Production of Pb-203 from Target Manufacturing to Chemical Separation Tl/Pb

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Production of Pb-203 from Target Manufacturing to Chemical Separation Tl/Pb

I have prepared subtitles for you because my accent is very strong. Indeed, I am hard of hearing. My presentation is titled : production of Pb-203 from target manufacturing to chemical separation thallium and lead.

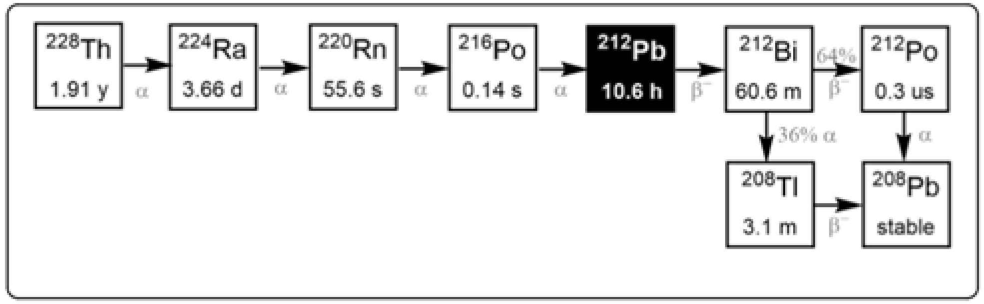
Why Pb-203?

Pb-203
$T_{1/2}$ = 51.92 h
Emission γ : energy = 279 keV
intensity = 81%

Pb-203 is compatible for SPECT

+ Pb-203 can be used for theranostic with Pb-212

Pb-212
$T_{1/2}$ = 10.64 h
Emission two β^- particles and one α particle during its decay
chain Pb-212 -> Bi-212 -> Pb-212/Tl-208 -> Pb208



Pb-212 is compatible for therapy β^- and α

Why Pb-203? This radioisotope has a half life period of 52 hours and emits the γ energy at 279 keV with an intensity of 81%, it is adapt for the medical imagery SPECT. Furthermore, it can be used for thernostic with the same element but different isotopic : Pb-212. This radioisotope emits two β^- particles and one α particle during its decay.

Why Pb-203?

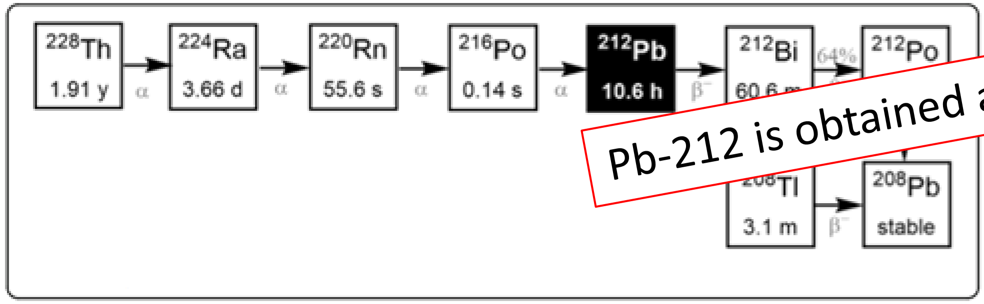
Pb-203
$T_{1/2}$ = 51.92 h
Emission γ : energy = 279 keV
intensity = 81%

Pb-203 is produced in accelerator

Pb-203 is compatible for SPECT

+ Pb-203 can be used for theranostic with Pb-212

Pb-212
$T_{1/2}$ = 10.64 h
Emission two β^- particles and one α particle during its decay chain Pb-212 -> Bi-212 -> Pb-212/Tl-208 -> Pb208



Pb-212 is obtained after decaying of Th-228

Pb-212 is compatible for therapy β^- and α

Pb-203 is produced in an acceletor and Pb-212 is obtained after Th-228 is decayed.

Which raw material to produce Pb-203?

200Pb 21.5 h ε = 100.00%	201Pb 9.33 h ε = 100.00%	202Pb 52.5E+3 y ε = 100.00%	203Pb 51.92 h ε = 100.00%	204Pb ≥ 1.4E+17 y 1.4% α	205Pb 1.73E+7 y ε = 100.00%	206Pb STABLE 24.1%
199Tl 7.42 h ε = 100.00%	200Tl 26.1 h ε = 100.00%	201Tl 3.0421 d ε = 100.00%	202Tl 12.31 d ε = 100.00%	203Tl STABLE 29.524%	204Tl 3.783 y β^- = 97.08% ε = 2.92%	205Tl STABLE 70.48%
198Hg STABLE 9.97%	199Hg STABLE 16.87%	200Hg STABLE 23.10%	201Hg STABLE 13.18%	202Hg STABLE 29.86%	203Hg 46.594 d β^- = 100.00%	204Hg STABLE 6.87%

^{200}Pb is the radioactive impurity principal, its production must be avoided

Which raw material should be used to produce Pb-203? Before answering this question, here is the chart of nuclides to study the different isotope of lead that can be produced after irradiation. We can see that we should avoid producing one isotope, Pb-200. Indeed, the half-life period of Pb-200 is the near half of the half-life period of Pb-203, the value is 21.5h.

Which raw material to produce Pb-203?

200Pb 21.5 h ε = 100.00%	201Pb 9.33 h ε = 100.00%	202Pb 52.5E+3 y ε = 100.00%	203Pb 51.92 h ε = 100.00%	204Pb ≥ 1.4E+17 y 1.4% α	205Pb 1.73E+7 y ε = 100.00%	206Pb STABLE 24.1%
199Tl 7.42 h ε = 100.00%	200Tl 26.1 h ε = 100.00%	201Tl 3.0421 d ε = 100.00%	202Tl 12.31 d ε = 100.00%	203Tl STABLE 29.524%	204Tl 3.783 y β^- = 97.08% ε = 2.92%	205Tl STABLE 70.48%
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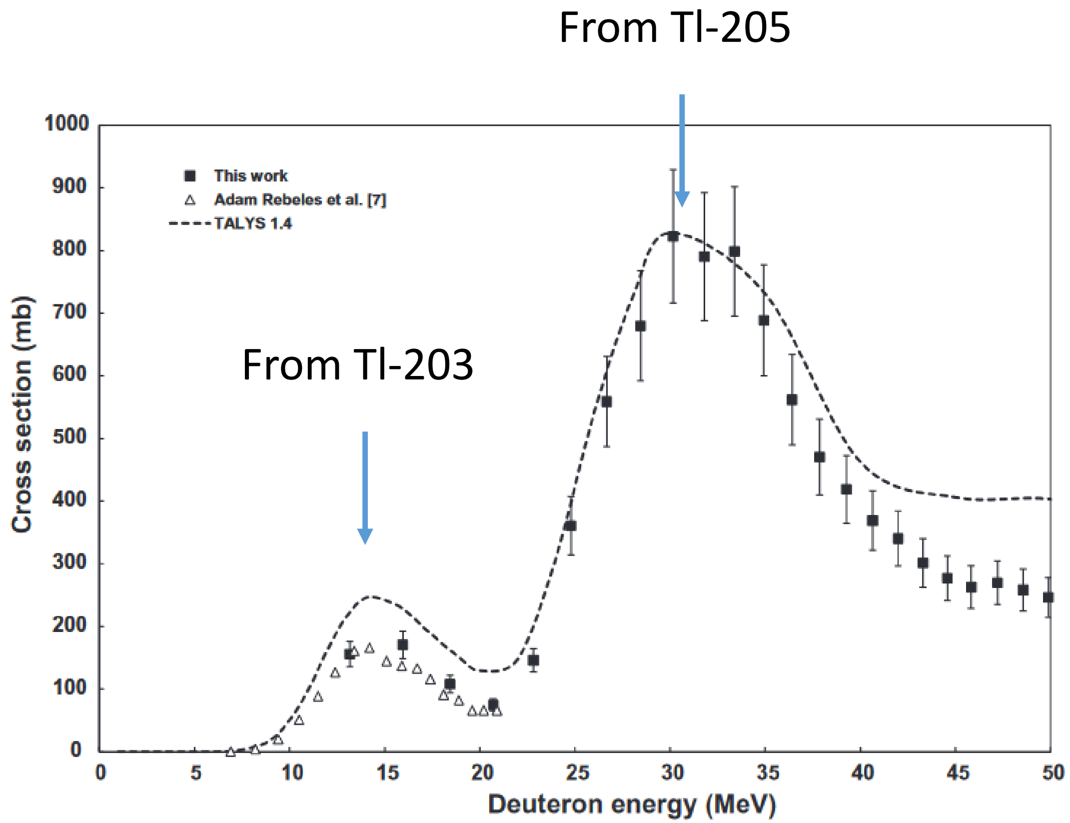
^{201}Pb is the radioactive impurity principal, its production must be avoided

