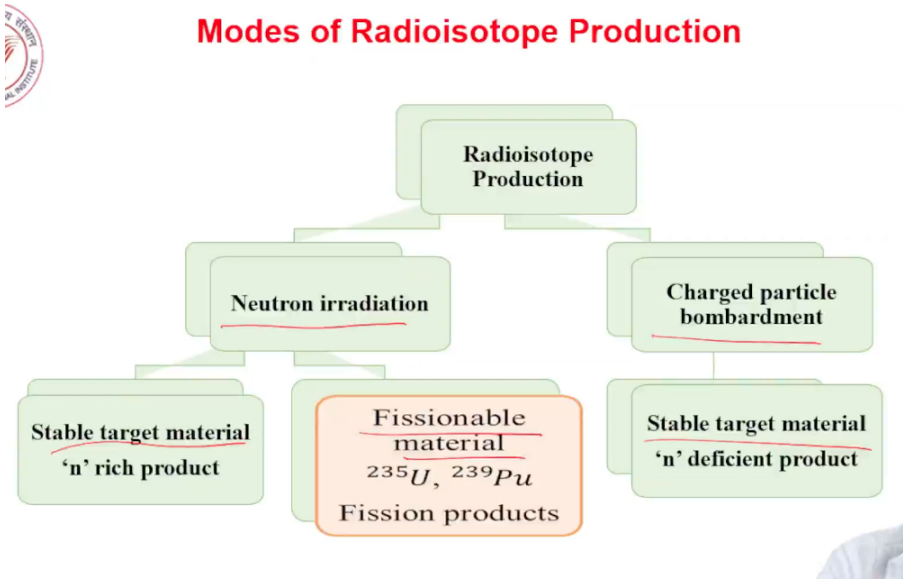
Production of radioisotopes by neutron irradiation

B.S.Tomar

Homi Bhabha National Institute

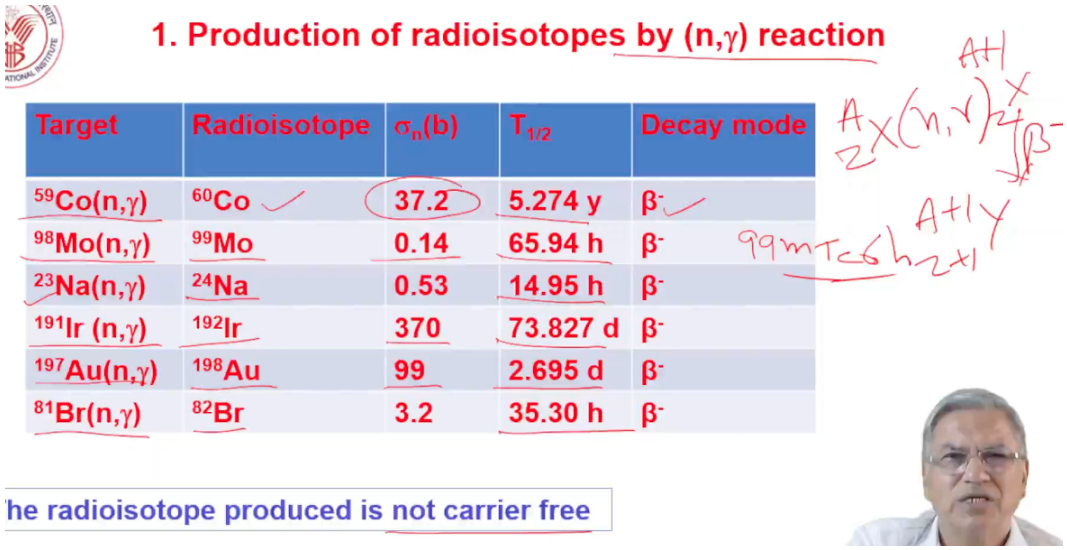
Lecture-15, Module-1

Hello everyone. So far, we have discussed the different aspects of nuclear chemistry including the fundamentals of radioactivity, nuclear structure, nuclear models, different types of decays, detection of different types of radiations and also nuclear reaction mechanisms, different types of nuclear reactions including fission and fusion. And in the nuclear fission and fusion, we discussed about how to tap the energy from the nuclei to produce electricity and that for the fusion efforts are going on. So we will now switch over to another aspect of nuclear science and technology that is the applications of the radioisotopes in different areas. So we will not go into details of applications, but how to produce the radioisotopes for applications in many, many areas. So radioisotopes applications are one area of nuclear science and technology, nuclear fission and fusion is another area. So we will now discuss the production of radioisotopes which are useful in many, many applications.



