# Investigation of activation cross-sections of proton induced nuclear reactions on $^{nat}Tl$ up to 42 MeV: review, new data and evaluation

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#### Abstract

Cross-sections of proton induced nuclear reactions on natural thallium have been studied for investigation of the production of the medical important  $^{201}Tl$  diagnostic radioisotope. The excitation functions of  $^{204m}Pb$ ,  $^{203}Pb$ ,  $^{202m}Pb$ ,  $^{201}Pb$ ,  $^{200}Pb$ ,  $^{199}Pb$ ,  $^{202}Tl$  (direct, cumulative),  $^{201}Tl$  (direct, cumulative),  $^{200}Tl$ (direct), and  $^{203}Hg$  were measured up to 42 MeV proton energy by stacked foil technique and activation method. The experimental data were compared with the critically analyzed experimental data in the literature, with the IAEA recommended data and with the results of model calculations by using the ALICE-IPPE, EMPIRE-II and TALYS codes.

*Keywords:* Tl targets, proton induced reactions, experimental cross-sections, model calculations, Pb, Tl and Hg radioisotopes, medical radionuclides

#### 1. Introduction

The radionuclide <sup>201</sup>*Tl* ( $T_{1/2} = 73h$ ) as a good potassium analog has been used widely in diagnostic nuclear medicine for forty years (Qaim, 2001). <sup>201</sup>*Tl* decays by EC (100%) and emits a 167 keV  $\gamma$ -ray used for SPECT imaging. The <sup>201</sup>*Tl* can be produced at charged particle accelerators. The investigated nuclear reactions are collected in Table. 1 (Adam-Rebeles et al., 2012; Al-Saleh et al., 2007; Bell and Skarsgard, 1956; Birattari et al., 1982; Blue et al., 1978; Comar and Crouzel, 1975; Dmitriev et al., 1976; Fernandes and da Silva, 1992; Gloris et al., 2001; Hermanne et al., 1980, 1981, 1978; Lebowitz et al., 1975; Nayak et al., 2002; Qaim et al., 1979; Sakai et al., 1965; Zaitseva et al., 1987). The theoretical excitation functions for the most important routes are shown in Fig. 1.

The final result of these studies and of the everyday practice is that the  ${}^{203}Tl(p, 3n){}^{201}Pb \longrightarrow {}^{201}Tl$ production route appears to be the most practical one taking into account the targetry, the production yields, the available accelerators, the radionuclide purity, etc. Presently the  ${}^{203}Tl(p, 3n)$  reaction is used exclusively for production of  ${}^{201}Tl$ . In the next chapters we are



Figure 1: Excitation functions (TENDL 2011) of the most important nuclear reactions for production of  $^{201}Tl$ 

discussing only this production route together with side reactions during bombardment of thallium target with protons. In Table 2 we have collected the most important physical parameters of the experiments related to the measurement of activation cross-section and yield of proton induced nuclear reactions on thallium (Al-Saleh et al., 2007; Blue et al., 1978; Bonardi, 1987; Dmitriev

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| Reaction  | References  | Comment                                   |
|---|---|---|
| $203 \operatorname{Tl}(p,3n)^{201} \operatorname{Pb} \longrightarrow 201 \operatorname{Tl}$ | Sakai (Sakai et al., 1965)                          | Only presently used in practice           |
|   | Lebowitz (Lebowitz et al., 1975)                    |   |
|   | Blue (Blue et al., 1978)                            |   |
|   | Lagunas-Solar (Lagunas-Solar et al., 1978)          |   |
|   | Qaim (Qaim et al., 1979)                            |   |
|   | Bonardi (Birattari et al., 1982)                    |   |
|   | Hermanne (Hermanne et al., 1992)                    |   |
|   | Al-Saleh (Al-Saleh et al., 2007)                    |   |
| $205 \text{ Tl}(p,xn)^{201} \text{ Pb} \rightarrow 201 \text{ Tl}$                          | Lagunas-Solar (Lagunas-Solar et al., 1980)          | Requires high energy beam                 |
| $^{203}$ Tl(d,4n) $^{201}$ Pb $\rightarrow$ $^{201}$ Tl                                     | Adam Rebles (Adam-Rebeles et al., 2012)             | Requires high energy beam                 |
|   | Blue (Blue et al., 1978)                            |   |
| 201 Hg(p,n)201 Tl   | Fernandes (Fernandes and da Silva, 1992)            | Low cross-section                         |
|   | Comar (Comar and Crouzel, 1975)                     |   |
| <sup>202</sup> Hg(p,2n) <sup>201</sup> Tl   | Birattari (Birattari et al., 1982)                  | 202 Tl impurity level                     |
| 200 <sub>Hg(d,n)</sub> 201 <sub>Tl</sub>  | Dmitriev (Dmitriev et al., 1976)                    | Low cross-section                         |
| <sup>201</sup> Hg(d,2n) <sup>201</sup> Tl   | Comar (Comar and Crouzel, 1975)                     | 202 Tl impurity level                     |
| nat Pb(p,x)201 Tl   | Zaitseva (Zaitseva et al., 1987)                    | Requires high energy beam                 |
|   | Lagunas-Solar (Lagunas-Solar et al., 1981)          |   |
|   | Gloris (Gloris et al., 2001; Kuhnhenn et al., 2001) |   |
|   | Bell (Bell and Skarsgard, 1956)                     |   |
| <sup>197</sup> Au( <sup>7</sup> Li,p2n) <sup>201</sup> Tl                                   | Nayak (Nayak et al., 2002)                          | Easy targetry                             |
|   |   | Poor availability of <sup>7</sup> Li beam |
| <sup>197</sup> Au( <sup>6</sup> Li,pn) <sup>201</sup> Tl                                    |   | Easy targetry                             |
|   |   | Poor availability of <sup>6</sup> Li beam |
|   |   |   |

et al., 1976; Hanna et al., 1977; Hermanne et al., 1992; Kernert et al., 1983; Lagunas-Solar et al., 1978; Lebowitz et al., 1975; Malinin et al., 1984; Milazzo-Colli et al., 1975; Oaim et al., 1979; Sakai et al., 1965; Sattari et al., 2003). According to Table 2, a large number of papers was published on  $Tl(p, x)^{201}Pb \longrightarrow^{201} Tl$ production and chemistry, on compilation of the earlier experimental data and on the calculation of crosssection by using different theoretical model codes. In Table 3 we have summarized the most important compilations and theoretical calculations (Canderias-Cruz and Okamoto, 1987; Dmitriev and Zaitseva, 1996; Groppi et al., 2001; Haji-Saeid et al., 2009; Hermanne et al., 2001; Iljinov et al., 1993; Kaplan et al., 2009; Koning and Rochman, 2011; Kurenkov et al., 1995; Lagunas-Solar et al., 1978; Mihailescu et al., 2007; Nowotny, 1981; Rurarz, 1994; Sheu et al., 2003; Shubin, 2001; Shubin et al., 1998; Szelecsényi et al., 1995; Takács et al., 2005; Tel et al., 2011). In spite of numerous earlier works we decided to make new measurements and new evaluations on the activation cross-sections of proton induced reactions on thallium. The aim of the present study and the report was manifold:

 Some part of the recently reported experimental data were measured long time ago (Hermanne et al., 1992). Only preliminary results were reported in equidistant energy scale obtained by fitting the experimental data, without any details on experiments and on the nuclear data used for the data evaluation. This fitted preliminary data were used for preparation of recommended data file of IAEA (Hermanne et al., 2001) for production of  $^{201}Tl$  and used in the upgraded data library (Takács et al., 2005) without correction of the decay data. In this work we decided to publish the original experimental data corrected with upgraded decay data for further use to prepare recommended data (see this work - ser. 1).

- During the last decade new experiments were performed, which could be included into the new evaluation. It is necessary to check the earlier works in respect of new monitor and decay data.
- 3. The theoretical model codes were significantly improved in the last decade and new input parameter library was developed. It was a challenge to see the predictivity of these codes in comparison with the experimental data and with the earlier model results.
- 4. We have performed experiments on deuteron induced reactions on  $^{nat}Tl$  up to 50 MeV (Adam-Rebeles et al., 2011). We thought it is worthwhile to repeat the experiments with proton beam using the same experimental technology (target preparation, spectra measurements, etc.).
- 5. Last but not least in connection with investigation of production possibility of  $^{201}Tl$  in Hungary, the ATOMKI group had direct practical interest in the targetry and the production yields (similar to the case of other routinely produced isotopes ( $^{11}C$ ,  $^{18}F$ ,  $^{67}Ga$ ,  $^{111}In$ ,  $^{123}I$ ). In such a way, to complete our systematical study of excitation functions for production of medical radioisotopes and to make new evaluation we decided to make new measurement relative to the well-established monitor reactions by using more modern experimental and eval-

uation techniques (see this work ser. 2).

#### 2. Experimental

We have performed two series of experiments. The first series was measured 20 years ago and the second was done in the last year.

