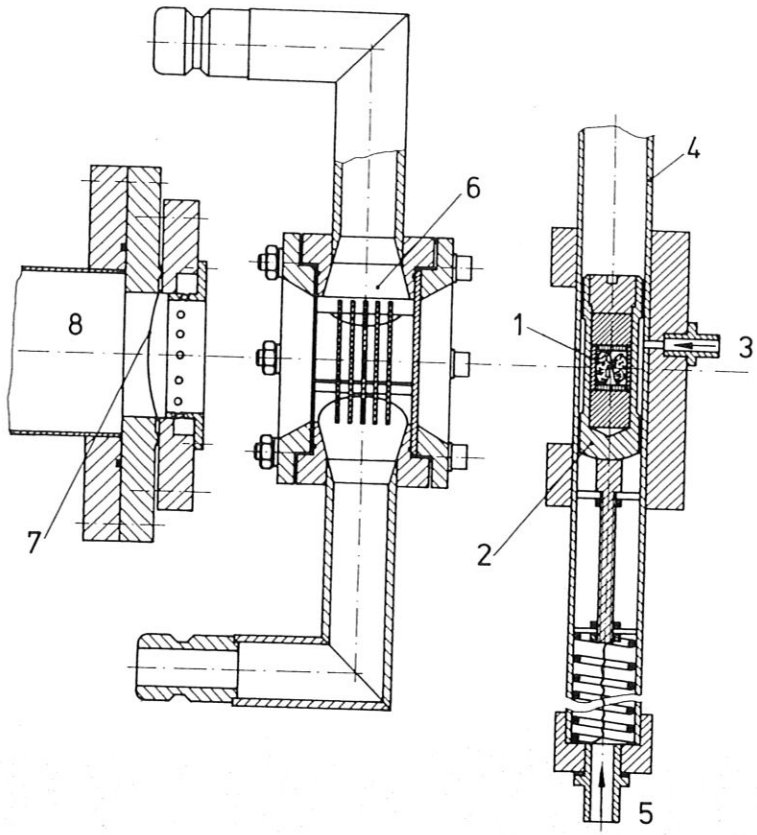

Fig. 3.19: Irradiation facility at the BAM Linac: explanations see text

3.5 Typical irradiation facility

As is explained above, a converter material of high atomic number is required. Furthermore, it has to resist long-time heating and large radiation doses. Moreover, chemical stability is obligate to avoid subsequent decomposition by chemical reaction with any matter which is in close contact with the target, e.g. cooling water.

The irradiation setup used at the 35 MeV electron linear accelerator of the Bundesanstalt für Materialprüfung in Berlin is shown in Fig.3.19. The electron beam ejected from the accelerator tube (8) through the titanium electron beam window (7) impinges on the target (6) after a distance of a few centimeters. The converter consists of seven tantalum discs with spaces of about one mm between so as to enable efficient cooling water flow. The power load of the converter is about 4 KW under routine operation of the accelerator. The total converter thickness is sufficient for complete absorption of the 35 MeV electron beam. About 6 cm behind the converter the sample position is located. The sample and the reference material are enclosed in an aluminium capsule and transported by compressed air to and from the irradiation position. The axis of the capsule is perpendicular to the photon beam. The terminal of the pneumatic tube (4) is equipped with an inlet for compressed air flow injected tangentially into the tube (3) so as to cool the sample rabbit (2). This air flow is also used to make the sample rabbit rotate so as to provide uniform irradiation of the sample to be analysed and the reference material (1). The electron beam and thus the bremsstrahlung cone are adjusted with respect to the sample using the steering coils of the accelerator. The beam position is monitored by a fluorescent view which is observed by a television camera.

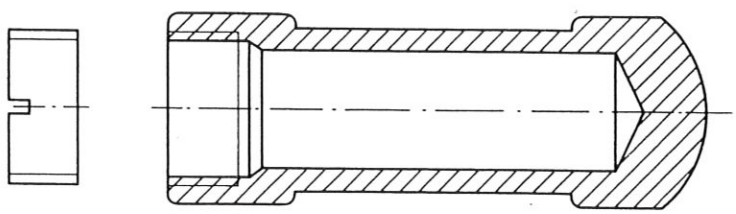

Fig. 3.20: Sample rabbit; inner diameter = 16 mm

At the lower end of the pneumatic tube another inlet for compressed air is provided (5) by which, after irradiation, the rabbit is transported to the terminal in the radiochemical laboratory. There the irradiated matter is removed from the rabbit and processed further as desired.

The inner space of the rabbit (see Fig.3.20) is about 5 cm long at a diameter of 16 mm. The rabbits as well as nearly all parts of the tube system near the converter are made of aluminium since this material is inexpensive and durable against heat, radiation and mechanical shock. Moreover, it does not accumulate cumbersome activity during activation; all radioisotopes produced through bremsstrahlung or photoneutron irradiation are either reasonably short-lived (^{26m}Al, ^{28}Al, ^{27}Mg) or extremely long-lived (^{26}Al) or the corresponding nuclear reactions have very small activation cross sections at the irradiation conditions (^{24}Na, ^{22}Na). No other material is thinkable which is comparably suitable.