So I have just given in a nutshell what are the different ways by which we can produce the radioisotopes. So mainly we have at our disposal the reactors or other types of neutron sources. So we can irradiate a target material with neutrons to produce radioisotopes or can have accelerators, cyclotrons, electrons, Van der Graaff, etc., wherein we bombard the target by charged particle to produce radioisotopes.

Using neutron radiation, when we irradiate the stable targets, we get neutron rich products which are invariably β- active. Or we can, suppose we irradiate the actinides, the fissionable materials, then by nuclear fission of these heavy actinide elements, we can produce fission products which are also many of them are useful in different areas. With charge particle, we irradiate the stable target materials and since we are bombarding with charged particles, we produce neutron deficient radioisotopes which will be emitting the β+, and /or electron capture. This in nutshell gives you different ways by which we can produce radioisotopes. So now we will first discuss the neutron based radioisotope production where there are different processes, different ways by which we can produce radioisotopes.



So we will now discuss using neutrons, what are the different ways by which we can produce radioisotopes. The first and the most common is by (n, γ) reaction. So you all know that if you irradiate a target material, let us say ${}^{A}X_{Z}$ by (n, γ), then we get ${}^{A+1}X_{Z}$ and this invariably is β- active, so we get ${}^{A+1}Y_{Z+1}$. So this is how we can in fact increase the atomic number of a target material by irradiating with neutron. So this is the most common route of radioisotope production in a reactor or it can be even other source like you can have photon neutron source or a D-T neutron source or you can have Californium-252 fission as source of neutrons.

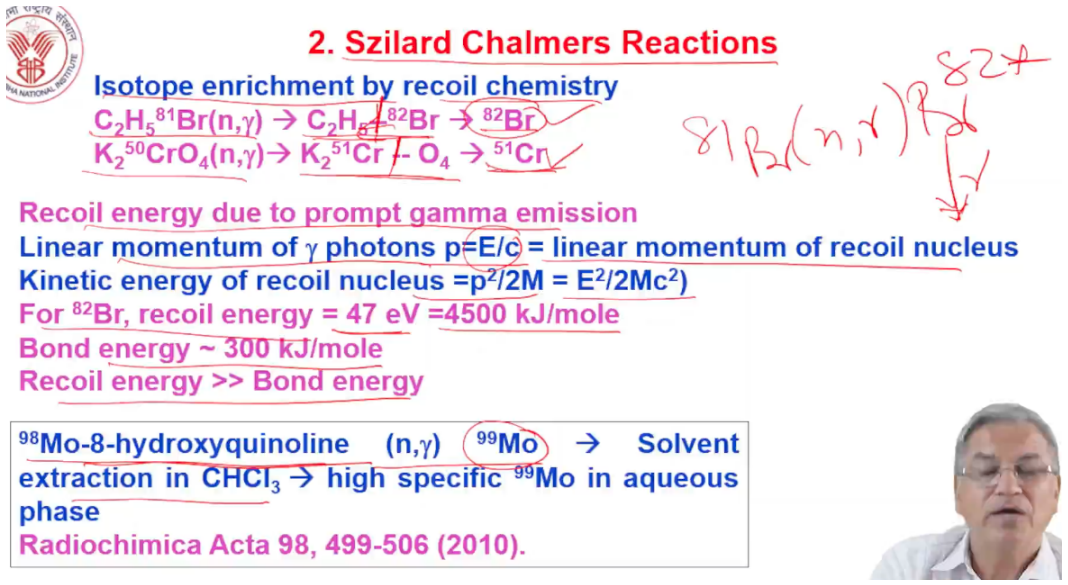
But the most common source of neutrons for radioisotope production is a nuclear reactor where you can at the same time irradiate several targets and produce large amount of activity that we discussed in the nuclear reactions. So what I have listed out is the isotopes which are very much used in some area or other. So, ^{59}Co(n, γ) gives you ^{60}Co having half life of 5.274 years and which is undergoing β- and this ^{60}Co then after β- ends up in the excited states of ^{60}Ni and which gives you two gamma rays 1172 and 1332 keV. These are the gamma ray, because of this gamma ray the ^{60}Co finds lot of

applications in irradiation of materials, many applications you will find in industry and other areas.