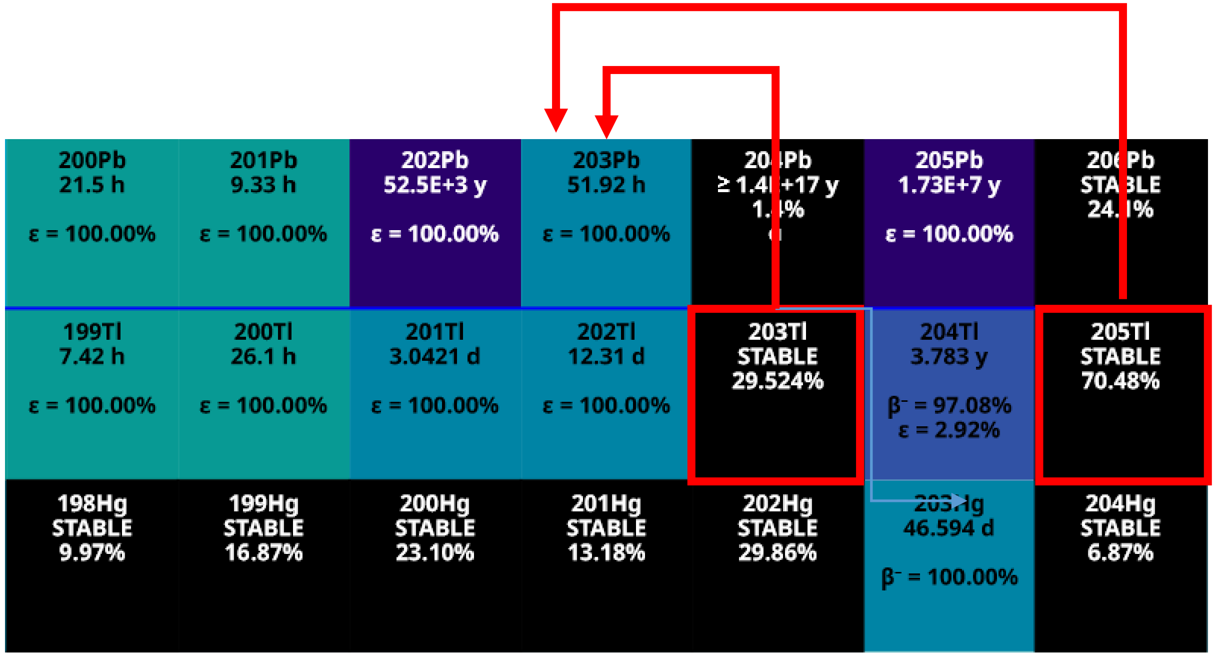
Now Pb-201 can be a problematic but its half-life period is 6 time less half life period of Pb-203, this value is 9h. This radioactive impurity can be largely decreased by leaving it enough time so that it becomes negligible.

Which raw material to produce Pb-203?



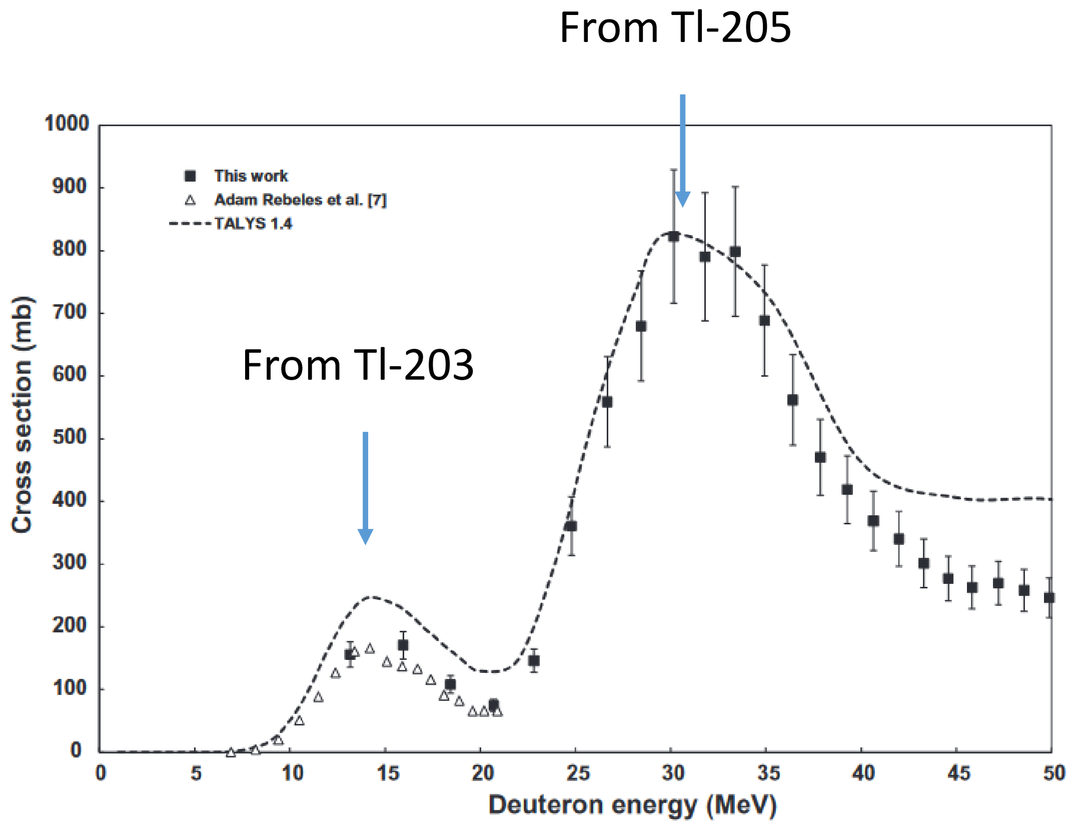
Excitation function for the $^{nat}Tl(d,xn)^{203m2+m1+g}Pb$ reaction.

Nuclear Instruments and Methods in Physics Research B 288 (2012) 94–101



Here is the excitation function of natural thallium to produce Pb-203 with deuteron particles. You can see two peaks at 30 MeV and 15 MeV which correspond to the probability of the production of Pb-203, with the highest production coming from Tl-205 and Tl-203 respectively. The value on mb at 30 MeV is 800 mb and at 15 MeV 250 mb. The natural abundance of Tl-205 is 70% but if we increase the value to 100% for Tl-205 and Tl-203, the production of Pb-203 will always be higher from Tl-205.

Which raw material to produce Pb-203?



Excitation function for the $^{nat}Tl(d,xn)^{203m2+m1+g}Pb$ reaction.

Nuclear Instruments and Methods in Physics Research B 288 (2012) 94–101

Threshold energy of nuclear reaction 203Tl(d,5n)Pb200 = 26,7 MeV

205Tl(d,7n)Pb200 = 40,9 MeV

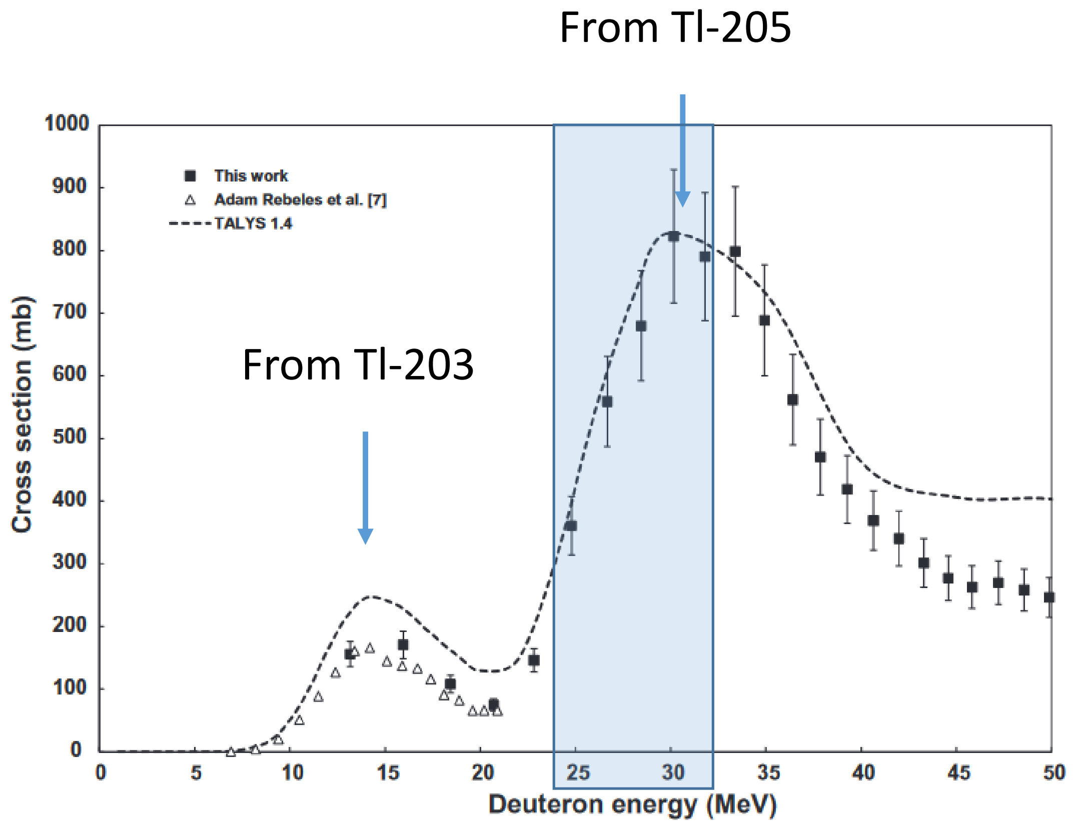
203Tl(d,4n)Pb201 = 19,6 MeV

205Tl(d,6n)Pb201 = 33,8 MeV

@ARRONAX cyclotron, maximal energy of deuteron beam is 35 MeV

The study of threshold energy is mandatory if we want to avoid the production of Pb-200 and Pb-201. From Tl-203, Pb-200 and Pb-201 can be avoided if the energy is below 27 MeV and 20 MeV respectively. And from Tl-205, it is 41 MeV and 34 respectively.