## 2.1. Experimental and data evaluation details of the first series of measurements:

Samples of natural Tl (50  $\mu m$ ) were prepared by electroplating on brass-foils (50  $\mu m$ ). No additional monitoring foils were inserted into the stacks. The irradiation was done at the VUB CGE 560 cyclotron at beam current of 3-6 µA, for 3-6 minutes. 13 stacks of 7-12 foils were irradiated at incident energies of 43-22 MeV. The number of incident particles was determined from the integrated charge measured in Faraday cup. Tl produced in irradiated samples was quantitatively dissolved after irradiation and aliquots of 1 ml sample were measured using a Ge(Li) detector calibrated with reference sources of  $^{22}Na$ ,  $^{152}Eu$ ,  $^{60}Co$  (2% error on standards). The proton energy degradation along the stack was computed using the stopping formulae coefficients of Andersen and Ziegler for Tl (Andersen and Ziegler, 1977) and the tabulated stopping powers of (Janni, 1966) for the brass backings. The production cross-section was calculated from activity. Quadratic summation of the uncertainties of the contributing parameters results an absolute error of 8.5% (target thickness (2%), detector efficiency (5%), physical decay data (5%), integrated beam current (2%), counting statistics (2%)). Nuclear decay data originally were taken from Erdtmann and Soyka (Erdtmann and Soyka, 1979). For preparation of the IAEA recommended data base (Takács et al., 2005) and for compilation into the EXFOR format the decay data were corrected according to the newest parameters (Browne and Firestone, 1986). In the recent work the cross-section data were corrected according to the new decay data, and the uncertainty of the number of incident particle on the basis of comparisons of the monitor reactions and the Faraday cup data was enlarged from 2 % to 7 %, resulting in 11 % total uncertainty. The results of the earlier data evaluation were corrected for the recent recommended data of the  $\gamma$ -ray intensities. No corrections were done for the half-lives (nonlinearly contributing factor), but there are only small changes in half-lives. In a few cases there are significant changes in  $\gamma$ -ray intensities (see Table 4). There are significant changes in decay data for <sup>201</sup>Pb, <sup>200</sup>Pb used by Lagunas Solar et al., for 401 keV  $\gamma$ -line of <sup>203</sup>Pb used by Al Saleh et al. (Al-Saleh et al., 2007). In the case of the <sup>199</sup>Pb the recent decay data also differ significantly from data used by Hermanne et al. (Hermanne et al., 1992) and by Qaim et al. (Qaim et al., 1979). The correction for the decay data is complicated when the authors used many  $\gamma$ -lines, for which different corrections are required according to the recent data. In this case the correction was done for the strongest  $\gamma$ -line. It should be mentioned that the automatic corrections of the of the cross-section and yield data for the new decay data is not a simple question in the case of many used  $\gamma$ -lines, taking into account the lack of the information in the original publications on the averaging of cross-section data obtained from different  $\gamma$ -lines, measured at different time, sometimes at different sample detector distances.

## 2.2. Experimental and data evaluation details of the second series of measurement:

Samples of natural Tl were prepared also by electroplating. Simultaneous plating of four 15  $\mu m$  thallium target layers (surface area 11.69  $cm^2$ ) on 12.5  $\mu m$  Cu foil were used. The plating method is described in detail in (Afarideh et al., 2004). The electroplated foils for preparation of target stack were cut for 10x10 mm pieces. No additional monitoring foils were inserted into the stacks, the nuclear reactions induced on the backing Cu foils were used as beam monitors. The irradiation was done at VUB CGE 560 cyclotron in a short Faraday cup at beam current 90 nA, for 75 minutes irradiation for time, at 37 MeV incident energy. The radioactivity of the irradiated sample and monitor foils was measured nondestructively using HPGe  $\gamma$ -spectrometry. Counting was started about 3-4 hours after the end of the bombardment (EOB). The proton energy degradation along the stack was computed using the stopping formulae coefficients of Andersen and Ziegler for Tl (Andersen and Ziegler, 1977), and corrected on the basis of a simultaneously measured  $^{nat}Cu(p, x)^{62}Zn$ ,  $^{65}Zn$  monitor reactions (Tárkányi et al., 2001). The uncertainties in the energy scale were estimated taking into account the uncertainty of the energy of the primary beam, the calculated beam straggling and the measures necessary for the energy corrections on the basis of simultaneously measured excitation functions of monitor reactions.

The intensity of the incident beam was obtained from the charge collected in a Faraday cup and corrected using the monitor reactions. The best agreement was found after small corrections on the beam intensity (7% with respect to the Faraday cup results) and on the primary beam energy (0.5 MeV). According to the Fig.



Figure 2: Application of monitor reactions for determination of proton beam energy and intensity

2 the agreement is good in the whole energy range. The production cross-section was calculated from activity. So called "elemental" cross-sections were determined, considering that the Tl targets are monoisotopic. The decay data were taken from NuDat database (NuDat, 2011) the reaction Q-values from (Pritychenko and Sonzogni, 2003). The used decay data are summarized in Table 5. The uncertainties were estimated in the standard way (of-Weights-and Measures, 1993) by combining the individual errors in quadrature. The main sources of error were: beam current (8%), counting statistics and peak area determination (1-7%), detector efficiency (5%), decay data (3%) and effective target thickness (5%)). A total uncertainty of 12-20% is obtained.

#### 3. Theoretical calculations

The measured cross-sections were compared to theoretical effective cross-sections calculated by using the ALICE-IPPE (Dityuk et al., 1998) and EMPIRE-II (Herman et al., 2007)( codes as well as with the data based on the latest version of the TALYS code (Koning et al., 2007), retrieved from the online TENDL 2011 file (Koning and Rochman, 2011). An a priori calculation was done without any knowledge of the experimental results and thus without any parameter adjustment. For each activation product the reaction cross-section on the individual target isotopes was calculated and a weighted summation (according to the abundance of natural occurrence) was made to obtain the total production crosssection.

#### 4. Results

#### 4.1. Excitation functions

The cross-sections for all the reactions studied are shown in Figures 3 - 13 and the numerical values are collected in Tables 6 - 8. The results of the two series of measurements are shown separately. Our results of the two series of measurements are in acceptable good agreement. In most cases the radioisotopes are formed via nuclear processes occurring on <sup>203</sup>Tl and  $^{205}Tl$ . The cross-sections presented here refer to so called effective "elemental" cross-sections, calculated on hypothetic monoisotopic thallium target having number of target nuclei equal to the number of the sum of the numbers of  ${}^{205}Tl$  and  ${}^{207}Tl$  isotopes. We discuss the different reaction products individually.

#### 4.1.1. Activation cross-sections of lead radioisotopes

The radioisotopes of lead are produced via (p,xn) reactions on <sup>203</sup>Tl and <sup>205</sup>Tl. The earlier experimental activation data on Tl are related mainly to the production of radioisotopes of lead, and was measured mostly up to 40 MeV.

### $^{nat}Tl(p, xn)^{204m}Pb$ reaction

This radioisotope has two states: a shorter-lived  $(T_{1/2})$ = 66.93 m,  $I^{\pi} = 9^{-}$ ) decaying by IT to the ground state. The only reaction leading to the formation of  $^{204m}Pb$ from natural thallium is the  ${}^{205}Tl(p, 2n)$  reaction (if we neglect the small contribution from  ${}^{203}Tl(p,\gamma)$ ). The calculations with various codes are compared with the present- and the literature experimental data are shown in Fig. 3. The experimental data show acceptable good agreement with the theory (except Lagunas Solar (Lagunas-Solar et al., 1978) data). Systematic energy shift to the higher energies can be observed for experimental data of Al-Saleh (Al-Saleh et al., 2007). No TENDL calculated data are presented for production of  $^{204m}Tl$ , only for total production cross-section of  $^{204}Tl$ .  $^{nat}Tl(p, xn)^{203}Pb$  reaction

The  $^{203}Pb$  radioisotope is formed by  $^{203}Tl(p,n)^{203}Pb$ and  ${}^{205}Tl(p,3n){}^{203}Pb$  reactions. The contributions of the reactions are well separated in Fig. 4. The available experimental data are in good agreement, except the data of Lagunas-Solar (Lagunas-Solar et al., 1978) and Sakai (Sakai et al., 1965) (see Fig. 4). The theoretical data in TENDL 2011 describe well the experimental results.

#### $^{nat}Tl(p, xn)^{202m}Pb$ reaction

The experimental and theoretical data for production of  $^{202m}Pb$  are shown in Fig. 5. We were able to measure the production cross-section only of the shorterlived metastable state (3.5 h), which decays mostly





Figure 3: Excitation function of  ${}^{nat}Tl(p, x)^{204m}Pb$  reaction



Figure 4: Excitation function of  $^{nat}Tl(p, x)^{203}Pb$  reaction

(90.5%) by internal transition to the longer-lived (5250 a) ground state. In the investigated energy range both the  ${}^{203}Tl(p, 2n)$  and the  ${}^{205}Tl(p, 4n)$  contribute to the formation of  ${}^{202m}Pb$  isomeric state. The comparison of the experimental data shows generally good agreement. At low energies the experimental data of Lagunas Solar (Lagunas-Solar et al., 1978) and (Al-Saleh et al., 2007) shows energy shift towards high energies. There are no theoretical data in the TENDL 2011 for isomeric states, only for the total production of  ${}^{202}Pb$ .

