

# Measurements of cross-sections of the proton-induced activation reactions

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Excitation functions for the  $^{89}\text{Y}(p,x)^{89,88,86}\text{Zr}$ ,  $^{89}\text{Y}(p,x)^{88,87,87m,86}\text{Y}$ ,  $^{89}\text{Y}(p,x)^{85,83,82}\text{Sr}$  and  $^{89}\text{Y}(p,x)^{84,83}\text{Rb}$  reactions were measured by a stacked foil activation technique in the energy range 15-80 MeV. The production for the long lived products like  $^{88}\text{Zr}$ , and  $^{88}\text{Y}$  are significantly larger than that of  $^{\text{nat}}\text{Mo}+p$ ,  $^{\text{nat}}\text{Nb}+p$  and  $^{\text{nat}}\text{Zr}+p$  processes. The productions of the medical isotopes,  $^{85}\text{Sr}$  and  $^{83}\text{Sr}$  are also effective by Y+p process using 80 MeV beam. The model calculations using ALICE-IPPE code compiled in MENDL-2P have the general trend of the measured results.

## 1. Introduction

Yttrium is widely applied to increase the strengths of alloys of important metals of nuclear technology (aluminum, magnesium and chromium). The proton-induced activation cross-section on this element is important for dose estimation in accelerator technology, for thin layer activation analysis (TLA) of yttrium alloys (especially of aluminum and magnesium) and of yttrium oxide ceramics. Yttrium is a monoisotopic element therefore ideal target material to test nuclear reaction theories.

The  $^{86,87,88}\text{Y}$  radioisotopes have value for investigation of the biodistribution of  $^{90}\text{Y}$  labeled therapeutic compounds.  $^{88}\text{Y}$  is a recognized standard for calibration of gamma-ray detector.  $^{88}\text{Y}$  is produced directly by particle-induced activation and/or via generator system from  $^{88}\text{Zr}$ . Yttrium can be used as target for the production of medically related radioisotopes,  $^{82}\text{Sr}$ ,  $^{85}\text{Sr}$ ,  $^{83}\text{Sr}$  and biological tracers,  $^{84}\text{Rb}$ ,  $^{83}\text{Rb}$  (water transports in plants, etc.) and for a large-scale production of  $^{88}\text{Zr}$  and  $^{88}\text{Y}$  activities.

Few authors have reported data for proton-induced activation reactions on yttrium at low energies limited to maximum energies 5 MeV to 45 MeV [1-5]. A Large discrepancy is found between them.

In this work we present the new cross-sections for Y+p reactions as a part of systematic studies on particle-induced activations [6-9]

## 2. Experimental technique and data evaluation

The independent and “cumulative cross-sections” of the proton-induced reactions on yttrium were measured as a function of proton energy using a conventional stacked foil activation technique. The irradiation, the activity measurement and the data evaluation were similar as described in more details in our recent works [6-9]. Here we summarize some features specific for the investigation. Two stacks of several identical groups containing high purity thin yttrium foils (110  $\mu\text{m}$  thick) were irradiated separately with 50 MeV (~165 nA for 30 min) and 80 MeV (~45 nA for 70 min) collimated protons, respectively using a K=90 MeV AVF cyclotron at Cyclotron and Radioisotope Center (CYRIC), Tohoku University, Sendai, Japan. In this way the proton beam energy range of 15-80 MeV was covered. Reactions induced on aluminum (100 and 1000  $\mu\text{m}$  thick) and copper (54  $\mu\text{m}$  thick) foils were used to monitor the parameters of the bombarding beam and as proton beam energy degrader.

The residual activity was measured nondestructively by the high-resolution HPGe gamma-ray spectroscopy. The efficiency *versus* energy curves of the HPGe-detector for various counting distances were established using calibrated standard gamma-ray point sources. The effective particle energy in the foils was calculated by the computer program SRIM-2003 [10] using measured thicknesses of the foils. The beam current was determined by the monitor reactions  $^{\text{nat}}\text{Cu}(p,x)^{56}\text{Co}$ ,  $^{62,65}\text{Zn}$  and  $^{27}\text{Al}(p,x)^{22,24}\text{Na}$  [11]. The decay data for the investigated radionuclides were taken from NUDAT database [12].

The peak area data were corrected for the sum-coincidence effect caused by the coincidental detection of two or more gamma rays in cascade by using the SUMECC code [13]. In some cases, the contributions from overlapping peaks were separated by using the independent gammas of the contributing nuclides.

The combined uncertainty on each cross-section was estimated by taking the square root of the quadratic sum of the following errors: statistical error (0.15-6 %), error of the monitor flux (3-5 %), the error in efficiency calibration of the gamma-ray (~3 %) and foil thickness (3 %).