Which raw material to produce Pb-203?



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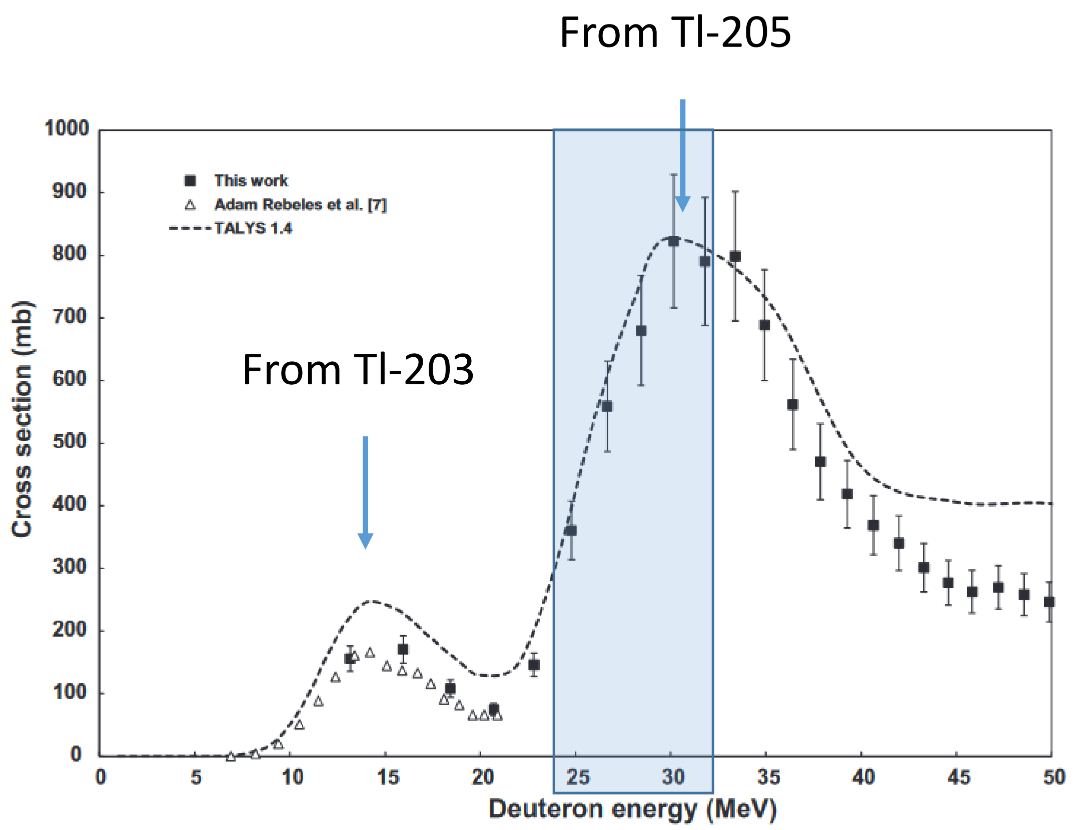
@ARRONAX cyclotron, maximal energy of deuteron beam is 35 MeV

It is clear that if we use Tl-203 there will not be production of Pb-200 and Pb-201 if the irradiation energy is below 19 MeV. However, the production of Pb-203 will be lower than if we use Tl-203. Furthermore, if we use Tl-205, Pb-201 can be avoid if the irradiation energy is below 33 MeV.

Which raw material to produce Pb-203?



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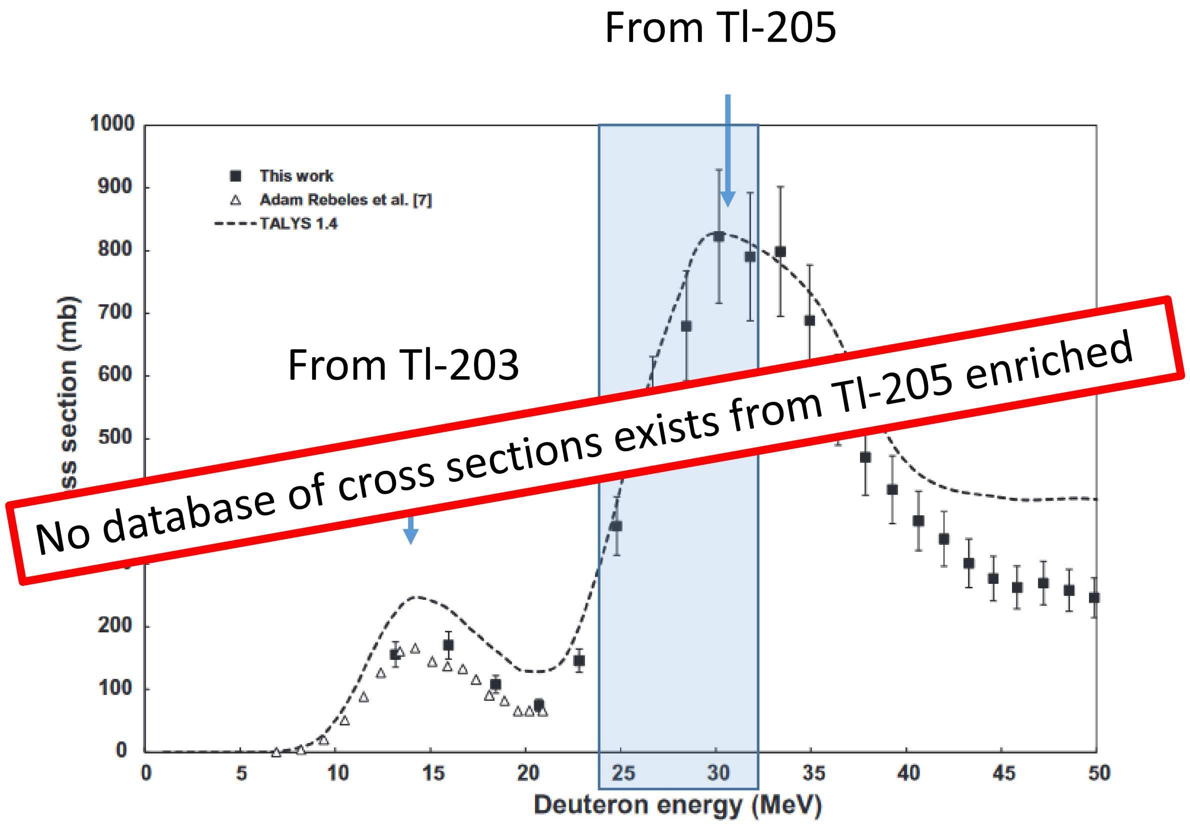
@ARRONAX cyclotron, maximal energy of deuteron beam is 35 MeV

To increase the production of Pb-203, we use enriched Tl-205 until 99.75% from Isolflex USA

If we irradiate with natural Tl below 33 MeV, the production of Pb-200 and Pb-201 will be inevitable. If we want to produce high quantity and purity of Pb-203, the raw material of Tl-205 must be enriched to 205. The enriched raw material of Tl-205 is purchased from Isoflex USA. The enrichment of Tl-205 is 99.75%. 0.25% unfortunately comes from Tl-203.

Which raw material to produce Pb-203?



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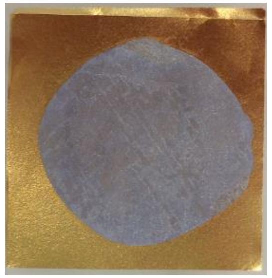
@ARRONAX cyclotron, maximal energy of deuteron beam is 35 MeV

It is necessary to study the excitation functions of all nuclear reactions from Tl-205 with deuteron particles to estimate the production of Pb-203 as well as all impurity productions especially Pb-200 and Pb-201. This study will tell us if the utilization of Tl-205 is a good choice.