### $^{nat}Tl(p,xn)^{201}Pb$ reaction

The obtained new data are shown in Fig. 6 in comparison with the literature experimental data and with the theory. The structure of the excitation function (similar to previous processes) shows two maxima in the in-

Figure 5: Excitation function of  $^{nat}Tl(p, x)^{202m}Pb$  reaction

vestigated energy range. The origin of the first peak is the  ${}^{2\bar{0}3}Tl(p, 3n)^{2\bar{0}1}Pb$  process. The second peak is due to the (p,5n) process on  $^{205}Tl$ . Our data in maximum is in good agreement with the results of Lagunas Solar (Lagunas-Solar et al., 1978), data of Bonardi measured on enriched target (Bonardi et al., 1982), of Al-Saleh (Al-Saleh et al., 2007) and Qaim (Qaim et al., 1979). It should be mentioned that the data of Qaim (Qaim et al., 1979) are very scattered with approximately 30% difference at the same energies (having 10-15% uncertainties). Our data support the results of higher values (the low and high energy data came probably from beam current measurement of irradiations of different stacks). As it was mentioned at the previous reactions, the Al-Saleh data (Al-Saleh et al., 2007) are shifted in energy near the threshold. The calculated cross-section curve follows the general trend of the experimental data.

#### $^{nat}Tl(p, xn)^{200}Pb$ reaction

Irradiating thallium targets having natural isotopic composition the  ${}^{203}Tl(p, 4n)$  (Q = -24514.2 keV) and the  ${}^{205}Tl(p, 6n)$  (Q = -38716.2 keV) reaction contribute to the production of  ${}^{200}Pb$ . According to Fig. 7 the agreement of experimental data is acceptable good, except the scattered data of Sakai et al (Sakai et al., 1965), and the contradicting data of Lagunas-Solar at high energies reported in (Lagunas-Solar et al., 1978) and his derived data (Lagunas-Solar et al., 1981). The theoretical data in TENDL 2011 describe well the low energy part where reliable experimental data exist.

#### $^{nat}Tl(p, xn)^{199}Pb$ reaction

In the investigated energy range only the  ${}^{203}Tl(p, 5n)$  process contributes to the production of  ${}^{199}Pb$ . Only one earlier data set was found in the literature, reported





Figure 6: Excitation function of  $^{nat}Tl(p, x)^{201}Pb$  reaction



Figure 7: Excitation function of  ${}^{nat}Tl(p, x)^{200}Pb$  reaction

by Qaim et al (Qaim et al., 1979). Our new data show acceptable agreement with these experimental data and with the prediction of TALYS calculation in TENDL 2011 (see Fig. 8).

#### 4.1.2. Activation cross sections of thallium radioisotopes

The radioisotopes of thallium are formed directly via (p,pxn) reaction and through the decay of parent lead radioisotopes.

#### $^{nat}Tl(p,xn)^{202}Tl$ reaction

Parallel with the direct production the radioisotope is produced indirectly via the decay of the  ${}^{202m}Pb$  ( $T_{1/2}$  = 3.54 h, EC(9.5%)) and of the decay of the long-lived  ${}^{202g}Pb$  ( $T_{1/2}$  = 52.5E+3 a, EC(100%)). The cross-

Figure 8: Excitation function of  $^{nat}Tl(p, x)^{199}Pb$  reaction

sections were calculated from spectra measured few days after EOB i.e. after complete decay of  $^{202m}Pb$ , but insignificant decay of  $^{202g}Pb$ . In such a way our cumulative cross-sections contain contributions from the direct decay of  $^{202m}Pb$ . In Fig. 9 we show our cumulative data and corrected data for direct production of  $^{202}Tl$  (when the contribution from the  $^{202m}Pb$  were subtracted). For comparison we also present the experimental data of Qaim (Qaim et al., 1979) corrected for the contribution of  $^{202m}Pb$  decay (direct cross-sections), and the theoretical data for the direct production. The comparison shows good agreement with the earlier data and with theory.



Figure 9: Excitation function of  $^{nat}Tl(p, x)^{202}Tl$  reaction

 $^{nat}Tl(p, xn)^{201}Tl$  reaction

The  ${}^{201}Tl(T_{1/2} = 3.0421 \text{ d})$  radioisotope is formed directly through the  ${}^{203}Tl(p, p2n)$ , and  ${}^{205}Tl(p, p4n)$  reactions and indirectly from the decay of  ${}^{201}Pb(T_{1/2} = 9.33)$ h). In Fig. 10 we present cumulative cross-section of  $^{201}Tl$  after complete decay of  $^{201}Pb$ , and cross-section for direct production when the contribution from the decay of parent was subtracted. Our direct production cross-section data are systematically higher compared to the results of Qaim (Qaim et al., 1979). The uncertainty of direct production is large, taking into account the large contribution of the parent.



Figure 10: Excitation function of  $^{nat}Tl(p, x)^{201}Tl$  reaction

#### $^{nat}Tl(p, xn)^{200}Tl$ reaction

In Fig. 11 we present direct production cross-section for production of  $^{200}Tl$  ( $T_{1/2} = 26.1$  h) after subtraction the contribution of the indirect production through the decay of  ${}^{200}Pb$  ( $T_{1/2} = 21.5$  h). We have only a few points near the threshold and the uncertainties are high due to the high contribution of the parent.



Figure 11: Excitation function of  $^{nat}Tl(p, x)^{201}Tl$  reaction

#### 4.1.3. Activation cross-sections of mercury radioisotopes

The radioisotopes of mercury are formed directly via (p,2pxn) reaction and through the decay of parent lead radioisotopes. We could measure activation crosssection only for production of <sup>203</sup>Pb. In principle the thresholds for production of  $^{199m}Hg$  (42.67 min, IT: 100%), <sup>197m</sup>Hg (23.8 h, IT: 91.4%) and <sup>197g</sup>Hg (64.14 h, EC 100%) are lower than our maximum energy, but due to the low cross-sections (theoretical estimation in Fig. 12), to the weak  $\gamma$ -lines and to the short half-life no reliable data were found.



Figure 12: Activation cross-sections for production of  $^{199m}Hg$ ,  $^{197m}Hg$  and  $^{197g}Hg$  isomeric states predicted in TENDL 2011

 $^{nat}Tl(p, xn)^{203}Hg$  reaction The  $^{203}Hg$  (46.594 d) is produced through the  $^{205}Tl(p, 2pn)$  reaction. It has the same energy  $\gamma$ -line (279 keV) as the  $^{203}Pb$  (51.92 h), therefore it was measured after complete decay of <sup>203</sup>Pb. Only a few experimental data points were measured above the effective threshold (Fig. 13).

#### 4.2. Integral yields

From fits to our experimental excitation functions thick target physical yields (Bonardi, 1987) were calculated (no saturation effects, obtained in an irradiation time that is very short compared to the half-life of the radionuclide). The integral yields for  ${}^{201}Pb$ ,  ${}^{202m}Pb$ ,  ${}^{200}Pb$ ,  ${}^{203}Pb$  and  ${}^{204m}Pb$  (related to production of  ${}^{201}Tl$ and <sup>203</sup>*Pb* ) are shown in Figs. 14-18 in comparison with the directly measured data in the literature. Only very few experimental thick target yield data were found in the literature. The data reported for enriched and natural targets were normalized according to the real isotopic





Figure 13: Excitation function of  ${}^{nat}Tl(p, x)^{203}Hg$  reaction

composition and to the contributing reactions. Our calculated integral yield values for the production of  $^{203}Pb$ and are higher than the direct yield data. In cases of  $^{202m}Pb$  and  $^{201}Pb$  our calculated data are surprisingly lower. No experimental thick target yield data were found for  $^{200}Pb$  in the literature.



Figure 14: Integral yields of <sup>201</sup>*Pb* calculated from the measured excitation functions in comparison with experimental thick target yields from literature

#### 5. Summary and conclusions

The principal aim of this investigation was remeasurement and evaluation of the cross-section data of the production of  $^{201}Tl$ .

Figure 15: Integral yields of  $2^{02m}Pb$  calculated from the measured excitation functions in comparison with experimental thick target yields from literature

- 1. We present new excitation curves for proton induced reactions on <sup>*nat*</sup>Tl targets up to 42 MeV leading to the production of <sup>204m</sup>Pb, <sup>203</sup>Pb, <sup>202m</sup>Pb, <sup>201</sup>Pb, <sup>200</sup>Pb, <sup>199</sup>Pb, <sup>202</sup>Tl (dir, cum), <sup>201</sup>Tl (dir, cum), <sup>200</sup>Tl(dir) and <sup>203</sup>Hg.
- Detailed literature search was done and the literature data were critically analyzed (the earlier errors were corrected, the derived data were changed to original experimental data).
- To upgrade the recommended cross-section data new experimental data were involved (Hanna (Al-Saleh et al., 2007; Hanna et al., 1977), this work).
- 4. To check the predictivity of the theoretical codes, new theoretical calculations were done based on new versions of the model codes.

#### 5.1. The new experimental data:

As our excitation functions are correlated to monitor reactions re-measured over the whole energy range simultaneously the uncertainties in incident energy, average energy in each foil and number of bombarding particles are diminished.

- 5.2. The evaluation process:
- 1. Except a few old works the experimental data shows surprisingly good agreement.
- 2. The description of the theoretical codes is acceptable good.
- 3. The new recommended data from point of view of production of  ${}^{201}Tl$  and the  ${}^{200}Tl$  and  ${}^{202}Tl$  impurities didn't differ significantly from recommended data in the IAEA medical library.