The entire sample transportation system is represented schematically in Fig. 3.21. The arrival of the rabbit at the target terminal is monitored acoustically by a microphone; this kind of monitoring has proved most reliable since the microphone does not have to be installed in the close vicinity of the radiation source and so does not suffer from heat and radiation attack.

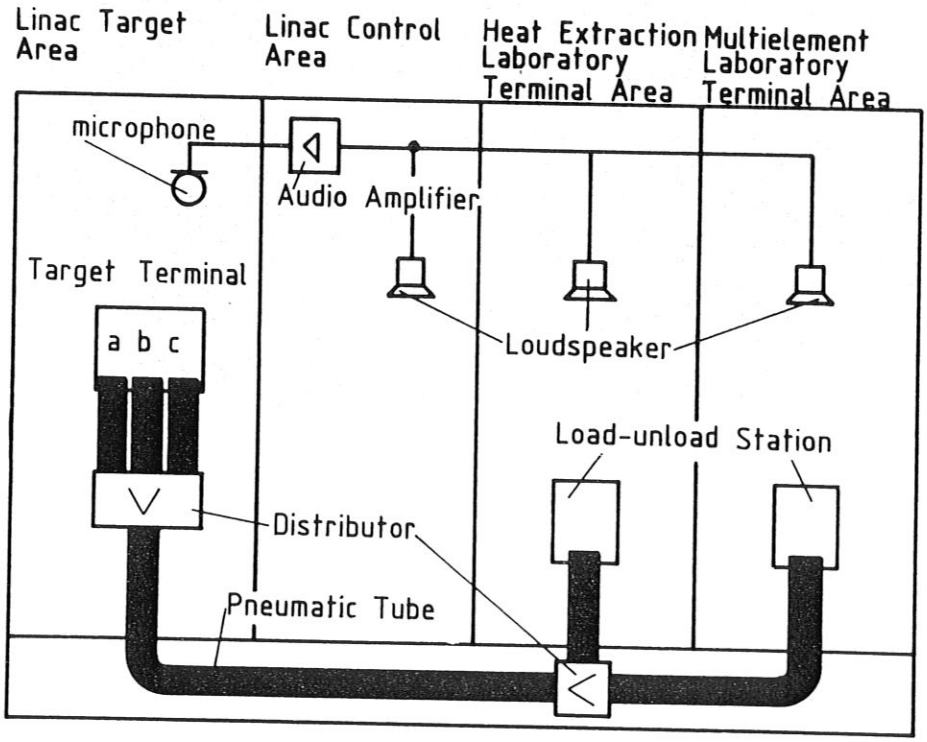

Fig. 3.21: Schematic representation of the pneumatic tube sample transfer system at the BAM Linac

Photoneutrons are also used for activation analysis (see 3.4). Therefore, the sample rabbit can be transported to different irradiation positions through pneumatic tube branches (a, b, c in Fig.3.21) selectable by a distributor. Figures 3.18a and b show the plastic material photoneutron moderator equipped with several irradiation positions so as to enable photoneutron irradiations under different conditions. In the tangential channel (4) a pneumatic tube terminal is installed in the "fast neutron" position (c in Fig.3.21). Here photoneutron irradiations can be conducted at the maximum achievable flux density (several 10^{10} cm^{-2} s^{-1}) at a cadmium ratio of about unity. In the "thermal neutron" position (a in Fig.3.21) a better cadmium ratio is achieved (about 20) favoured by a plastic layer of about 5 cm water-equivalent between the photoneutron source and the sample position. A third irradiation position (5) is provided to enable large volume (maximally about 150 ml) irradiations. The positions 3 and 4 (and, of course, the photon irradiation position behind the target; 2 in Fig.3.21) are equipped with pneumatic tube terminals. Another pneumatic tube distributor is provided so as to enable transfer of the irradiated sample to either the heat extraction laboratory where the light elements (C, N, O, F) exclusively are analysed (see 6.1) or the multielement laboratory where instrumental multi-element analyses with help of semiconductor spectrometers are performed (see 6.2).

Other irradiation facilities used for photon activation analysis at electron linear accelerators are described in Refs.[325-329] and many others.

3.6 Conclusion

At this point an obvious difference between activation analysis with reactor neutrons and methods requiring accelerator irradiation should be pointed out. Both particle sources are basically designed for use in a physical or, in the case of accelerators, in a medical laboratory. However, whilst neutron activation analysis is a priori included in the construction concept of a nuclear research reactor, activation analysis normally has a comparatively low priority level in the daily operation schedule of an accelerator (see also[330]). Usually radiochemistry - activation analysis in particular - is only restrictedly allowed in accelerator laboratories in terms of irradiation time and space requirement. As far as the authors know, there is only one electron linac which is used almost exclusively for photon activation analysis, namely the accelerator in the Bundesanstalt für Materialprüfung which was used for the present work.

This remarkable difference is not explainable by the historical development of the methods; all kinds of activation analysis have about the same date of birth (see Ch.1). Probably the extremely high sensitivity of thermal neutron activation analysis for a number of elements plays a certain role.

However, in some cases photon activation analysis is much more suitable than neutron activation methods as is shown in the following chapters.