Cross sections of all nuclear reactions from Tl-205 with deuteron particles

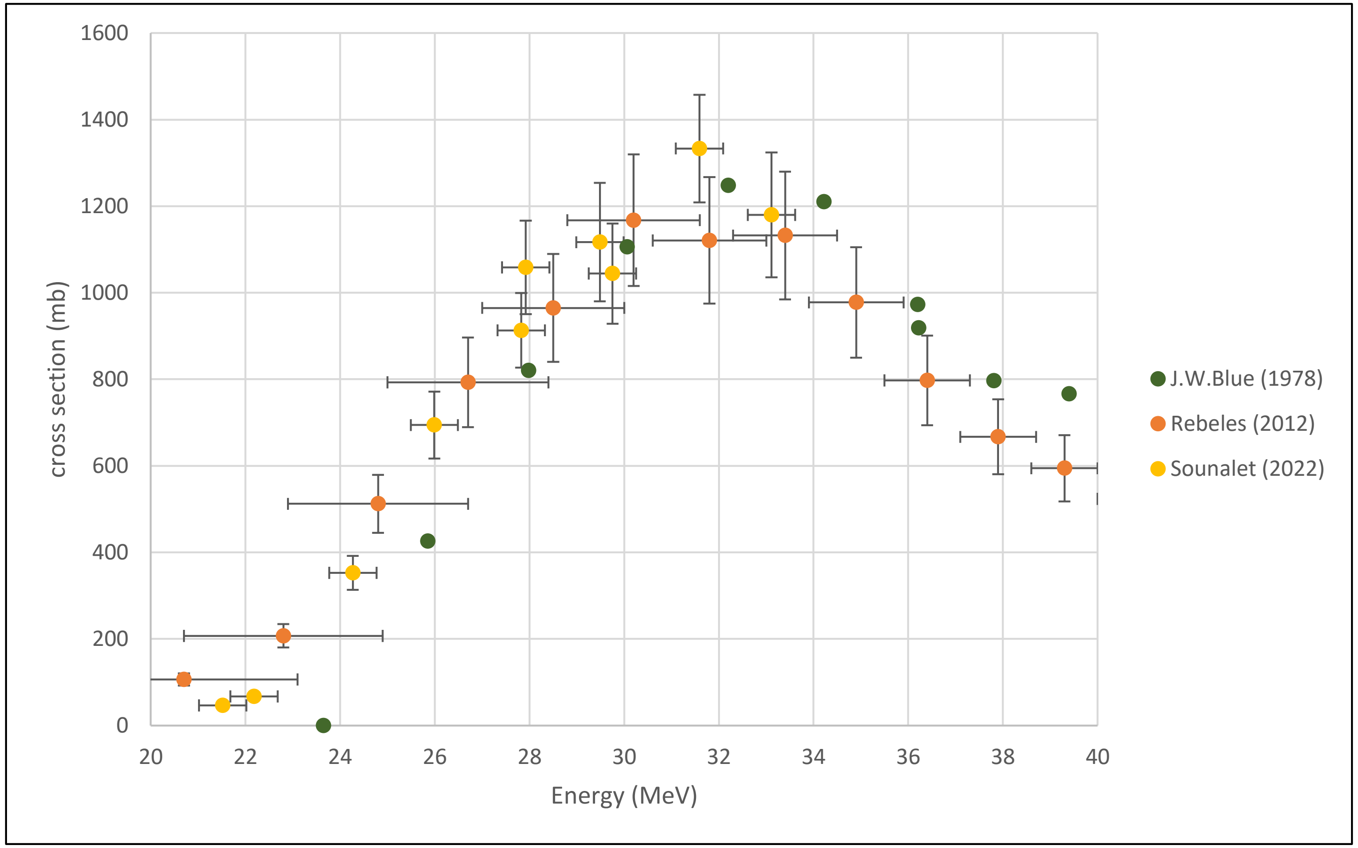


^{205}Tl is manufactured by electrodeposition on gold foil
Thickness of ^{205}Tl : 10 µm < e < 20 µm
Thickness of gold foil: 15 µm

To obtain the excitation functions, we have used the technique of "stack-foils". It consists of irradiating several thin foils in the stack. The foils are the target of Tl-205, monitors and degraders. The target Tl-205 is manufactured by the electrodeposition technique on gold foil. The thickness obtained is between 10 µm and 20 µm. The experiments are carried out at ARRONAX cyclotron.

Cross sections of all nuclear reactions from Tl-205 with deuteron particles

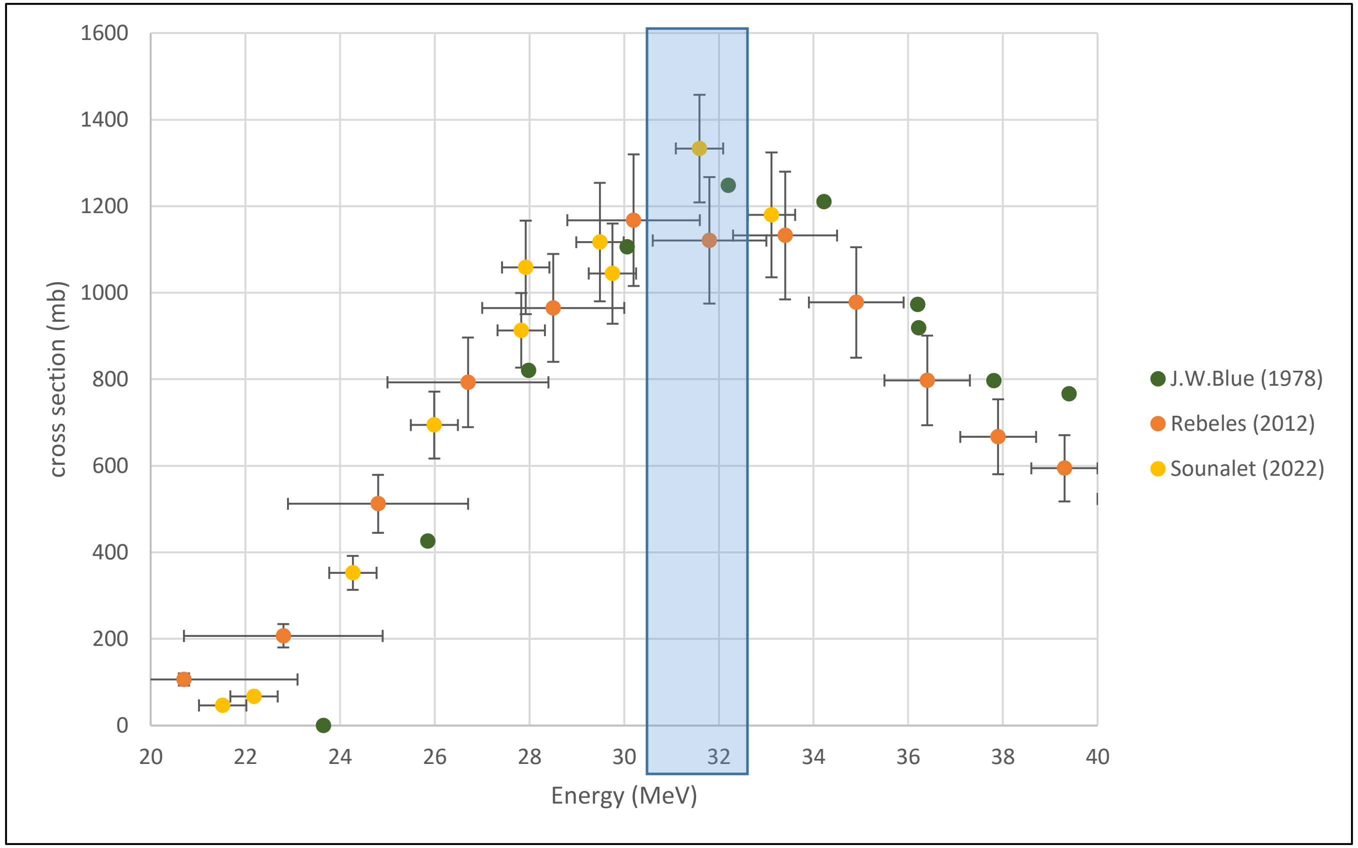
Production of Pb-203



Here we can see the cross section of the nuclear reaction from Tl-205 to produce Pb-203. I have taken into account the isotopic abundance of Tl-205 until 100% to obtain the excitation functions as you can see in the figure above for all authors, including myself. Our points are yellow under the name “Sounalet (2022)” and are consistent with Rebeles (2012). The points of Blue (1978) seems to move the energy to right of 1.5 MeV.