Figure 16: Integral yields of  ${}^{200}Pb$  calculated from the measured excitation functions in comparison with experimental thick target yields from literature

- 4. No proper thick target yield data were measured for validation of the excitation functions (low beam intensity measurements at around 30 MeV).
- There are good review works discussing the optimization of the <sup>201</sup>*Tl* production and the chemical processes (Afarideh et al., 2004; Al-Saleh et al., 2007; Birattari et al., 1982; Fernandes and da Silva, 1992; Groppi et al., 2001; Haji-Saeid et al., 2009; Hanna et al., 1977; Lagunas-Solar et al., 1981, 1978; Malinin et al., 1984; Qaim et al., 1979; Sattari et al., 2003; Sheu et al., 2003)(.
- Except excitation functions of Lagunas Solar (Lagunas-Solar et al., 1978) measured up to 60 MeV for production of <sup>200–204</sup>*Pb*, no experimental data exists above 40 MeV
- 5.3. Application of the new experimental and evaluated data:
  - 1. Optimization of the production of the medically used  ${}^{201}Tl$ .
  - The measured excitation functions can be used for determination of thallium in different materials by proton activation analysis (De Brucker et al., 1989; Wauters et al., 1987).
  - 3. To estimate the radiation dose caused by proton activation of the materials with thallium content (glasses, semiconductors, particle detectors, high temperature superconductors, etc.)
  - 4. Application of  ${}^{203}Pb$  emitting imaginable  $\gamma$ -ray (279 keV) for bio-distribution and targeting studies of the cytotoxic alpha-emitting  ${}^{212}Pb$ -radiolabeled



Figure 17: Integral yields of <sup>203</sup>*Pb* calculated from the measured excitation functions in comparison with experimental thick target yields from literature



Figure 18: Integral yields of  $2^{04m}Pb$  calculated from the measured excitation functions in comparison with experimental thick target yields from literature

compounds and antibodies (Chappell et al., 2000; Garmestania et al., 2005) . Preparation of  $^{203}Pb$  compounds for studies on pathway and effects of lead pollution (Girardi et al., 1975).