### 3. Results and discussion

The measured excitation functions are shown in Figs. 1-9 together with literature values. The results of this work are compared with reported experimental data and ALICE-IPPE calculation compiled in MENDL-2P [14].

#### 3.1 $^{89}\text{Y}(p,xn)^{89,88,86}\text{Zr}$

The measured excitation function for the production of  $^{89}\text{Zr}$  is shown in Fig.1. The contribution from the decay of the 4.18 min isomeric state  $^{89m}\text{Zr}$  is included in the measured cross-section. Saha et al. [15] reported the  $^{89}\text{Zr}$  production cross-section in our investigated energy range. The results of Saha et al. moved upward in the range 18-30 MeV and out of this energy range showing consistency with this work. The results of this work are in good agreement with Birattari et al [1], Johnson et al. [2], Levkovski [3] and Mustafa et al. [4] in the lower energy region and with MENDL-2P and that fact confirm our measurements as reliable.

The independent cross-sections for the  $^{88}\text{Zr}$  production through  $^{89}\text{Y}(p,2n)$  reaction were measured that are shown in Fig.2. About 98 % consistencies of the measured values for the  $^{88}\text{Zr}$  production with the experimental values of Saha et al. are found above 40 MeV, but large discrepancies are present in lower energy region. The results of this work are about 62 % lower and 8 % higher than Saha et al. and ALICE-IPPE calculation in peak region, respectively. The measured data are in good agreement with Mustafa et al. who reported below 40MeV. The  $^{88}\text{Zr}$  production cross-sections reported by Levkovski are unexpectedly rising in peak region.

The measured cross-sections for the  $^{89}\text{Y}(p,4n)^{86}\text{Zr}$  reaction are shown in Fig.3. The present results are consistent with Saha et al. in shape, but not in magnitudes. The shape of excitation function for the production  $^{86}\text{Zr}$  obtained from the ALICE-IPPE calculation is not acceptable. Caretto et al. [16] reported only two points, which have common trend with this work.

#### 3.2 $^{89}\text{Y}(p,x)^{88,87,87m,86}\text{Y}$

The cross-sections for the production of  $^{88}\text{Y}$  shown in Fig.4 were measured. The contributions in the measured values are the internal transition decay of the short-lived ( $T_{1/2} = 13.9$  ms)  $^{88m}\text{Y}$  isomer. The measurement was done one day after EOB. The half-life of  $^{88}\text{Zr}$  is 83.4 days that is so much higher than cooling time. Hence the effect of the decay of  $^{88}\text{Zr}$  on the  $^{88}\text{Y}$  production cross-section is very low that has been separated. No cumulative data are found in literature for the  $^{88}\text{Y}$  production in the investigated energy range.

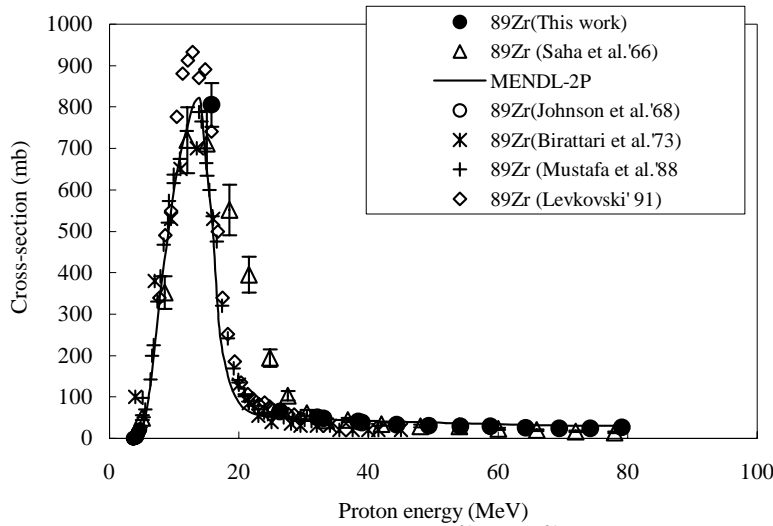


Fig. 1 Excitation functions of the  $^{89}\text{Y}(p,x)^{89}\text{Zr}$  reaction.

The measured  $^{87m}\text{Y}$  production cross-sections (Fig.5) are due to the direct process induced on  $^{89}\text{Y}$  and the decay of  $^{87}\text{Zr}$ . The overlapping peaks for  $^{83}\text{Sr}$  and  $^{86}\text{Y}$  on the 380.7 keV gamma energy of  $^{87m}\text{Y}$  were separated by using other independent gammas of the contributing nuclides. A good consistency was found between the results of this work and Michel et al. [17].