Cross sections of all nuclear reactions from Tl-205 with deuteron particles

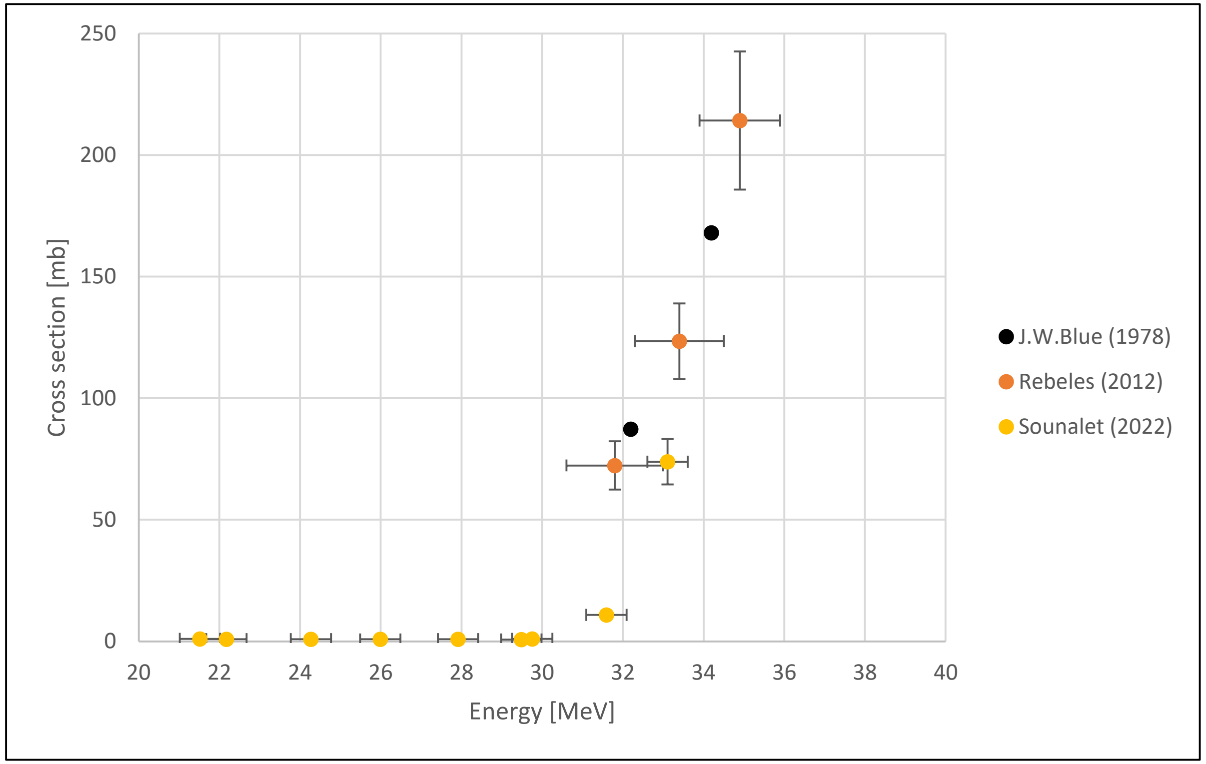
Production of Pb-203



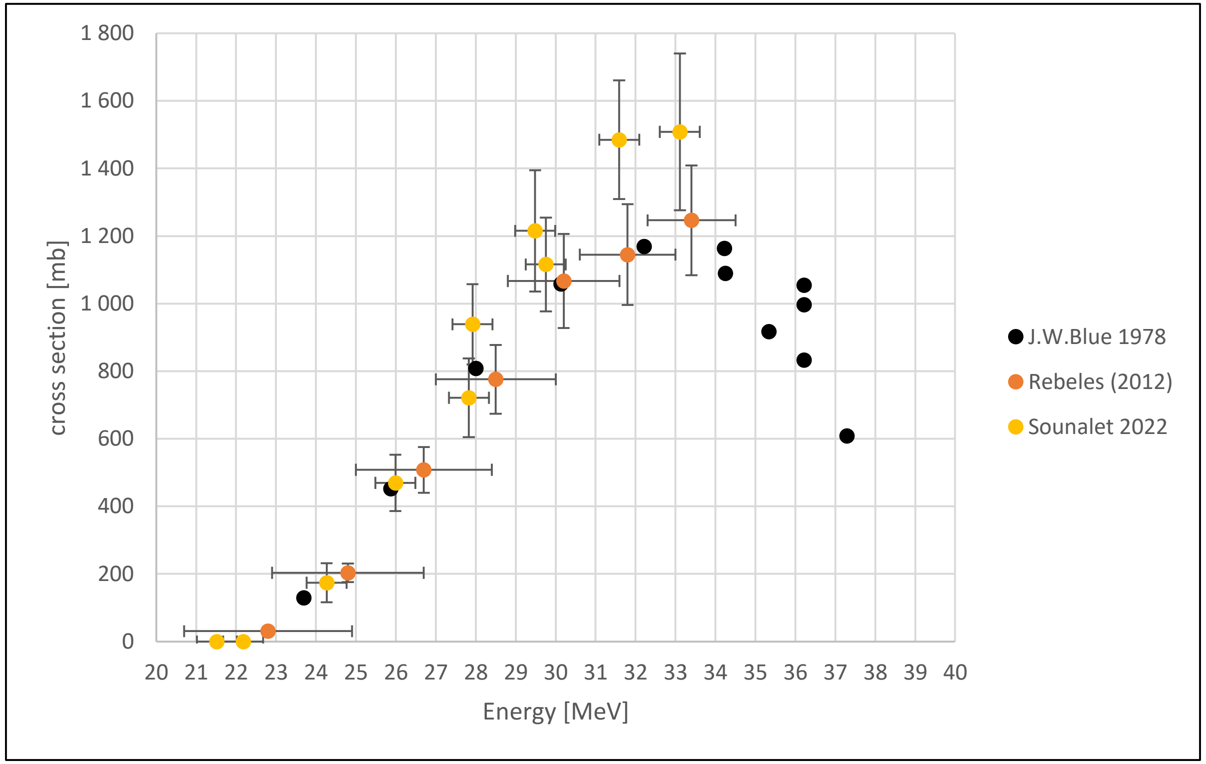
The maximum cross section value is 1333 mb at 32 MeV.

Cross sections of all nuclear reactions from Tl-205 with deuteron particles

Production of Pb-202m and Pb-201



$^{205}Tl(d,5n)^{202m}Pb$

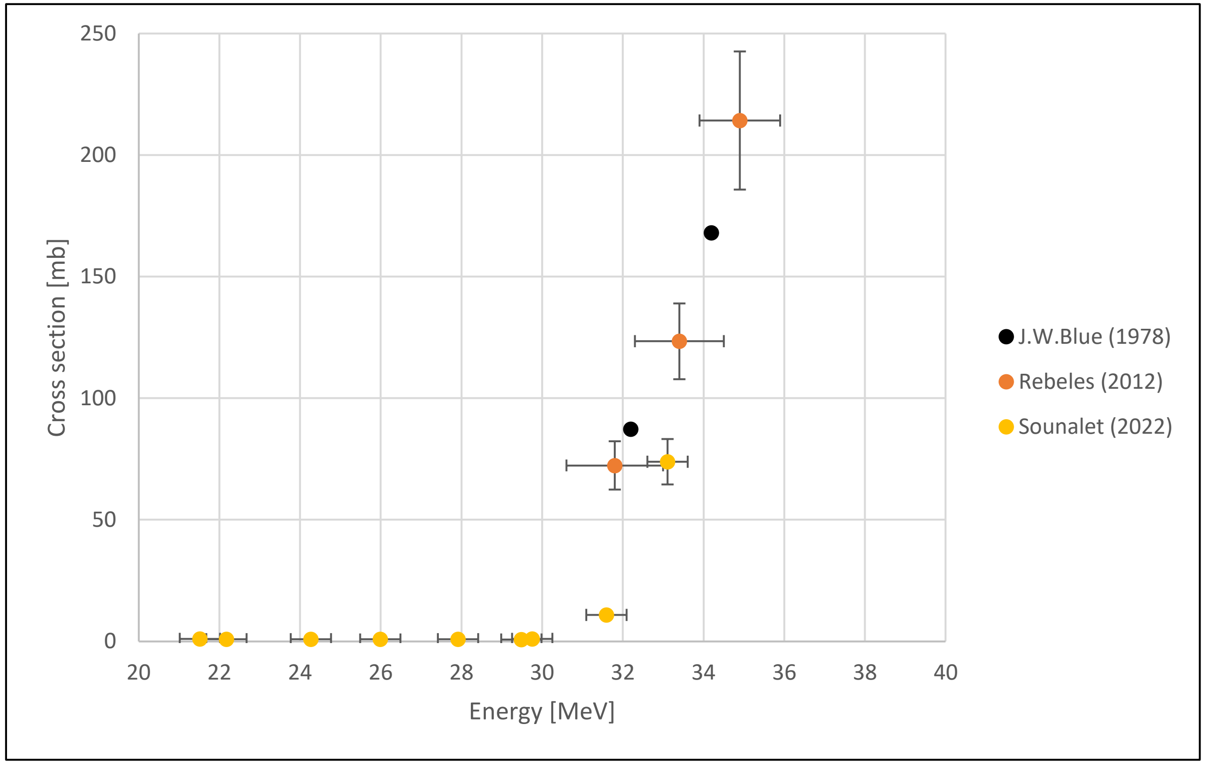


$^{203}Tl(d,4n)^{201}Pb$

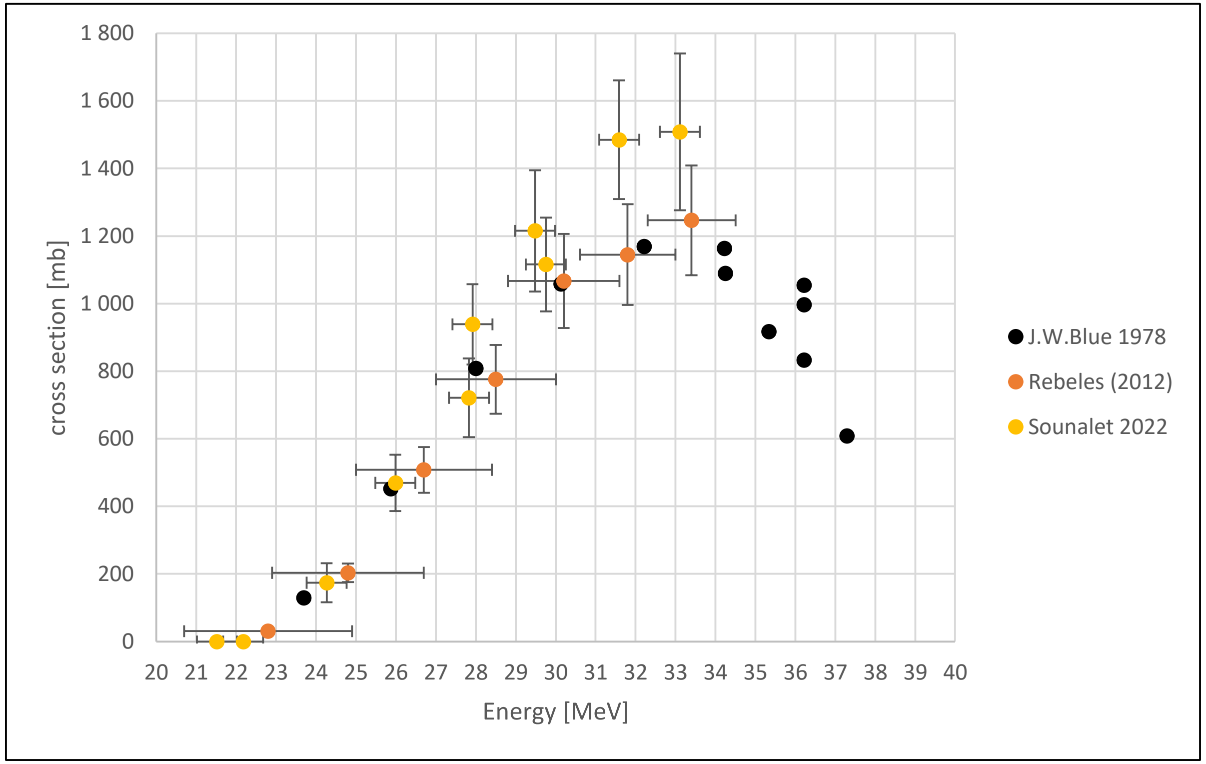
Now we turn our interest to the production of impurities, like, here, Pb-202m and Pb-201. These two radioisotopes were present in the target of Tl-205 after irradiation. The figure on the left is the cross section of the nuclear reaction to produce Pb-202m and that on the right Pb-201.