This is a gamma source, then we have ^{99}Mo by natural ^{98}Mo(n, γ) reaction. It can also be produced by other routes which we will discuss later on. It has got a low cross section 0.141 barns, half life is 66 hours. It is a parent of the ^{99m}Tc which is 6-hour half life and that is used in nuclear medicine for diagnosis. Again ^{23}Na(n, γ)^{24}Na, ^{24}Na is a hard gamma emitter which emits high energy gamma rays and having a half life of 15 hours it can be used as a tracer in many applications.

^{192}Ir produced by ^{191}Ir(n, γ) is another isotope having 73 days half life and cross section is quite good. So iridium-192 actually emits different energies in hundreds of keV and for radiography commonly industrial radiography of many products like non-destructive testing of equipment is used. If you want to do radiography of a heavy material, heavy machine tool, go for high energy gamma ray from cobalt 60. If it is a small one, you want low energy gamma rays so use ^{191}Ir. ^{197}Au gives you (n, γ) ^{198}Au, 2.695 days, cross section is also quite good and ^{198}Au is used as a tracer in many applications in industry. ^{81}Br(n, γ)^{82}Br you can even prepare a gaseous product isotope tracer or you can use ethyl bromide as tracer so if you want to trace the path of a liquid or a gas you can use ^{82}Br tracer. So this is 35 hours. In industry ^{82}Br has lot of applications when you want to trace the path of a petrochemical. There is a leak of petrochemical in a long underground pipeline and you want to know where the leakage is. You use this ^{82}Br as a tracer and you can trace where the leakage is. So these are the different ways of (n, γ) way by which you can produce different isotopes.

And one important aspect of this is that you irradiate the target material, the radioisotope also is of the same element and so bulk of the material remains the same elements and therefore they are not carrier free. All the isotopes are all the atoms are not of radioisotope but there is a bulk. Suppose you irradiate 1 gram of cobalt then only maybe picogram or nanogram or macrogram of cobalt will be converted into cobalt 60. Bulk of it is still remains cobalt 59. So that is what the meaning that it is not a carrier free. So there is a carrier associated. Carrier means the stable target element. So this is the drawback of this (n, γ) route that you do not get a carrier free radioisotope. And carrier free means only the radioisotopes, other isotopes or stable isotopes are not available in that.



Okay, the another way is called the Szilard-Chalmers reaction. So the necessity to produce carrier free radioisotope but using (n, γ) reaction, you know, led to this kind of developments in the radioisotope production. So this can also be called as isotope enrichment by the recoil chemistry. That means you take such a compound that after the n,γ reaction, the radioisotope is detached from the compound and you have a carrier free radioisotope. So this is more of ideal way of speaking but we will see that you can increase the specific activity of the radioisotope by using Szilard-Chalmers.

When I say specific activity means per gram of the target, how many atoms are radioactive? So what is the activity per gram of target that is for the specific activity. So just to give you an example of Szilard-Chalmers, Szilard and Chalmers actually developed this methodology of producing radioisotope. So for example, you want to get ^{82}Br in the carrier free form. By ^{81}Br (n, γ), bulk of the bromine remains ^{81}Br and so ^{82}Br will be present in a large amount of ^{81}Br. But if you take organic molecule like ethyl bromide, now bromine is bound to ethyl group by a covalent bond and when you have an n,γ reaction, then one of the bromine atom will become ^{82}Br. And what happens that because of the recoil, when this (n, γ) reaction product, ^{82}Br, will be excited. So ^{81}Br (n, γ) ^{82}Br and it is excited. So when it is emitting a gamma ray, this gamma ray will give a recoil ^{82}Br and it will be snapped from the organic moiety. So you will have ^{82}Br which is not bound to ethyl group. So now bulk of the material remains ethyl bromide, which has got different chemistry and the ^{82}Br may become bromide ion and which has different chemistry. So you can separate ^{82}Br from ethyl bromide and you will get ^{82}Br.