$^{87}\text{Y}$  is produced by  $^{89}\text{Y}(p,p2n)$  reaction and by 98.43 % internal transition of the metastable state  $^{87m}\text{Y}$  ( $T_{1/2}=13.37$  h). It was paid attention for the complete internal decay of metastable state to measure cumulative production cross-sections shown in Fig.5. Michel et al. reported cumulative cross-section. The results of this work

are in good agreement with Michel et al. Saha et al. reported about two times lower values than this work. The result of ALICE-IPPE calculation shows similar overall general behavior as the experimental data.

The measured values for the production of  $^{86}\text{Y}$  shown in Fig.6 contain two contributions like  $^{89}\text{Y}(p,p3n)$  direct process and the decay of metastable state. The present results are similar to Michel et al. and MENDL-2P in shape.

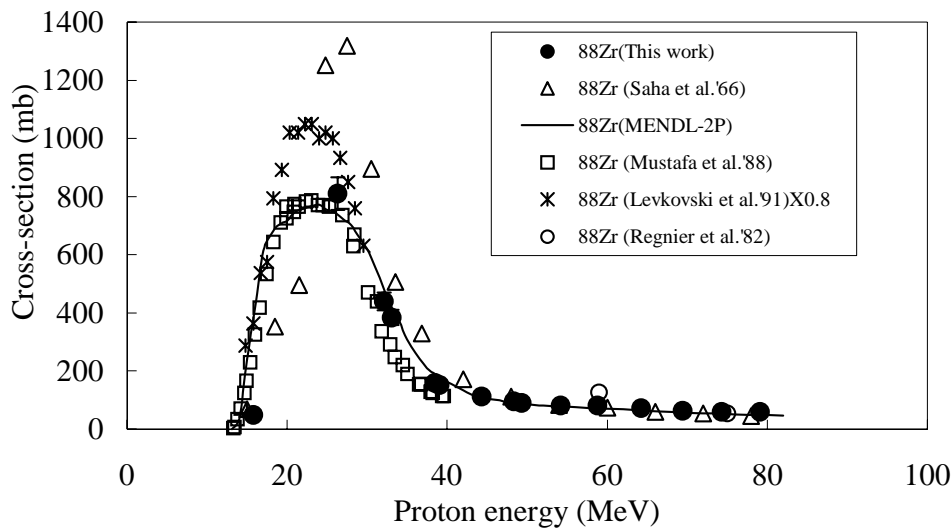


Fig. 2 Excitation functions of the  $^{89}\text{Y}(p,x)^{88}\text{Zr}$  reaction.

### 3.3 $^{89}\text{Y}(p,x)^{82,83,85}\text{Sr}$

$^{85}\text{Sr}$  has a large importance in nuclear medicine as diagnostic radioisotope and for the endotherapy. The  $^{85}\text{Sr}$  nuclide is also an important impurity in the application of the medical radioisotopes  $^{82}\text{Sr}$  and  $^{83}\text{Sr}$ . The  $^{85}\text{Sr}$  production cross-section was measured using 514 keV gamma. To separate the 514 keV gamma line from the strong annihilation peak (511 keV), the measurement was done about 40 days after end of irradiation and obtained separate peak through our high resolution gamma spectroscopy system. This work gives the cumulative cross-section shown in Fig.7 contained the contributions of direct process, decay of isomeric state and EC decay of  $^{85}\text{Y}$ . The results of this work are in good agreement with Michel et al. The calculated results compiled in MENDL-2P are also consistent with this work both in shape and magnitudes above 40 MeV

The  $^{82}\text{Sr}$  ( $T_{1/2}= 25.55$  d) production cross-sections shown in Fig.8 were determined by measuring 776 keV gamma line produced through the decay of its daughter radioisotope  $^{82}\text{Rb}$  ( $T_{1/2}= 1.273$  min). The  $^{86}\text{Y}$  ( $T_{1/2}= 14.74$  h),  $^{83}\text{Sr}$  ( $T_{1/2}= 1.35$  d) and  $^{82\text{m}}\text{Rb}$  ( $T_{1/2}= 6.47$  h) radionuclides emitted gamma-rays having very close energies to 776 keV which were difficult to separate by the graphical analysis of the multicomponent decay curve. After >40 days, these contributing radionuclides completely decayed out and the remaining activity was due to decay of the daughter nuclide  $^{82}\text{Rb}$  in transient equilibrium with the parent  $^{82}\text{Sr}$  radionuclide. The measured values are in good agreement with Michel et al. The results of ALICE-IPPE calculations have similar shape, but overestimate the experimental values.