Cross sections of all nuclear reactions from Tl-205 with deuteron particles

Production of Pb-202m and Pb-201



$^{205}Tl(d,5n)^{202m}Pb$

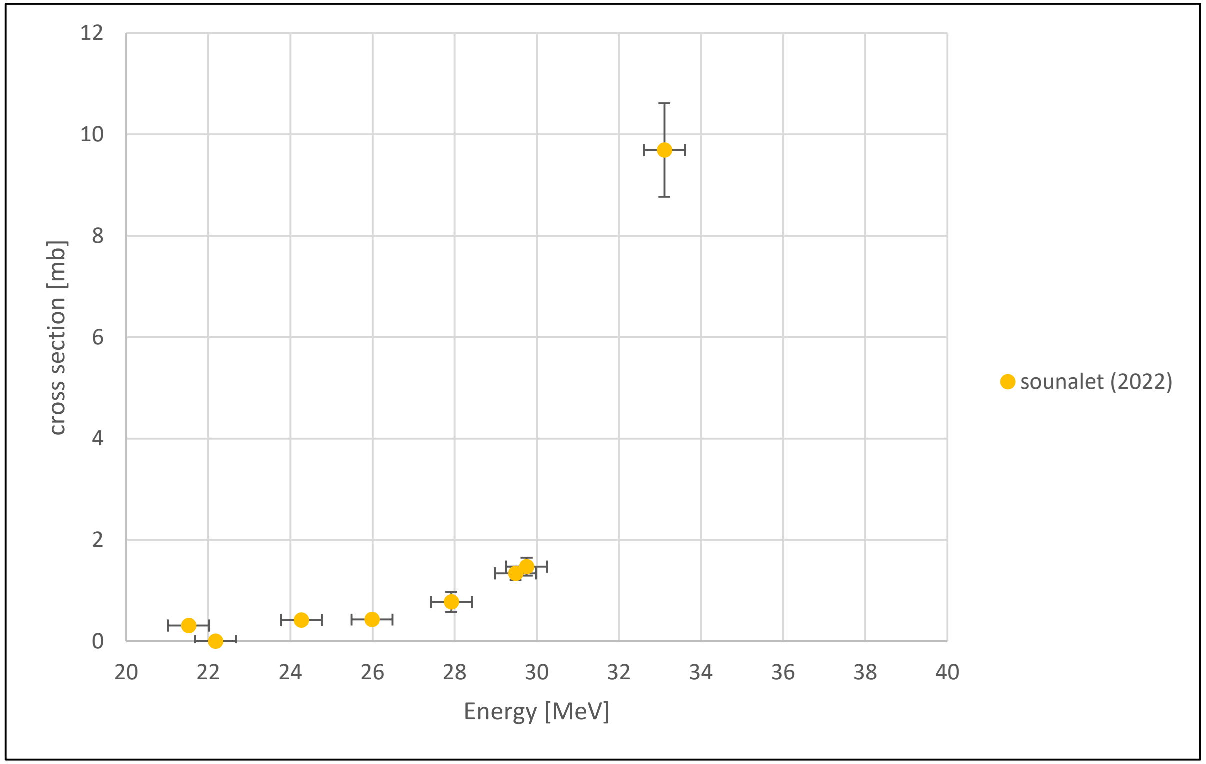


$^{203}Tl(d,4n)^{201}Pb$

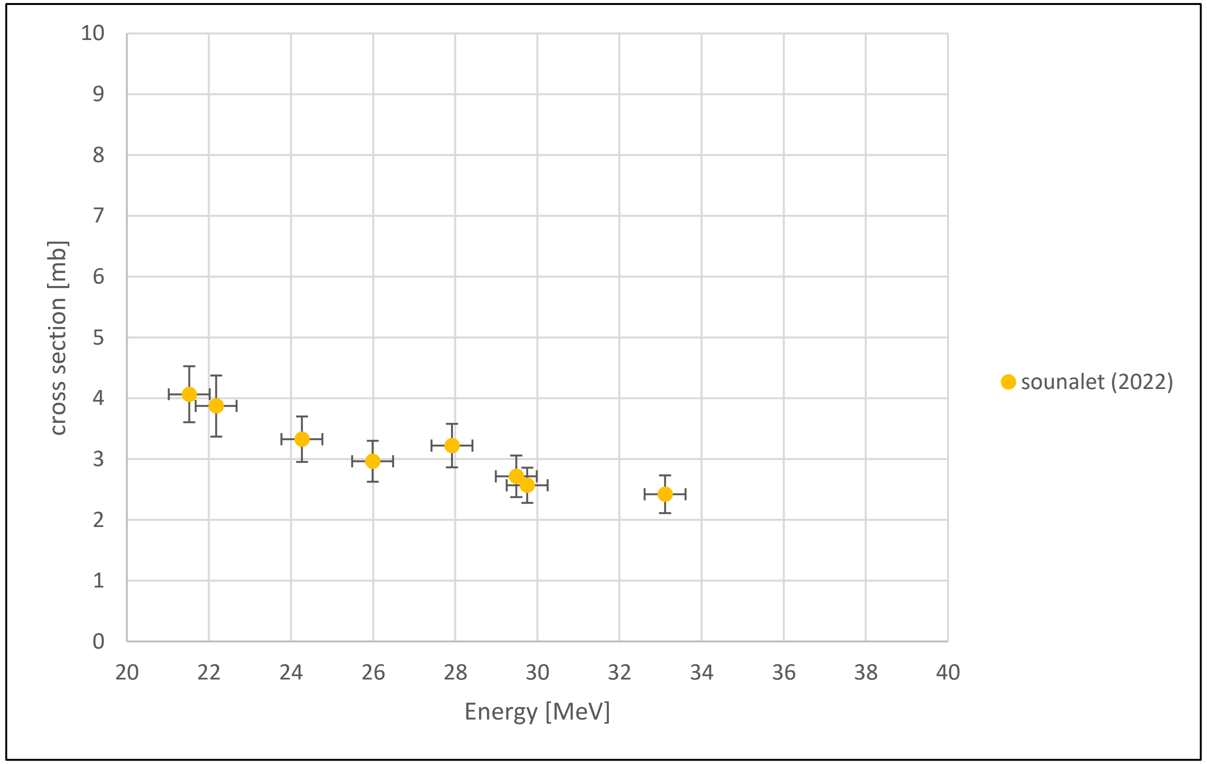
Our results, yellow points, are consistent with Blue (1978) and Rebeles (2012). Unfortunately, even if we have 0.25% of Tl-203, this low percentage doesn't escape the production of Pb-201. However, the radioisotope of Pb-200 wasn't detected with our γ detector. Even if the threshold energy indicated us that the Pb-200 production begins from 27 MeV, its production must be too low to be detected by our γ detector.

Cross sections of all nuclear reactions from Tl-205 with deuteron particles

Production of Tl-202 and Hg-203



$^{205}Tl(d,4n+p)^{202}Tl$



$^{205}Tl(d,2n+2p)^{203}Hg$

Two other radioisotopes have been detected by our γ detector, Tl-202 and Hg-203 for all energy. These two radioisotopes have a long half-life period, 12 days and 47 days. Even if the values of cross sections to produce Tl-202 and Hg-203 are low, it is important to take into account these radioactive impurities which must be avoided in the solution of high purity of Pb-203 for nuclear medecine.