Similarly, potassium chromate, $K_2{}^{50}CrO_4$ you irradiate with neutrons to get with $K_2{}^{51}CrO_4$. Now this chromate ion, $(CrO_4)^{2-}$, that Cr-O bonds may get broken when the gamma ray gives a recoil to Cr^{51} and because of the breaking of this bond, you will see free ^{51}Cr which may stabilize at CrO_3^+ and therefore this has got different chemistry than

the bromate ion which is CrO_4^{2-} and that is how the target and the radioisotope have different chemistry, then you can do separation and get career free ^{51}Cr.

So what is the concept behind the recoil chemistry? This is because the prompt gamma that is emitted from the excited nucleus formed by n,γ reaction, then when the gamma ray gives a recoil to the atom then the linear momentum of the gamma photon, the momentum is conserved. So we can calculate what is energy that the recoil atom will get.

So the momentum is nothing but E_γ/c, energy of the gamma ray photon and the speed of light and that same will be the momentum of the recoil. So whatever the momentum of the photon, the recoil nucleus will get that much momentum to preserve the momentum. So if you see this is the momentum of the recoil then the energy will be $P^2/2m$, so it will be $E_\gamma^2/2mc^2$. We can see here we have a 5 MeV gamma ray getting emitted from the compound nucleus. The 5 MeV gamma ray will give it a recoil.

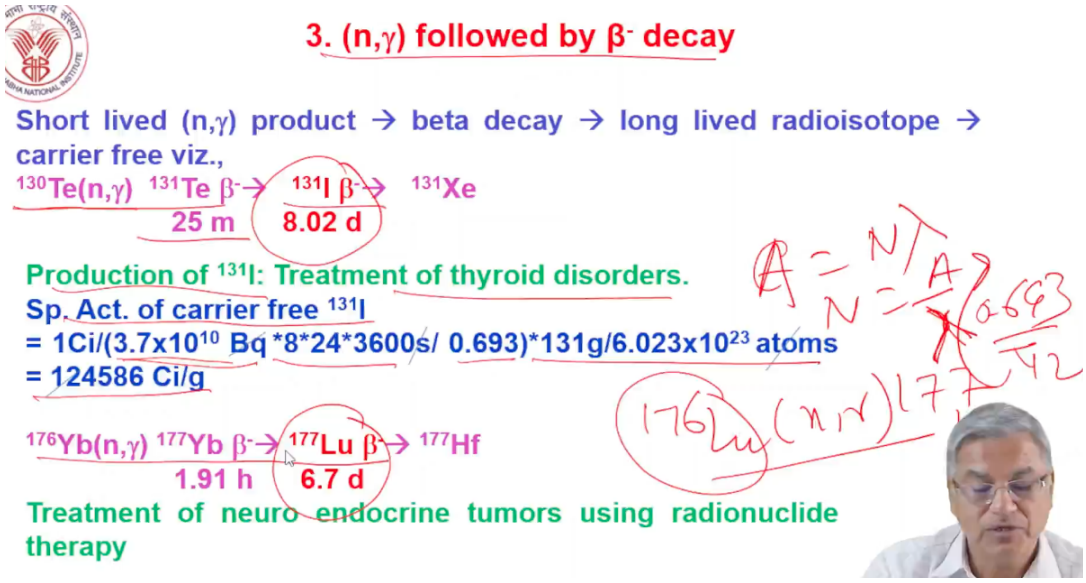
Let us see what is the value. So for ^{82}Br, the recoil energy will be 47 eV. You can see here the proper gamma ray of emitted from excited ^{82}Br will give a recoil to this let us say ethyl bromide and then this rather than ^{82}Br will get a kick of 47 eV and that will lead to the detachment of the ^{82}Br. And this is per atom, so in terms of chemical energy, kilojoule per mole, it corresponds to 4500 kilojoule per mole. We can see that typical bond energies are of the order of hundreds of kilo Joule per mole and the recoil energy received by the bromine will be thousands of kilojoule per mole.

The net result of this recoil will be that since the recoil energy is more than the bond energy, the recoil atoms will get detached from the parent molecule and you will have now to stop as an accelerated atom and it will stabilize in its most stable oxidation state. So like bromine will become bromide, if it is an organic solution, so you will find that bromine will have different chemistry than ethyl bromide. Similarly, the chromium will have different oxidation state than chromate, so we will have Cr^{3+} positive ion, bromine as Br^- ion. So then we can go for chemistry, separate the two atoms from the stable target.