5. To upgrade the nuclear reaction model codes and their input parameters.

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| Author                               | Target                          | Irradiation   | Beam current                     | Measurement            | Nuclear reaction,  | Covered energy |
|--------------------------------------|---------------------------------|---------------|----------------------------------|------------------------|--|----------------|
|                                      |                                 |               | measurement                      | of activity            | measured quantity,   | range          |
|                                      |                                 |               | and                              | and                    | number of measured data points   | (MeV)          |
|                                      | natro                           |               | monitor reaction                 | separation method      | natmy 200 pt   | 10.5.51.6      |
| Sakai,                               |                                 | cyclotron     | Faraday cup                      | chemical separation    | $nat TI(p,x)^{200}$ Pb, s, 6   | 10.5-51.6      |
| (Sakai et al., 1903)                 |                                 | stacked foll  |                                  | 7-1Na(1)               | $nat_{T(p,x)} = rb, s, b$  | 10.4-51.7      |
| Labouitz                             | nat <sub>T1</sub>               | avalotron     | Foreday aup                      | chamical constation    | 203 <sub>Tl</sub> (p,x) 10, 8, 0   | 10.28.22.15    |
| (Lebowitz,<br>(Lebowitz et al. 1975) | ~0.2 mg/cm <sup>2</sup>         | stacked foil  | Faraday cup                      | v-Ge(Li)               | 1(p,5ii) 10, s, 0  | 19.20-32.13    |
| Milazzo-Colli                        | 203 TI 205 TI                   | cyclotron     |                                  | a-Si surface barrier   | 203 Tl(n a)200 Hg s 1  | 20             |
| (Milazzo-Colli et al. 1975)          | 300-400 ug/cm <sup>2</sup>      | cyclouoli     |                                  | a bi sanace barrier    | $205_{Tl(p,a)}^{202}Hg s 1$  | 20             |
| Dmitriev                             | nat TI                          | cyclotron     | Faraday cun                      | no chemical separation | 203 Tl(p,p) 202 Tl TTV 1   | 20             |
| (Dmitriev et al. 1976)               | thick target                    | single foil   | T araday cup                     | v-Ge (Li)              | 203 <sub>Tl</sub> (p, 3p) <sup>201</sup> g ph TTY 1  | 22             |
| (,, _, _, )                          |                                 |               |                                  | ,                      | $203 \text{Tl}(p,2n)^{202m}$ Pb, TTY, 1  | 22             |
|                                      |                                 |               |                                  |                        | $nat_{Tl(p,x)}^{203g}$ Pb, TTY, 1  | 22             |
| Hanna,                               | nat Tl                          | cyclotron     | Faraday cup                      | chemical separation    | nat Tl(p,x) <sup>200</sup> Pb,TTD, REL, 4  | 26.89-34.84    |
| (Hanna et al., 1977)                 | 275 mg/cm <sup>2</sup>          | EN tandem VdG |                                  | γ-Ge (Li)              | nat Tl(p,x) <sup>201</sup> Pb,TTD, REL, 6  | 20.4-34.87     |
|                                      |                                 | stacked foil  |                                  |                        | nat Tl(p,x)202 Tl, TTD, REL, 7   | 16.27-34.84    |
| J. W. Blue,                          | nat Tl foil                     | cyclotron     | Faraday cup                      | no chemical separation | <sup>203</sup> Tl(p,3n) <sup>201</sup> Pb, s, 12   | 18.47-35.19    |
| (Blue et al., 1978)                  | 110µm                           | stacked foil  |                                  | γ-Ge(Li)               | <sup>203</sup> Tl(P,4N) <sup>200</sup> Pb, s, 6  | 27.91-35.1     |
| Lagunas-Solar                        | nat Tl                          | cyclotron     | Faraday cup                      | no chemical separation | <sup>nat</sup> Tl(p,x) <sup>200</sup> Pb, PY, FCT, 31  | 28.4-59.3      |
| (Lagunas-Solar et al., 1978)         | 0.16-0.20 g/cm <sup>2</sup>     | stacked foil  |                                  | γ-Ge(Li)               | <sup>203</sup> Tl(p,x) <sup>200</sup> Pb, s, DERIV, 14   | 30.0-40.9      |
|                                      |                                 |               |                                  |                        | narTl(p,x) <sup>201</sup> Pb, PY, FCT, 68  | 17.2-59.0      |
|                                      |                                 |               |                                  |                        | <sup>205</sup> Tl(p,x) <sup>201</sup> gPb, M+, s, DERIV, 39  | 18.3-33.6      |
|                                      |                                 |               |                                  |                        | $203m_{10} + 202m_{11} + 1179, DT, 41$   | 11.8-35.9      |
|                                      |                                 |               |                                  |                        | $p_{atri} = 203 \text{ m} \text{ ps}, \text{ s, DERIV, 41}$  | 17.0-27.4      |
|                                      |                                 |               |                                  |                        | $nat_{T(r,r)}^{203}$ Pb, r 1, PC1, 45  | 17.0-59.1      |
|                                      |                                 |               |                                  |                        | $nat_{Tl(p,x)}^{204m}$ pb py FCT 30  | 13.6-35.8      |
|                                      |                                 |               |                                  |                        | $nat_{Tl(p,x)}^{204m}$ Pb. s. DERIV. 30  | 15.3-35.       |
|                                      |                                 |               |                                  |                        | 205 <sub>Tl</sub> (p,x) <sup>200</sup> Pb, s, DERIV, 11  | 46.8-59.3      |
|                                      |                                 |               |                                  |                        | 205 Tl(p,x) <sup>201g</sup> Pb, M+, s. DERIV. 23   | 38.2-59.0      |
|                                      |                                 |               |                                  |                        | <sup>205</sup> Tl(p,x) <sup>202m</sup> Pb, s, DERIV, 23  | 27.4-35.9      |
| Qaim et al.                          | nat Tl                          | cyclotron     | 63Cu(p,2n)62Zn                   | no chemical separation | 205 <sub>Tl(p,2n)</sub> 204m <sub>Pb, s, 27</sub>  | 12.1-44.2      |
| (Qaim et al., 1979)                  | 50-125µm                        | stacked foil  | 63Cu(p,p2n)61Cu                  | γ-Ge(Li)               | nat Tl(p,x) <sup>203</sup> Pb, IND, s, 34  | 7.49-44.4      |
|                                      |                                 |               |                                  |                        | nat Tl(p,x) <sup>202m</sup> Pb, IND, s, 31   | 9.59-44.2      |
|                                      |                                 |               |                                  |                        | <sup>nat</sup> Tl(p,x) <sup>201</sup> Pb, IND, s, 25   | 17.2-44.1      |
|                                      |                                 |               |                                  |                        | <sup>nat</sup> Tl(p,x) <sup>200</sup> Pb, IND, s, 14   | 28.2-44.4      |
|                                      |                                 |               |                                  |                        | <sup>nat</sup> Tl(p,x) <sup>199</sup> Pb, IND, s, 5  | 38.6-44.3      |
|                                      |                                 |               |                                  |                        | nat Tl(p,x) = 202 Tl, IND, 18  | 16.2-44.2      |
|                                      |                                 |               |                                  |                        | nat Tl(p,x) <sup>201</sup> Tl, IND, s, 11  | 19.8-42.8      |
| Bonardi                              | nat Tl foil                     | cyclotron     | Faraday cup                      | γ-Ge(Li)               | $hat_{Tl}(p,x)^{200}$ Pb, TTD, 7   | 27.696-42.24   |
| (Bonardi et al., 1982)               |                                 | stacked foil  |                                  |                        | nat T1(p,x) <sup>201</sup> Pb, TTD, TM, 12   | 18.823-42.067  |
|                                      |                                 |               |                                  |                        | $nat TI(p,x)^{202}TI, TID, TTY, TM, 13$  | 16.677-42.143  |
|                                      |                                 |               |                                  |                        | 203m(, )200pi mmp (  | 7.5794-42.177  |
|                                      |                                 |               |                                  |                        | 203 T(p,x) <sup>201</sup> pb, TTD, 11  | 28.234-30.021  |
|                                      |                                 |               |                                  |                        | $203 T(p,x)^{202m}$ pb, TTD, 14  | 20.755-55.984  |
|                                      |                                 |               |                                  |                        | $203 T (p x)^{203} Pb TTD 14$  | 14.001-35.983  |
|                                      |                                 |               |                                  |                        | (1), (1), (1), (1), (1), (1), (1), (1),  | 11001 00.000   |
| Kernert (Kernert et al., 1983)       | <sup>203</sup> Tl (99%)         | cyclotron     | Faraday cup                      |                        | <sup>203</sup> Tl(p,x) <sup>201</sup> Pb, TTY, 1   | 29             |
|                                      | nat Tl                          | single target |                                  |                        | <sup>203</sup> Tl(p,x) <sup>202m</sup> Pb, TTY, 1  | 29             |
|                                      | thick target                    |               |                                  |                        | <sup>203</sup> Tl(p,x) <sup>203</sup> Pb, TTY, 1   | 29             |
|                                      |                                 |               |                                  |                        | <sup>203</sup> Tl(p,x) <sup>204</sup> <sup>m</sup> Pb, TTY, 1  | 29             |
|                                      |                                 |               |                                  |                        | $hat_{Tl}(p,x)^{201}$ Pb, TTY, 1   | 29             |
|                                      |                                 |               |                                  |                        | $nat_{Tl}(p,x)^{202m}Pb, TTY, 1$   | 29             |
|                                      |                                 |               |                                  |                        | $nat_{Tl}(\mathbf{p}, \mathbf{x})^{205}$ Pb, TTY, 1<br>$nat_{Tl}(\mathbf{p}, \mathbf{x})^{204}m_{Tl}$ mat_r  | 29             |
|                                      | 203 m (oc ogt)                  |               |                                  |                        | 203mi 201mi mili 1   | 29             |
| Malinin<br>(Malinin et al. 1084)     | 0.25.0.4 a/am2                  | cyclotron     | Faraday cup                      | chemical separation    | 2005 TI(p,x)201 Pb, TTY, I   | 30.5           |
| Hermanne                             | nat T1 (500 m)                  | cyclotron     | Faraday cup                      | no chemical separation | 203 <sub>Tl(n 2n)</sub> 202mph s DERIV 12  | 15-27          |
| (Hermanne et al. 1992)               | 11 (J0µ III)                    | stacked foil  | r araday cup                     | v-Ge(Li)               | 203 Tl(n, 3n)201 Ph. s. DERIV 22   | 19-42          |
| (1001111111 Ct al., 1772)            |                                 | Succeed for   |                                  | / 56(24)               | 203 TI(P.4N) 200 Pb. s. DERIV 12   | 29-42          |
|                                      |                                 |               |                                  |                        | 205 <sub>Tl(p,2n)</sub> 204m <sub>Pb</sub> , s, DERIV. 29  | 12-42          |
|                                      |                                 |               |                                  |                        | <sup>203</sup> Tl(p,3n) <sup>203</sup> Pb, s, DERIV, 24  | 17-40          |
|                                      |                                 |               |                                  |                        | <sup>203</sup> Tl(p,4n) <sup>202m</sup> Pb, s, DERIV, 12   | 29-40          |
|                                      |                                 |               |                                  |                        | <sup>203</sup> Tl(p,2n) <sup>201</sup> Pb, s, DERIV, 4   | 38-42          |
| Sattari                              | 203 Tl(enr.)                    | cyclotron     | Faraday cup                      | γ-HpGe(Li)             | <sup>203</sup> Tl(p,x) <sup>201</sup> Pb, TTY, REL, 4  | 28-30          |
| (Sattari et al., 2003)               | 18.3 µ m                        | single target |                                  |                        | nates. 201 - 202 201   |                |
| Al-Saleh                             | 10.15 1.1.1.1.                  | cyclotron     | Faraday cup                      | no chemical separation | $r_{10} Tl(p,x)^{201} Pb, s = {}^{203} Tl(p,3n)^{201} Pb, s, 17$   | 19.3-27.2      |
| (AI-Saleh et al., 2007)              | $10-15\mu$ m thick on $10\mu$ m | stacked foil  | Cu(p,x) <sup>05</sup> Zn monitor | g-HpGe                 | $11(p,x)^{-0.2m}$ Pb, s = $205$ Tl $(p,2n)^{2.02m}$ Pb, s + $205$ Tr $(-4.5202m$ Pb, s = $202$   | 15.7-27.2      |
|                                      | Cu                              |               |                                  |                        | $p_{0,1} = p_{0,1} = p_{0$ | ( ) )7 )       |
|                                      |                                 |               |                                  |                        | $II(p,x) = PD, s = 205 II(p,n) = 205 Pb, s + 205 II(-2\pi) = 203 Db = 2.4$   | 0.2-27.2       |
|                                      |                                 |               |                                  |                        | natru 204m pi 205mi 2 204m pi  | 11 2 27 2      |
|                                      |                                 |               |                                  |                        | $II(p,x)^{20}$ III Pb, s = $205$ $II(p,2n)^{20}$ III Pb, s, 28   | 11.5-27.2      |
| this work Ser 1                      | nat Tl (50µ m)                  | cvclotron     | Faraday cup                      | no chemical separation | nat Tl(p,x) <sup>204m</sup> Pb, 105  | 12.2-41.7      |
|                                      | on                              | stacked foil  |                                  | γ-Ge(Li)               | nat Tl(p,x) <sup>203</sup> Pb. 73  | 10.8-41.7      |
|                                      | Cu (50µ m)                      |               |                                  |                        | nat <sub>Tl(p,x)</sub> 202mpb, N110  |                |
|                                      |                                 |               |                                  |                        | nat Tl(p,x) <sup>201</sup> Pb, 81  |                |
|                                      |                                 |               |                                  |                        | nat Tl(p,x) <sup>200</sup> Pb, 67  | 10.8-41.7      |
|                                      |                                 |               |                                  |                        | nat <sub>Tl(p,x)</sub> 199Pb, 17   | 17.1-41.7      |
| this work Ser. 2                     | nat Tl (20.8 μ m)               | cyclotron     | Faraday cup                      | no chemical separation | nat <sub>Tl(p,x)</sub> 204m <sub>Pb</sub> , 20   | 27.1-41.5      |
|                                      | on                              |               | natCu(p,x)62,65Zn                | γ-HpGe                 | nat Tl(p,x) <sup>203</sup> Pb, 20  |                |
|                                      | Cu (12.5 µ m)                   |               | 10                               |                        | nat Tl(p,x) <sup>202m</sup> Pb, 20   |                |
|                                      |                                 |               | 10                               |                        | <sup>nat</sup> Tl(p,x) <sup>201</sup> Pb, 20   |                |
|                                      |                                 |               |                                  |                        | nat T1(p,x) <sup>200</sup> Pb, 20  |                |
|                                      |                                 |               |                                  |                        | nat TI(p,x)=02 TI, 20<br>nat mic 201 mi 20   |                |
|                                      |                                 |               |                                  |                        | nat TI(p,x)=01 TI, 20<br>nat TI(p,x)=01 TI, 20   |                |
|                                      |                                 |               |                                  |                        | II(p,x)=0011, 20<br>nat Ti(p,x)=0011, 20   |                |
| 1                                    | 1                               | 1             | 1                                | 1                      | ····· 11(p,x)=05 Hg, 20  | 1              |

| Table 3: Th | e most important compilations and theoretical calculations on cross-sections and yields o | of $^{nat}$ Tl(p,x) <sup>201</sup> Pb? <sup>20</sup> | <sup>1</sup> Tl and the side reactions |
|-------------|---|--|--|
|             |   |  |  |