Cross sections of all nuclear reactions from Tl-205 with deuteron particles

^{205}Tl

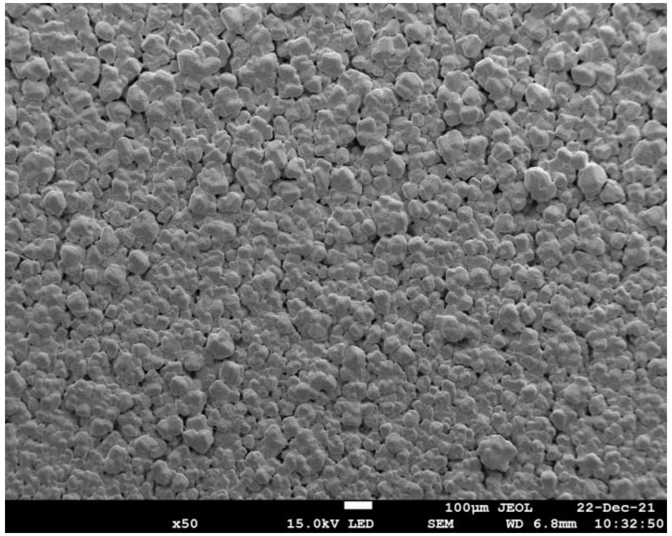
^{203}Tl

^{nat}Tl

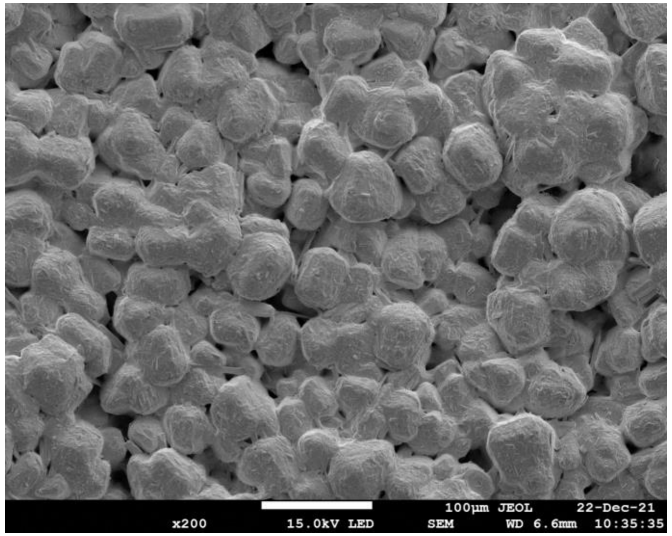
We have seen that the utilization of enriched Tl to 205 is promising if the energy of deuteron is below 33 MeV.

Chemical separation Tl/Pb

Tl deposit for the production



x50

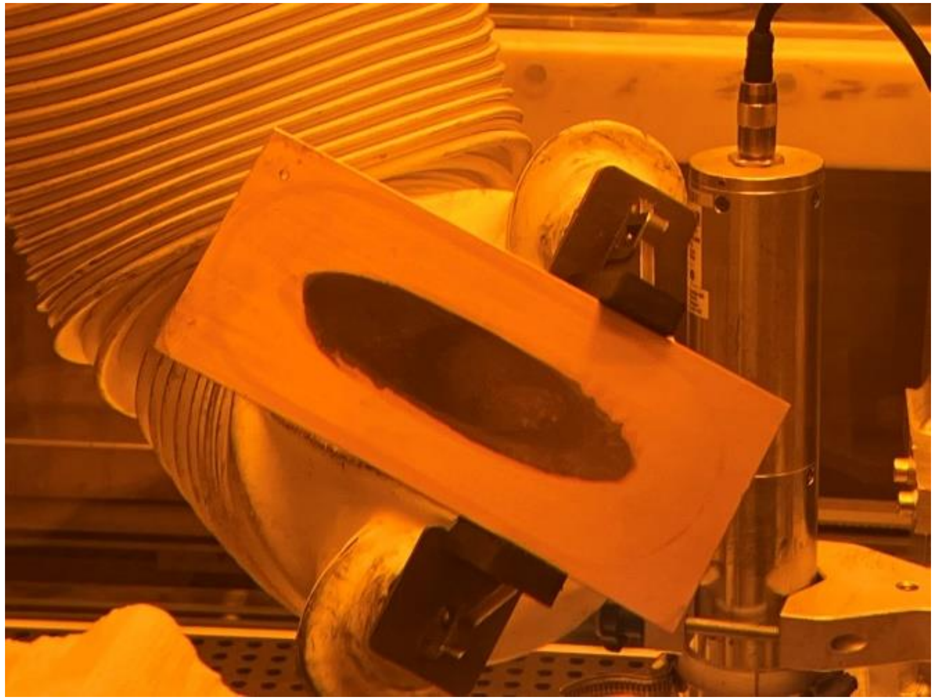


x200

You can see the photo of the deposit of Tl which will be used for the routine production. The shape is elliptical with semi minor axis at 1.1 cm and semi major axis at 4 cm. This deposit is characterized with SEM at magnification x50 and x200, the deposit is formed in reality, the agglomeration of grain and the adhesion on the substrate decrease after 40 µm.

Chemical separation Tl/Pb

Test of irradiated Tl deposit



We have irradiated the deposit of natural Tl at ARRONAX cyclotron. Not with thallium enriched to 205. 2 irradiations have been done with the duration of 10 min and the intensity at 10 µA and 30 µA. The objective of this work is double : one to know if the deposit has resisted under irradiation and one to know if the Pb resin purchased from Triskem is adapted for production. Pb resin is used to separate Tl and Pb. For that, the activities of Pb-203 and Tl-202 are followed like radio-tracers which allow us to determine the rate of recover of Pb and of recycling of Tl.

Chemical separation Tl/Pb

Separation rate of lead

	Separation rate of lead	Separation rate of Thallium
10 μA	78.7% ± 10.2% 2,6% ± 0.8% of ^{202}Tl	97.6% ± 10.5% No Pb impurity
30 μA	80.2% ± 5.7% 0.6% ± 0.1% of ^{202}Tl	96.9% ± 10.4% No Pb impurity

In general, the value of separation rate of lead is 80% with less 10% of uncertainty for both irradiations. However, the impurity of Tl-202 is present in solution, less 3%. Here, we have used natural thallium, if we use thallium enriched to 205, the production of Tl-202 will be low.

Chemical separation Tl/Pb

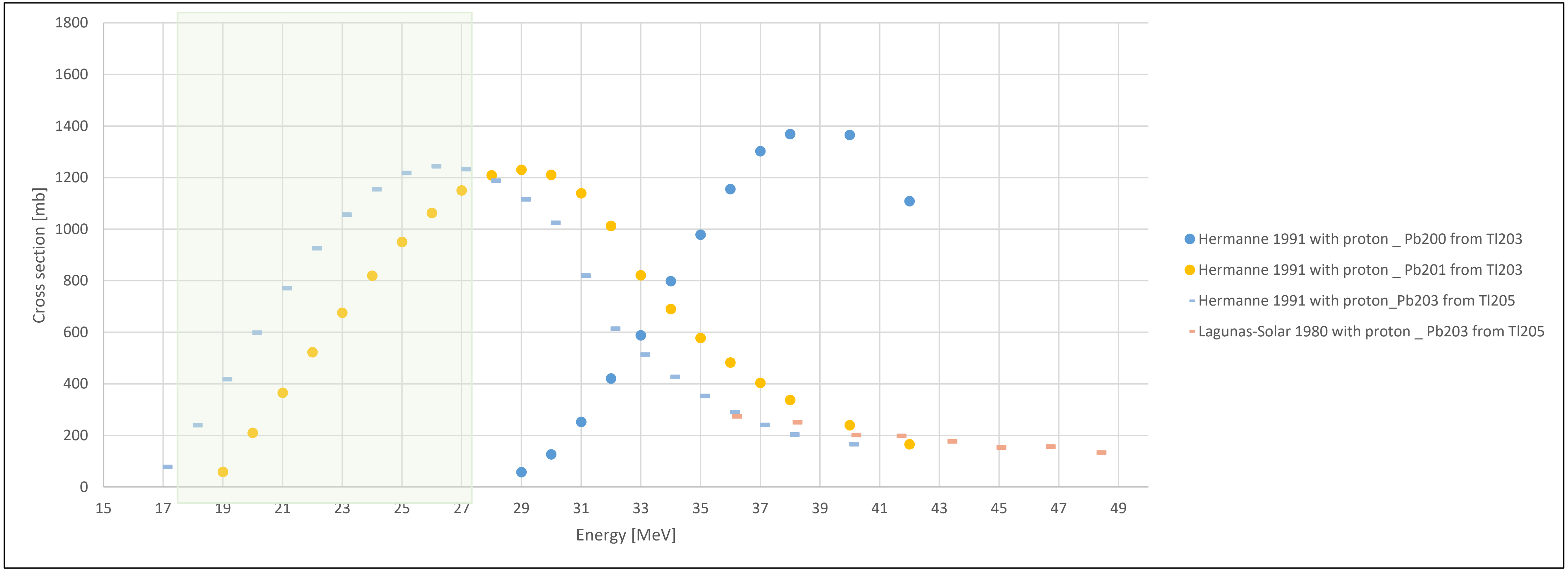
Separation rate of thallium

	Separation rate of lead	Separation rate of Thallium
10 µA	78.7% ± 10.2% 2,6% ± 0.8% of ^{202}Tl	97.6% ± 10.5% No Pb impurity
30 µA	80.2% ± 5.7% 0.6% ± 0.1% of ^{202}Tl	96.9% ± 10.4% No Pb impurity

The value of separation rate of thallium is more than 95% with less 10% of uncertainty for both irradiations. Furthermore, the solution of thallium is highly pure, there is not lead. We can recover near 100% of thallium to recycle and do again the thallium electrodeposition.