In fact, a very interesting experiment that we had done one such experiment was to get molybdenum-99, molybdenum-98(n,γ) gives you molybdenum-99 and bulk of the molybdenum will be natural molybdenum, but molybdenum-98 is very small abundance.

And so if you irradiate molybdenum in a complex organic molecule like 8-hydroxyquinoline, then this (n, γ) reaction will give recoil to molybdenum-99 and the ^{99}Mo detached from the organic complex will be at a different oxidation state. So you can actually do solvent extraction of this molybdenum-hydroxyquinoline chloroform and molybdenum-99 will be left in the aqueous solution. So you can enrich molybdenum-99 in aqueous solution, whereas the bulk of the molybdenum-98 hydroxyquinoline will

remain in the organic phase. So that is how you can increase the specific activity of a (n, γ) product.



Another way of producing the carrier-free radioisotopes is (n, γ) followed by beta decay. It is not always possible, but there are many examples where it is possible and I will give you some examples of that. For example, ^{130}Te (n, γ) gives you ^{131}Te and which is also radioactive and undergoes β- decay to iodine-131. So iodine-131 is the isotope of interest for thyroid imaging because it is emitting 364 keV gamma ray. So if you want to image the thyroid of a patient, you just give sodium iodide solution tagged with ^{131}I activity.

So how do you get a carrier-free iodine-131? You simply irradiate tellurium metal target and tellurium will give you iodine, which easily you can do radiochemical separation of iodine from the tellurium because they have totally different chemistry. So this is (n, γ) followed by β- decay. The (n, γ) product of ^{131}Te, which is 25 minutes' half-life. You allow for this to decay. So in about four half-lives, it can decay to ^{131}I and then you can do the iodine chemistry from the tellurium target.

So tellurium-131 is actually widely used in treatment of thyroid disorders and typical specific activity of this ^{131}I you can calculate. Suppose you have all now here, there are no other atoms of iodine other than 131. So 100% carrier-free.

So if you have one Curie of ^{131}I, then you can calculate the number of atoms. So you can see, so activity (A) equal to Nλ. So N = A/λ. So you know the half-life, so you can substitute for the λ in terms of the $0.693/T_{1/2}$ and this $T_{1/2}$ will go up. That's what I have done here.

Sp. Act. of carrier free ^{131}I

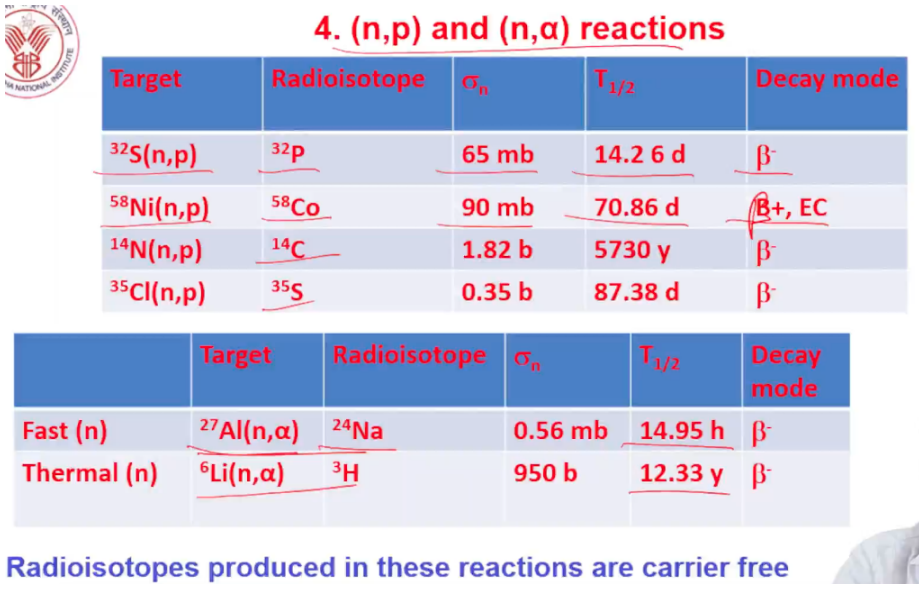
= 1Ci/(3.7x10^{10}*8*24*3600/ 0.693 atoms)*131g/6.023x10^{23} atoms = 124586 Ci/g
So the specific activity of iodine-131 generated in this tellurium irradiation will be about 1.24 lakh Curie per gram, very, very high specific activity because all radioactive atoms are of 131 iodine. There is no stable iodine that is ^{127}I in the ^{131}I sample.