| Author                                     | Model code            | Reactions                                 | Compilation | Recommended |
|--|-----------------------|---|-------------|-------------|
| Canedrias Ruz (Canderias-Cruz and          |                       | nat Tl(p,x) <sup>201</sup> Pb             | X           |             |
| Okamoto, 1987)                             |                       |   |             |             |
| Rurarz (Rurarz, 1994)                      |                       | nat Tl(p,x) <sup>201</sup> Pb             | X           |             |
| Kurenkov (Kurenkov et al., 1995)           | ALICE-87, STAPRE      | 203,205Tl(p,xn)                           |             |             |
| IAEA TECDOC (Hermanne et al., 2001)        | ALICE-IPPE, ALICE-HMS | nat Tl(p,x) <sup>201</sup> Pb             | X           | X           |
|  |                       | nat Tl(p,x) <sup>202</sup> Pb             |             |             |
|  |                       | nat Tl(p,x) <sup>200</sup> Pb             |             |             |
| Takcs (Takcs et al., 2005)                 |                       | nat Tl(p,x) <sup>201</sup> Pb             | X           | X           |
|  |                       | nat Tl(p,x) <sup>202</sup> Pb             |             |             |
|  |                       | nat Tl(p,x) <sup>200</sup> Pb             |             |             |
| Tel (Tel et al., 2011)                     | ALICE/ASH             | nat Tl(p,x) <sup>201</sup> Pb             |             |             |
| Haji Said (Haji-Saeid et al., 2009)        |                       | nat Tl(p,x) <sup>201</sup> Pb             | X           |             |
| Sheu (Sheu et al., 2003)                   | ALICE-01, Fluka       | nat Tl(p,x) <sup>201</sup> Pb             | X           |             |
| Landolt Bornstein (Iljinov et al., 1993)   |                       | nat Tl(p,x) <sup>201</sup> Pb             | X           |             |
|  |                       | nat Tl(p,x) <sup>202</sup> Pb             |             |             |
|  |                       | nat Tl(p,x) <sup>200</sup> Pb             |             |             |
| Mihilescu (Mihilescu et al., 2007)         |                       |   |             |             |
| Lagunas-Solar (Lagunas-Solar et al., 1978) | STAPRE                |   |             |             |
| Groppi (Groppi et al., 2001)               |                       | nat Tl(p,x) <sup>201</sup> Pb             | X           |             |
| Shubin (Shubin, 2001)                      |                       |   |             |             |
| Nowotny (Nowotny, 1981)                    |                       | nat                                       |             |             |
| MENDL -2P (Shubin et al., 1998)            | ALICE-IPPE            | nat Tl(p,x) <sup>201</sup> Pb             |             |             |
|  |                       | nat Tl(p,x) <sup>202</sup> Pb             |             |             |
|  |                       | nat Tl(p,x) <sup>200</sup> Pb             |             |             |
| Koning (Koning and Rochman, 2011)          | TALYS                 | nat Tl(p,x) <sup>201</sup> Pb             |             |             |
|  |                       | nat Tl(p,x)202 Pb                         |             |             |
|  |                       | nau Tl(p,x) <sup>200</sup> Pb             |             |             |
| Szelecsényi (Szelecsényi et al., 1995)     |                       |   | V           |             |
| Dmitriev (Dmitriev and Zaitseva, 1996)     | CEM                   | 203mi 203mi                               | A           |             |
| (Kaplan et al. 2009)                       | GDH model             | 203m(, 2)202m                             |             |             |
| (Rapian et al., 2005)                      | Hybrid model          | 203 m 2 201 m                             |             |             |
|  | Equilibrium model     | 203 m (p,3n) 201 pb                       |             |             |
|  |                       | 205 m(c, 2, )203 pt                       |             |             |
|  |                       | 205 TI(p,3n)205 Pb                        |             |             |
|  |                       | 205 TI(p,4n)202 Pb                        |             |             |
|  |                       | 205 TI(p,5n)201 Pb<br>205 200             |             |             |
|  |                       | <sup>203</sup> Tl(p,6n) <sup>200</sup> Pb |             |             |

| NuclideJ <sup>p</sup>                  | Half-life | E <sub>γ</sub> (keV)<br>NUDAT  | I <sub>γ</sub> (%)  | I <sub>γ</sub> (%)<br>Leg Sol | $I_{\gamma}$ (%) Qaim | I <sub>γ</sub> (%) Al Saleh     | I <sub>γ</sub> (%) Bonardi    | I <sub>γ</sub> (%) Dmitriev | I <sub>γ</sub> (%)   |
|--|-----------|--|---|-------------------------------|-----------------------|---------------------------------|-------------------------------|-----------------------------|----------------------|
| (MeV)                                  |           | NUDAI  | NUDAI   | Lag-301                       |                       |                                 |                               |                             | Hermanne 1991        |
| 204m <sub>Pb</sub><br>9-<br>2185.88    | 66.93 m   | 374.76<br>899.15<br>911.74   | 94.20<br>99.174<br>91.5                                       | 99.2                          | 95<br>100<br>100      | 89<br>99                        |                               |                             | 94.2<br>99.3         |
| <sup>203</sup> Pb<br>5/2-              | 51.92 h   | 279.1952<br>401.320  | 80.9<br>3.35  | 81.0                          | 80.7                  | 99<br>90.7                      | 80.8<br>3.35                  | 81<br>3.8                   | 80<br>3.8            |
| 202 <i>m</i> pb<br>9-<br>2169.83       | 3.54 h    | 422.12<br>657.49<br>786.99<br>960.70<br>389.94<br>459.72<br>490.47                     | 84<br>31.7<br>49<br>89.9<br>6.6<br>9.2<br>9.8                 | 85.3                          | 90<br>56<br>89        | 96<br>50                        | 85.66<br>32.4<br>49.8<br>91.3 |                             | 86<br>35<br>50<br>89 |
| 201 <sub>Pb</sub><br>5/2- <sup>+</sup> | 9.33 h    | 331.15<br>361.25<br>405.96<br>584.60<br>692.41<br>767.26<br>826.26<br>907.67<br>945.96 | 77<br>9.5<br>2.03<br>3.6<br>4.3<br>3.28<br>2.38<br>6.1<br>7.2 | 61.4                          | 82<br>10.7            | 79<br>9.9                       | 3.56<br>4.27<br>5.70<br>7.36  |                             | 82<br>10.7           |
| 200 <sub>Pb</sub><br>0+                | 21.5 h    | 147.63<br>235.62<br>257.19<br>268.36<br>450.56   | 38.2<br>4.35<br>4.52<br>4.01<br>3.37                          | 28.4                          | 30.8                  |                                 | 37.73<br>4.3<br>4.46<br>3.96  |                             | 29                   |
| 199 <sub>Pb</sub><br>3/2-              | 90 m      | 366.90<br>720.24<br>1135.04  | 44 %<br>6.5<br>7.8  |                               | 11.6<br>14            |                                 |                               |                             | 79<br>11.4           |
| 202 <sub>T1</sub><br>2-                | 12.31 d   | 439.510  | 91.5  |                               | 95                    | 91.4                            |                               |                             |                      |
| 201 <sub>Tl</sub><br>1/2+              | 3.0421 d  | 135.34<br>167.43   | 2.565<br>10.00  |                               | 2.65                  | 2.565                           |                               |                             |                      |
| 200 <sub>Tl</sub><br>2-                | 26.1 h    | 367.942<br>579.300<br>828.27<br>1205.75  | 87<br>13.7<br>10.8<br>30                                      |                               |                       | 87.2<br>13.78<br>10.81<br>29.91 |                               |                             |                      |
| <sup>203</sup> Hg<br>5/2-              | 46.594 d  | 279.1952   | 81.56   |                               |                       |                                 |                               |                             |                      |

#### Table 4: Comparison of the $\gamma$ -ray intensities used by different authors

| Nuclide           | Half-life | Decay mode (%)   | Εγ       | $I_{\gamma}(\%)$ | Contributing            | Q-value (keV) |
|-------------------|-----------|------------------|----------|------------------|-------------------------|---------------|
| $\mathbf{J}^p$    |           |                  | (keV)    | ,                | reactions               |               |
| level energy      |           |                  |          |                  |                         |               |
| (MeV)             |           |                  |          |                  |                         |               |
| 204m Pb           | 66.93 m   | IT(100)          | 374.76   | 94.20            | 203 Tl(p,g)             | 6637.51       |
| 9-                |           |                  | 899.15   | 99.174           | 205 TI(p 2p)            | -7564.517     |
| 2185.88           |           |                  | 911.74   | 91.5             | 11(p,211)               |               |
| 203 Ph            | 51.92 h   | EC(100)          | 279 1952 | 80.9             | 203 Tl(n n)             | -1756.97      |
| 5/2=              | 51.521    | 20(100)          | 401 320  | 3 35             | 205 TI(n 2n)            | -15959.0      |
| 202m              |           |                  | 101.520  | 5.55             | 203                     | 1575710       |
| 202mPb            | 3.54 h    | FT(90.5)         | 422.12   | 84               | 205-T1(p,2n)            | -8681.26      |
| 9-                |           | EC(9.5)          | 657.49   | 31.7             | <sup>205</sup> Tl(p,4n) | -22883.29     |
| 2169.83           |           |                  | 786.99   | 49               |                         |               |
|                   |           |                  | 960.70   | 89.9             |                         |               |
|                   |           |                  | 389.94   | 6.6              |                         |               |
|                   |           |                  | 459.72   | 9.2              |                         |               |
| 201               |           |                  | 490.47   | 9.8              | 202                     |               |
| 201 Pb            | 9.33 h    | EC(100)          | 331.15   | 77               | 205 Tl(p,3n)            | -17428.3      |
| 5/2-*             |           |                  | 361.25   | 9.5              | <sup>205</sup> Tl(p,5n) | -31630.3      |
|                   |           |                  | 405.96   | 2.03             |                         |               |
|                   |           |                  | 584.60   | 3.6              |                         |               |
|                   |           |                  | 692.41   | 4.3              |                         |               |
|                   |           |                  | 767.26   | 3.28             |                         |               |
|                   |           |                  | 826.26   | 2.38             |                         |               |
|                   |           |                  | 907.67   | 6.1              |                         |               |
| 200               |           |                  | 945.96   | 7.2              | 202                     |               |
| 200 Pb            | 21.5 h    | EC(100)          | 147.63   | 38.2             | <sup>205</sup> Tl(p,4n) | 24514.2       |
| 0+                |           |                  | 235.62   | 4.35             | <sup>205</sup> Tl(p,6n) | -38716.2      |
|                   |           |                  | 257.19   | 4.52             |                         |               |
|                   |           |                  | 268.36   | 4.01             |                         |               |
|                   |           |                  | 450.56   | 3.37             |                         |               |
| <sup>199</sup> Pb | 90 m      | EC(100)          | 366.90   | 44 %             | <sup>203</sup> Tl(p,5n) | -33600.8      |
| 3/2-              |           |                  | 720.24   | 6.5              | <sup>205</sup> Tl(p,7n) | -47802.9      |
|                   |           |                  | 1135.04  | 7.8              |                         |               |
| 202 <sub>Tl</sub> | 12.31 d   | EC(100)          | 439.510  | 91.5             | <sup>203</sup> Tl(p,pn) | -7849.2       |
| 2-                |           |                  |          |                  | 205 Tl(p,p3n)           | -22051.3      |
|                   |           |                  |          |                  | $202m_{\rm Pb}$ decay   |               |
| 201 m             | 2.0421.1  | EC(100)          | 125.24   | 25/5             | 203 TI( 2)              | 14721.9       |
| 1/2               | 5.0421 d  | EC(100)          | 155.54   | 2.303            | 205                     | -14/21.8      |
| 1/2+              |           |                  | 107.45   | 10.00            | 20.3 Tl(p,p4n)          | -28923.8      |
|                   |           |                  |          |                  | <sup>201</sup> Pb decay |               |
| 200 <sub>Tl</sub> | 26.1 h    | EC(100)          | 367.942  | 87               | 203 Tl(p,p3n)           | -22927.04     |
| 2-                |           |                  | 579.300  | 13.7             | 205 TI(p p5p)           | -37129.08     |
|                   |           |                  | 828.27   | 10.8             | r (p,pon)               |               |
|                   |           |                  | 1205.75  | 30               |                         |               |
| 203 Hg            | 46.594 d  | $\beta^{-}(100)$ | 279.1952 | 81.56            | 205 Tl(p.2pn)           | -13911.76     |
| 5/2-              |           | P ()             |          |                  | ···,_,,,                |               |