Quid with proton irradiation?

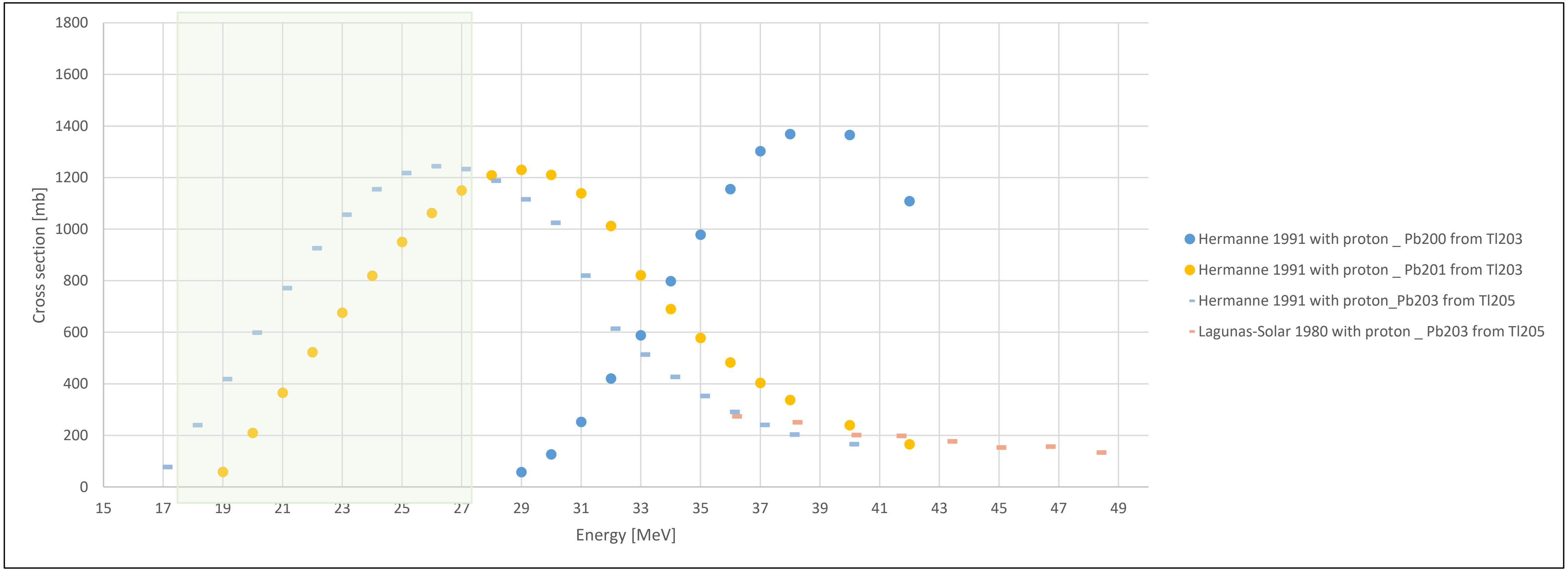
Cross sections extrated in Exford library



Pb-203 can be produced with proton irradiation. 2 authors, Hermannne and Lagunas-Solar have studied the function excitations, but without the values of uncertainty, the curves are extracted from Exford library. The circle point corresponds to the nuclear reactions from Tl-205 enriched to 100% and the dash point corresponds to the nuclear reactions from Tl-203 enriched to 100%.

Quid with proton irradiation?

Cross sections extrated in Exford library



To avoid the production of Pb-200, the proton energy must be below 28 MeV. However, in the region between 17 MeV and 28 MeV, there are two productions, Pb-203 and Pb-201. If the raw material of Tl-205 is enriched to 99.75% like that I have used for our experiments, the cross section values of Pb-201 are low. At 27 MeV, with 100% of Tl-203, the value is 1230 mb but with 0.25%, it will be 3 mb. This value is low and the utilization of proton is also promising for production.

Quid with proton irradiation?

deuteron beam

ARRONAX cyclotron can deliver the energy at 33 MeV

proton beam

ARRONAX cyclotron cannot deliver the energy at 28 MeV
=> The utilisation of degrader is mandotory

But, if we have chosen deuteron, it is because the ARRONAX cyclotron can deliver the energy at 33 MeV, we did not need to use the degrader while with proton, the utilisation of degrader is mandatory to decrease until 28 MeV. Furthermore, the activity of Pb-203 is actually sufficient. Of course, if the demand of Pb-203 will increase with the activity, we move from deuteron to proton.

Estimation of production of ^{203}Pb and ^{201}Pb

With deuteron beam
99,75% ^{205}Tl
0,25% ^{203}Tl

t = 13h
I = 100 uAe
e(Tl) = 110 um

E in = 33 MeV
E out = 26 MeV

→

Act EOB (^{203}Pb) = 1.63 e+5 MBq
Act EOB (^{201}Pb) = 1.63 e+3 MBq

→

After 43h End of Beam, < 0.07% of ^{201}Pb

I have estimated the production of Pb-203 and Pb-201 with deuteron irradiation. For the intensity, duration of irradiation and thickness of Tl, 100 µAe, 13 h and 110 µm, the activities at EOB of Pb-203 and of Pb-201 are 1.63*10^{5} MBq and 1.63*10^{3} MBq respectively. After 43h of EOB, the activity of Pb-201 is negligible, less 0,07%, the limit of acceptable for nuclear medicine use.

Conclusion

- ✓ Cross sections of nuclear reactions of Tl-205 with deuteron particle are done
- ✓ The activity of Pb-203 for production is sufficient with deuteron beam
- ✓ Only Pb-201 will be produced but after some hours, its quantity is below 0.07%, the limit of acceptable for nuclear medicine use.
- ✓ Easily obtained thickness and good coating quality of Tl until 40 µm
- ✓ Chemical separation Tl/Pb is good and simple

- ➢ Pb-201 will always be produced with deuteron and proton beam
- ➢ Thallium is toxic and the handling must be controlled
- ➢ Tl deposit must be protected if the thickness is above 40 µm

- ❖ Cross sections of nuclear reactions of Tl-205 with proton must be realized to compare with two authors Hermanne and Lagunas-Solar

Thank you for your attention

Thank you to :

N.Audouin[1], M.Tarois[1], K.Kamalakannan[2] , F.Haddad[2], G.Arnaud[2], E.Nigron[2], B.Bozovic[3] , R.Dureau[3]

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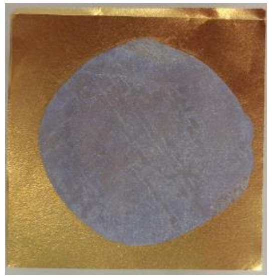
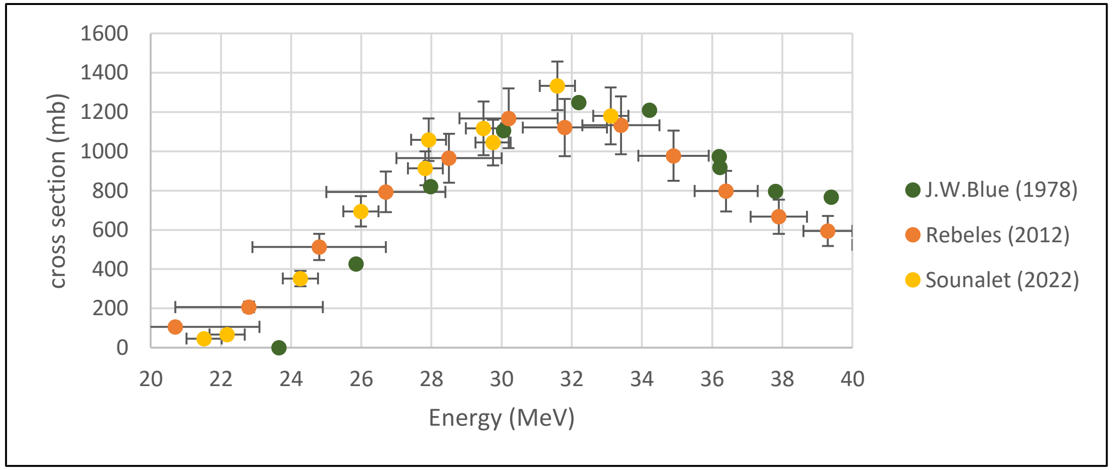
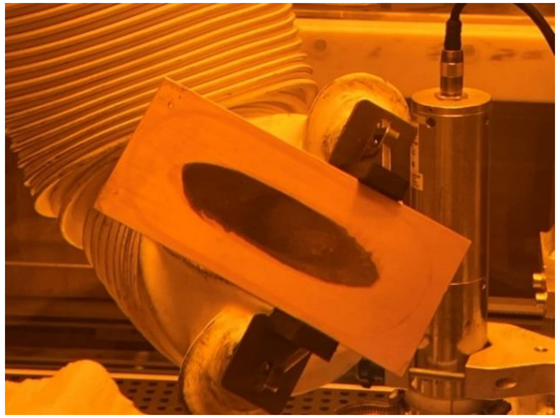
[3] ORANO med







Thank you to the whole team of GIP ARRONAX and Subatech
And INTDS conference at PSI

To answer your questions, I propose that you write them on a piece of paper and discuss them together during the break.