So these are the methods by which you can get carrier-free radioisotopes by neutron activation.

Another example of this is ^{177}Lu that is produced by ^{176}Lu(n, γ), but this lutetium will be having bulk of ^{176}Lu and also you have ^{175}Lu which is the dominant. But here you will find that if you irradiate ^{176}Yb and ^{176}Yb(n, γ)^{177}Yb which is undergoing β- decay to ^{177}Lu.

So though they are adjacent rare earths, you can still separate them by careful radiochemical separations and so you can have carrier-free ^{177}Lu, 6.78 days half-life which is used in treatment of neuroendocrine tumors, basically for the diagnosis and therapy of the tumors in the glands.

These are the examples of (n, γ) followed by β- decay of the production of radioisotopes. In the case of light Z elements, like low Z, so up to you can say mass number 40, 50 or so where the barrier for the emission of charged particles are low, so you can have (n, p) and (n, α) type of reactions.



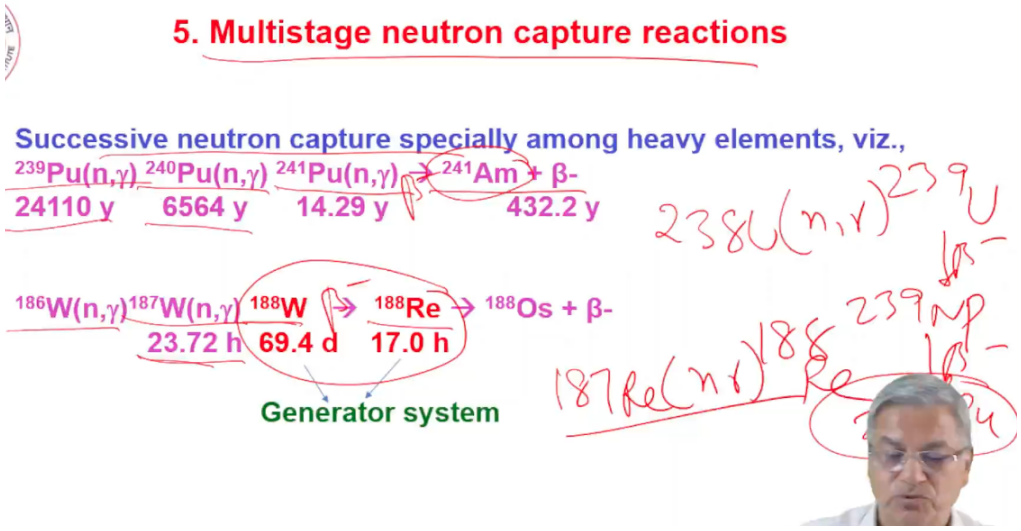
So these reactions are possible only for the lighter targets because for heavier targets emission of proton and alpha is hindered due to high emission barrier.

So you can irradiate like sulfur, if you irradiate sulfur compound, sulfate, sodium sulfate, you will get phosphorus-32

$^{32}S(n,p)\ ^{32}P$,

and then phosphorus-32, the cross-sections are given, half-life of phosphorus-32 and the mode of decay is there. So phosphorus-32 is utilized in many applications given for the skin, skin catches for the treatment of skin cancer. Similarly, you have ^{58}Co from ^{58}Ni (n, p) reaction, it is a small cross-section and you have the 70 days half-life of ^{58}Co and it is emitting beta plus or an electron capture. ^{58}Co is used as a tracer in many applications. Then you have carbon-14 and sulfur-35 which are also used as radio tracers in organic chemistry you can use carbon-14 in organic reactions, sulfur-35 also, sulfur-35 labeled compounds you can use in synthesis of compounds containing sulfur.