Table 5: Nuclear data of the investigated reactions for  $^{\it nat}Tl+~p$ 

Abundances (%):  ${}^{203}Tl$  (29.524),  ${}^{205}Tl$  (70.476) Increase Q-values if compound particles are emitted: np-d, +2.2 MeV; 2np-t, +8.48 MeV; n2p- ${}^{3}$ He, +7.72 MeV; 2n2p- $\alpha$ , +28.30 MeV Decrease Q-values for isomeric states with level energy of the isomer

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| 26.1 1.4 20.3 2.2 10.5 1.16 316.1 34.77  |       |
|  |       |
| 26.5 1.4 22.1 2.4 10.0 1.10 345.2 37.97  |       |
| 26.6 1.4 21.3 2.3 964.0 106.0 11.4 1.25 366.5 40.32<br>26.7 965.1 106.0 11.4 1.25 366.5 40.32  |       |
| $\begin{array}{c ccccccccccccccccccccccccccccccccccc$  |       |
| 27.2 1.3 19.0 2.1 889.3 97.8 8.9 0.98 417.3 45.90 5.3 0.01   |       |
| 27.6 1.3 20.4 2.2 882.2 97.0 10.4 1.15 375.9 41.34 1.9 0.21  |       |
| 27.6 1.3 18.0 2.0 972.9 107.0 12.4 1.36 406.3 44.70 2.2 0.25   |       |
| $\begin{array}{c ccccccccccccccccccccccccccccccccccc$  |       |
| 27.7 1.5 $77.6$ $77.6$ $77.0$ $72.0$ $100.5$ $12.8$ $1.41$ $400.4$ $44.53$ $2.6$ $0.50$ $2.2$ $0.24$   |       |
| 28.2 1.2 16.3 1.8 11.3 1.24 5.5 0.60   |       |
| 28.7 1.2 16.7 1.8 931.7 102.5 17.8 1.96 8.5 0.94   |       |
| 28.7 1.2 16.1 1.8 849.0 93.4 20.3 2.24 9.2 1.02 9.2 1.02 9.2 1.02  |       |
| 28.7 1.2 15.5 1.7 17.5 192 441.7 48.59 9.0 0.99 28.9 12 859.7 94.6 21.6 238 402.9 44.32 15.2 1.67  |       |
| 29.0 1.2 21.6 2.37 403.2 44.35 11.7 1.28   |       |
| 29.2 1.2 13.7 1.5 21.2 2.33 411.6 45.27 11.0 1.21  |       |
| 29.3 1.2 15.3 1.7 23.5 2.59 14.1 1.55  |       |
| $\begin{array}{c ccccccccccccccccccccccccccccccccccc$  |       |
| 29.8 1.1 10.5 1.2 792.3 87.2 41.9 4.61 406.0 44.66 30.8 3.38   |       |
| 29.9 1.1 17.6 1.9 741.1 81.5 44.4 4.88 425.7 46.83 31.7 3.48   |       |
| 30.0 1.1 737.4 81.1 419.9 46.19 25.2 2.77  |       |
| $ \begin{array}{c ccccccccccccccccccccccccccccccccccc$   |       |
| 30.3 1.1 1.3.8 1.5 48.0 5.28 1.00 37.0 4.06  |       |
| 30.6 1.1 14.9 1.6 70.2 7.72 379.4 41.73 62.7 6.90  |       |
| 30.7 1.1 18.0 2.0 684.7 75.3 74.1 8.15 65.4 7.19   200 1 1 16.0 2.0 684.7 75.3 74.1 8.15 65.4 7.19   |       |
| 30.8 1.1 13.0 1.7 1.9 7.91 404.8 44.33 00.4 7.30<br>30.0 11 167 1.8 750 41.25 3750 41.25 771 7.03  |       |
| 31.0 1.1 16.5 1.8 704.3 77.5 85.4 9.39 369.2 40.61 71.8 7.90   |       |
| 31.6 1.0 16.0 1.8 499.1 54.9 108.1 11.89 116.7 12.83   |       |
| $\begin{array}{c ccccccccccccccccccccccccccccccccccc$  |       |
| 51.7 1.0 15.5 1.7 465.9 55.2 101.1 11.12 511.4 54.23 100.1 11.01<br>31.8 10 15.0 1.7 438.6 48.2 105.7 11.62 313.5 34.48 115.3 12.68  |       |
| 31.9 1.0 14.1 1.6 100 101 1100 12.10 299.6 32.96 106.4 11.70   |       |
| 32.0 1.0 14.5 1.6 399.6 44.0 96.1 10.57 325.4 35.80 105.2 11.57  |       |
| 32.0 0.9 14.2 1.6 396.6 43.6 139.3 15.33 273.5 30.08 153.3 16.86   32.0 0.9 14.2 1.6 396.6 139.3 15.33 16.86   |       |
| $\begin{array}{c ccccccccccccccccccccccccccccccccccc$  |       |
| 33.6 0.9 14.8 1.6 325.7 35.8 174.1 19.15 235.2 25.65 203.7 12.40   |       |
| 33.8 0.9 14.5 1.6 316.4 34.8 179.3 19.72 224.8 24.73 198.3 21.81   |       |
| 339 0.8 12.9 1.4 299.7 33.0 152.7 16.79 216.9 23.85 193.5 21.28  |       |
| 55.7 U.0 15.7 1.3 180.2 19.82 210.7 25.18 224.0 24.64   34.6 0.8 13.4 1.5 100.4 20.95 234.7 25.07 25.47  |       |
| 34.7 0.8 11.9 1.3 243.3 26.8 193.7 21.31 176.1 19.37 227.9 25.07   |       |
| 34.8 0.8 14.6 1.6 237.6 26.1 189.8 20.88 166.9 18.36 251.0 27.61   |       |
| 34.8 0.8 14.9 1.6 263.5 29.0 218.2 24.00 165.5 18.20 28.34 31.17   |       |
| 35.5 0.7 14.7 1.6 208.2 22.9 227.3 23.00 180.0 19.80 281.7 30.99   35.6 0.7 10.4 1.2 206.4 27.7 200.2 15.8 17.14 240.7 20.09   |       |
| 35.7 0.7 13.4 1.5 232.3 25.6 192.9 21.22 137.7 15.15 260.6 28.66   |       |
| 35.7 0.7 13.8 1.5 241.4 26.56 128.6 14.15 320.4 35.25  |       |
| 36.3 0.7 13.2 1.5 236.9 2606 304.3 33.47   |       |
| 30.5 U./ 15.8 1.5 202.4 22.3 253.8 25.72 137.9 15.17 291.6 32.07 36.4 102 21.1 202.4 1202.5 204.47 136.6 15.02 292.1 21.4  |       |
| $ \begin{array}{cccccccccccccccccccccccccccccccccccc$  |       |
| 36.6 0.7 12.9 1.4 185.0 20.4 204.6 22.50 126.8 13.95 273.8 30.12   |       |
| 36.6 0.7 12.5 1.4 240.5 26.45 118.6 13.04 309.8 34.08 2.2  | 0.2   |
| 30.9 U0 12.3 1.4 198.0 21.78 143.5 15.78 1<br>37.4 0.6 10.5 1.2 10 198.0 21.75 10.17 12.78 200.0   |       |
| $\begin{array}{c ccccccccccccccccccccccccccccccccccc$  | 0.3   |
| 37.8 0.6 12.7 1.4 162.6 17.9 216.0 23.76 154.3 16.98 50.17 53.19 2.5   | 0.0   |
| 38.3   0.5   11.7   1.3   154.3   17.0   252.4 <b>↓ 4</b> 7.76   137.6   15.14   343.3   37.76   12.9  | 1.4   |
| 38.3 0.5 13.4 1.5 167.0 18.4 261.6 28.77 225.1 24.76 357.8 39.35 9.8   | 1.1   |
| $\begin{array}{c ccccccccccccccccccccccccccccccccccc$  | 1.7   |
| $\begin{array}{cccccccccccccccccccccccccccccccccccc$   | 2.3   |
| 39.3 0.5 10.5 1.2 128.9 14.2 206.3 22.69 259.5 28.54 20.01 2 | 3.3   |
| 39.9 0.4 12.5 1.4 127.9 14.1 198.7 21.86 275.0 30.25 294.7 32.42 34.2  | 3.8   |
| 39.9 0.4 10.6 1.2 1000 321.4 35.35 290.4 31.95 41.5  | 4.6 2 |
| $ \begin{array}{c ccccccccccccccccccccccccccccccccccc$   | 5.1   |
| $\begin{array}{cccccccccccccccccccccccccccccccccccc$   | 7.0   |
| 40.9 0.4 8.6 0.9 98.9 10.9 160.1 17.61 436.9 48.05 70.6  | 7.8   |
| 415 0.3 11.0 1.2 111.1 12.2 180.5 19.85 274.6 30.20 91.5   415 0.3 10.2 1.1 11.1 12.2 180.5 19.85 1.2 274.6 30.20 91.5   | 10.1  |
| $\begin{array}{cccccccccccccccccccccccccccccccccccc$   | 10.9  |

Table 6: Measured cross-sections of the  ${}^{204m}$ Pb,  ${}^{203}$ Pb,  ${}^{202m}$ Pb,  ${}^{201}$ Pb and  ${}^{199}$ Pb reactions (ser. 1). Different cross-sections by the same energy come from different stack/irradiation

| $E \pm \Delta E(Me)$ | eV) | <b>Cross-sections</b> $(\sigma) \pm \Delta \sigma$ ( <b>mb</b> ) |      |                   |      |                    |      |                   |      |                   |      |
|----------------------|-----|--|------|-------------------|------|--------------------|------|-------------------|------|-------------------|------|
|                      |     | <sup>204m</sup> Pb   |      | <sup>203</sup> Pb |      | 202m <sub>Pb</sub> |      | 201 <sub>Pb</sub> |      | 200 <sub>Pb</sub> |      |
| 36.4                 | 0.3 | 11.4   | 1.4  | 204.2             | 23.0 | 227.2              | 24.6 | 113.1             | 12.7 | 270.7             | 29.3 |
| 35.2                 | 0.3 | 11.5   | 1.3  | 232.6             | 26.0 | 197.7              | 21.4 | 130.8             | 14.3 | 251.0             | 27.2 |
| 34.0                 | 0.3 | 13.6   | 1.5  | 275.8             | 30.5 | 176.2              | 19.1 | 166.6             | 18.1 | 206.1             | 22.3 |
| 32.8                 | 0.3 | 12.6   | 1.4  | 362.0             | 40.3 | 137.3              | 14.9 | 217.5             | 23.6 | 211.1             | 22.9 |
| 31.5                 | 0.4 | 14.3   | 1.6  | 468.1             | 51.0 | 103.6              | 11.2 | 275.5             | 29.9 | 88.5              | 9.6  |
| 30.2                 | 0.4 | 14.7   | 1.6  | 621.7             | 67.8 | 59.3               | 6.5  | 316.1             | 34.3 | 44.1              | 4.8  |
| 28.8                 | 0.4 | 16.8   | 1.9  | 769.6             | 83.7 | 26.4               | 2.9  | 355.0             | 38.5 | 11.7              | 1.4  |
| 27.4                 | 0.5 | 18.8   | 2.6  | 836.4             | 90.9 | 10.2               | 1.2  | 345.7             | 37.6 | 0.5               | 0.5  |
| 25.9                 | 0.5 | 22.5   | 2.7  | 858.9             | 93.4 | 11.8               | 1.4  | 338.7             | 37.0 |                   |      |
| 24.4                 | 0.6 | 28.0   | 3.1  | 768.7             | 83.5 | 17.0               | 1.9  | 287.3             | 31.3 |                   |      |
| 22.8                 | 0.6 | 45.7   | 5.2  | 686.2             | 74.7 | 26.6               | 2.9  | 223.7             | 24.3 |                   |      |
| 21.1                 | 0.7 | 73.7   | 8.2  | 532.2             | 58.0 | 31.0               | 3.4  | 127.8             | 14.0 |                   |      |
| 19.3                 | 0.8 | 96.2   | 10.8 | 244.2             | 26.8 | 28.6               | 3.2  | 22.8              | 3.5  |                   |      |
| 17.8                 | 0.9 | 75.8   | 8.2  | 65.1              | 8.2  | 25.7               | 2.8  |                   |      |                   |      |
| 16.2                 | 0.9 | 63.4   | 6.9  | 11.1              | 3.4  | 12.8               | 1.4  |                   |      |                   |      |
| 14.6                 | 1.0 | 33.4   | 3.6  | 20.3              | 2.9  | 10.4               | 1.2  |                   |      |                   |      |
| 12.7                 | 1.1 | 11.8   | 1.5  | 36.5              | 4.2  |                    |      |                   |      |                   |      |
| 10.6                 | 1.3 | 0.6  | 0.3  | 40.2              | 4.5  |                    |      |                   |      |                   |      |
| 9.3                  | 1.4 |  |      | 27.8              | 3.3  |                    |      |                   |      |                   |      |
| 7.9                  | 1.5 |  |      | 10.1              | 1.1  |                    |      |                   |      |                   |      |

Table 7: Measured cross-sections of the <sup>204m</sup>Pb,<sup>203</sup>Pb,<sup>202m</sup>Pb, <sup>201</sup>Pb, and <sup>200</sup>Pb reactions (ser. 2)

Table 8: Measured cross-sections of the  $^{202}$ Tl,  $^{201}$ Tl,  $^{200}$ Tl, and  $^{203}$ Hg reactions (ser.2)

| $E \pm \Delta E(M$ | leV) | <b>Cross-sections</b> ( $\sigma$ ) $\pm \Delta \sigma$ ( <b>mb</b> ) |     |                   |      |                                     |     |                   |      |  |
|--------------------|------|--|-----|-------------------|------|-------------------------------------|-----|-------------------|------|--|
|                    |      | 202 <sub>Tl</sub>  |     | 201 <sub>T1</sub> |      | 201 <sub>Tl</sub> 200 <sub>Tl</sub> |     | <sup>203</sup> Hg |      |  |
| 36.4               | 0.3  | 80.1   | 8.9 | 54.8              | 23.8 | 5.1                                 | 0.6 | 0.36              | 0.05 |  |
| 35.2               | 0.3  | 77.8   | 8.7 |                   |      | 2.5                                 | 0.3 | 0.26              | 0.07 |  |
| 34.0               | 0.3  | 67.6   | 7.6 | 51.8              | 28.4 | 2.9                                 | 0.3 | 0.25              | 0.07 |  |
| 32.8               | 0.3  | 64.8   | 7.3 |                   |      | 1.5                                 | 0.2 | 0.13              | 0.07 |  |
| 31.5               | 0.4  | 58.4   | 6.4 | 85.3              | 27.1 | 1.5                                 | 0.2 | 0.07              | 0.06 |  |
| 30.2               | 0.4  | 46.6   | 5.4 | 79.9              | 27.0 | 0.6                                 | 0.1 |                   |      |  |
| 28.8               | 0.4  | 46.0   | 5.3 | 99.6              | 29.9 |                                     |     |                   |      |  |
| 27.4               | 0.5  | 41.7   | 4.8 | 149.5             | 28.9 |                                     |     |                   |      |  |
| 25.9               | 0.5  | 33.7   | 4.0 | 98.9              | 23.9 |                                     |     |                   |      |  |
| 24.4               | 0.6  | 30.8   | 3.6 | 72.0              | 23.9 |                                     |     |                   |      |  |
| 22.8               | 0.6  | 26.4   | 3.2 | 112.8             | 29.2 |                                     |     |                   |      |  |
| 21.1               | 0.7  | 20.5   | 2.6 | 38.6              | 11.8 |                                     |     |                   |      |  |
| 19.3               | 0.8  | 14.6   | 1.8 | 1.0               | 8.7  |                                     |     |                   |      |  |
| 17.8               | 0.9  | 10.3   | 1.3 |                   |      |                                     |     |                   |      |  |
| 16.2               | 0.9  | 5.4  | 0.8 |                   |      |                                     |     |                   |      |  |
| 14.6               | 1.0  | 2.2  | 0.3 |                   |      |                                     |     |                   |      |  |
| 12.7               | 1.1  | 0.8  | 0.2 |                   |      |                                     |     |                   |      |  |
| 10.6               | 1.3  |  |     |                   |      |                                     |     |                   |      |  |
| 9.3                | 1.4  | 0.3  | 0.1 |                   |      |                                     |     |                   |      |  |
| 7.9                | 1.5  | 0.4  | 0.1 |                   |      |                                     |     |                   |      |  |

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