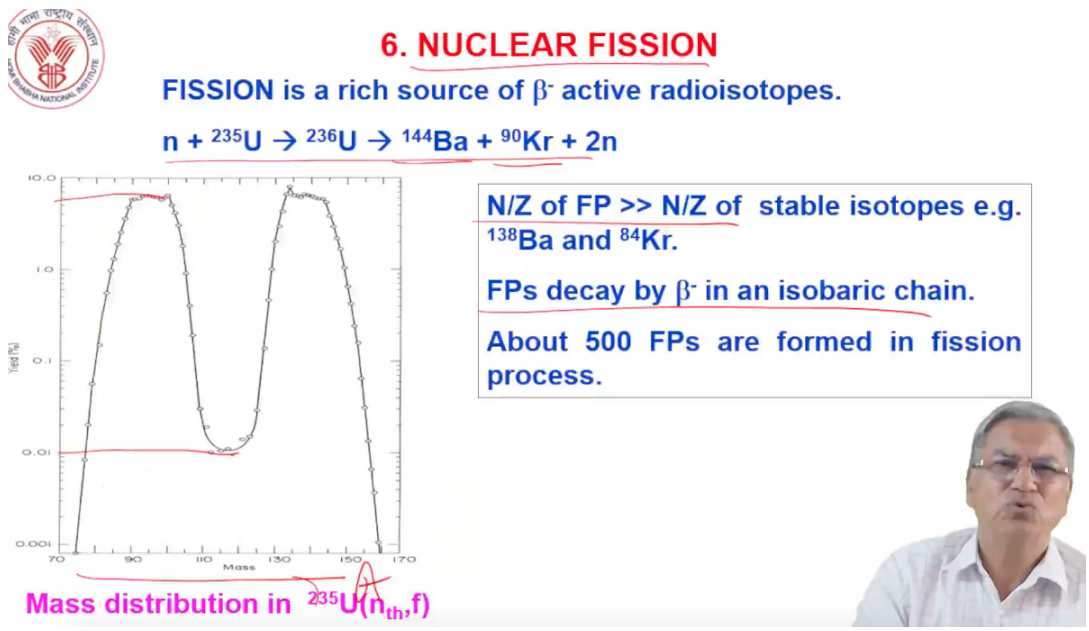
So for neutron induced reactions wherein the proton and alpha are emitted mostly involved in the lighter elements and certain isotopes are very useful so you go for these methods. Then with the aluminum you have (n, α) but that requires fast neutrons because they have got threshold reactions so Q value is negative so thermal neutrons you will not do, you require higher energy neutrons, energy more than a keV we call fast neutrons and so you want to produce sodium-24. See normally sodium-24 you can produce by $^{23}Na(n, \gamma)$ reaction but if you want carrier-free sodium-24 you go for this route, $^{27}Al(n, \alpha)^{24}Na$, 15 hours half-life and even with thermal neutrons you can produce tritium then $^{6}Li(n, \alpha)^{3}H$ you can get tritium and this tritium in fact this route you use to produce tritium for strategic applications and for that matter you know even for normal applications you can use tritium. So you can see here if you do this method (n,p), (n,alpha) type of reactions then by this method since the target element and the radioisotope produced have different chemistry due to different Z value so you can do chemistry and then you have these isotopes as carrier free. These are the methods by which you can even use neutrons to produce carrier-free radioisotopes.



Then there are certain methods called multi-stage neutron capture. Multi-stage means after one neutron capture whatever isotope is formed it will continue to capture neutrons and you will get a much higher isotope of same element or there can be beta decay. So successive neutron capture happens when the flux is very high so that the radioisotopes that are produced can further capture neutrons to next isotope. So this in fact is happening you know if you have a nuclear reactor, power reactor. For example, if you have $^{238}U(n,\gamma)^{239}U$. If you see it will undergo beta minus to ^{239}Np, which in turn will undergo beta minus to ^{239}Pu. So in the reactor if you have bulk target natural uranium has 99.3 percent ^{238}U, so you have a continuous production of ^{239}Pu. And this ^{239}Pu has a half life of 24,000 years.

It will capture neutron to get ^{240}Pu. ^{240}Pu is having half life 6500 years. It will capture another neutron ^{241}Pu and which is emitting beta minus to ^{241}Am. You can see all the isotopes have half lives in years. So if you want to produce ^{241}Am which is very, very useful in many applications. So in the reactor you will find lot of plutonium being produced you have a ton of fuel then about a kg of plutonium will be formed. And this is continuously capturing neutrons to form higher isotopes. So spent fuel of a reactor, such reactor or power reactor will always have plutonium and americium. This is the way by multiple neutron capture.

Similarly, in fact the same concept we do it in producing ^{188}W. ^{188}W is a parent of ^{188}Re and ^{188}Re can also be produced by (n, γ). But this is not carrier free and you require for many applications carrier free radioisotope. So if you irradiate ^{186}W by (n, γ) you get ^{187}W having half life of 23.7 hours and this can capture another neutron to get ^{188}W. This ^{188}W will undergo the beta minus decay to ^{188}Re. So this is like a generator system. 69 days half life of ^{188}W going ^{188}Re. So you can have a column containing ^{188}W and you can milk ^{188}Re. So this is the way whatever rhenium you will get will be carrier free. But since the half life is short it requires a very high flux reactor. Otherwise the production rates will be much lower than the decay rate. So typically 10^{15} $n/cm^2/s$ is the flux required to produce this kind of isotopes.

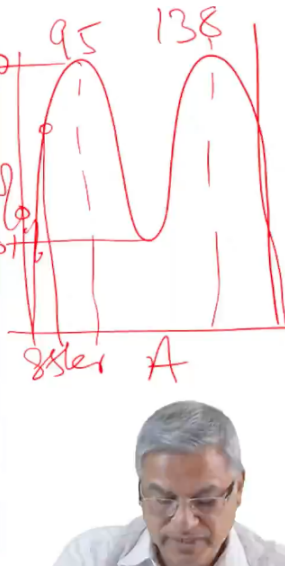


Of course we have discussed nuclear fission in various lectures also. So nuclear fission is a very, very rich source of β- radioactive isotopes. You already know that irradiation of uranium with thermal neutrons gives several pairs of fission products like one pair being $^{144}Ba+^{90}Kr$. And this is the mass distribution as a function of mass number, the fission yield. Fission yield is maximum about 8%, minimum about 0.01 at symmetry. The lower yields are also there. So it is asymmetric mass distribution. So you will have a large number of fission products produced in fission and many of them are long lived and have a lot of applications. So normally the fission products have higher N/Z compared to their stable isotopes and so most of them are β- active. There is a chain of isobars wherein the beta decay takes place and you will have about 500 fission products that will be formed in the fission process.

So if you can pick up the ones which find applications in many areas and then you can go subsequently for the radiochemical separation of individual isotopes used in different areas.

Long lives fission products

Fission product	$T_{1/2}$	Decay mode	Other route
^{85}Kr	10.756 y	β-, γ	(n,γ)
^{90}Sr	28.79 y	β-	
^{99}Mo	65.94 h	β-, γ	(n,γ)
^{106}Ru	373.6 d	β-, γ	
^{131}I	8.04 d	β-, γ	Te(n,γ)
^{140}Ba	12.752 d	β-, γ	
^{140}La	1.678 d	β-, γ	(n,γ)
^{144}Ce	284.9 d	β-, γ	
^{147}Pm	2.62 y	β-	



Now this is the list of radioisotopes that are produced in fission, long-lived fission products. ^{85}Kr, so you can see here if you try to draw the mass yield curve, A vs yield, typically about 8% to 0.01%. Now you can see typically this is around 95 and this is around 138. So that gives you an idea what will be the yield of the ^{85}Kr. ^{85}Kr will be somewhere here. And this is a gaseous radiotracer. So suppose you want to do a tracer application wherein you want to see the leakage in a gas pipeline, you can use ^{85}Kr.

You will have half-life of 10 years. You can also produce by ^{84}Kr(n, γ) but irradiating a gaseous target in the reactor. Instead of that you can have a fission product that will irradiate the nuclear uranium and you can just do distillation of the fission product Kr. Very high activity of ^{85}Kr can be produced.

^{90}Sr is another isotope which is having very long half-life. So it is present in the irradiated fuel and it is parent of ^{90}Y which is 64 hours so it is a generator system for producing fission products ^{90}Y.

^{99}Mo also can be produced in fission and you will have fission molybdenum. In fact, there are plants which are available for producing molybdenum-99 by fission route. ^{106}Ru is used in ocular cancer treatment, ^{131}I for treatment of thyroid disorders, ^{140}Ba as tracer and then ^{140}La is used as excellent tracer for silt movement, ^{144}Ce also for the tracer and ^{147}Pm is a pure beta emitter and it is used in the luminescent paints. So there are many applications of radioisotopes and most of them are available in the spent fuel. So if you irradiate uranium you get all of them and you can then do chemistry depending upon their half lives you can separate them and use them in different applications.

So I will stop here and I will take up the other method of charged particle irradiation in the next part. Thank you.