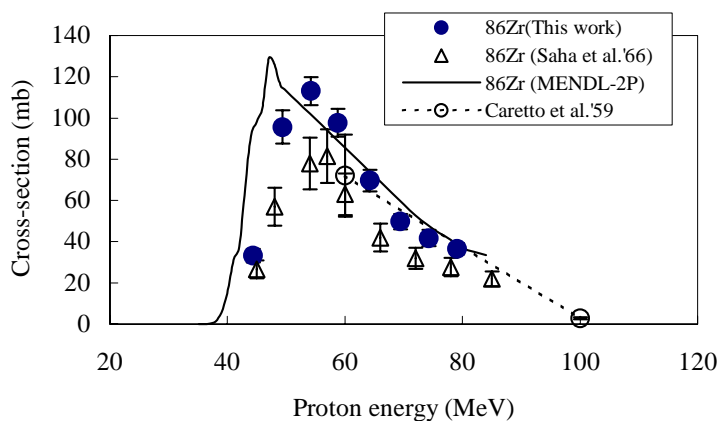


Fig. 3 Excitation functions of the  $^{89}\text{Y}(p,x)^{86}\text{Zr}$  reaction.

$^{83}\text{Sr}$  is a suitable positron emitter analog of the  $\beta^-$ -emitting  $^{89}\text{Sr}$  ( $T_{1/2}= 50.5$  d) used for endotherapy. The isomeric state  $^{83\text{m}}\text{Sr}$  have too short half-life ( $T_{1/2}= 4.95$  s) that decayed completely to the ground state and its role as isomer in production of the ground state is negligible.  $^{83}\text{Sr}$  is produced via  $^{89}\text{Y}(p,2p5n)$  reaction and the decay of  $^{83}\text{Zr}$  and  $^{83}\text{Y}$ . The results of this work are shown in Fig.8 together others. Michel et al. and ALICE-IPPE calculation well reproduce the shape of the measured excitation function.

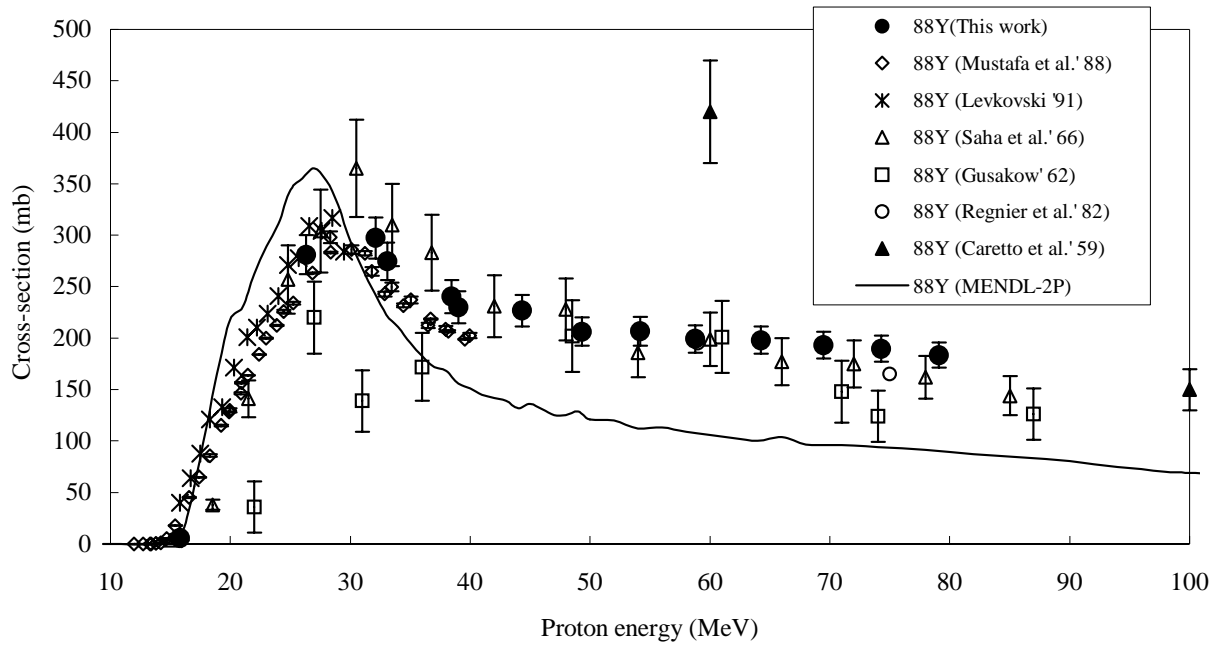


Fig.4 Excitation functions of the  $^{89}\text{Y}(p,x)^{88}\text{Y}$  reaction.

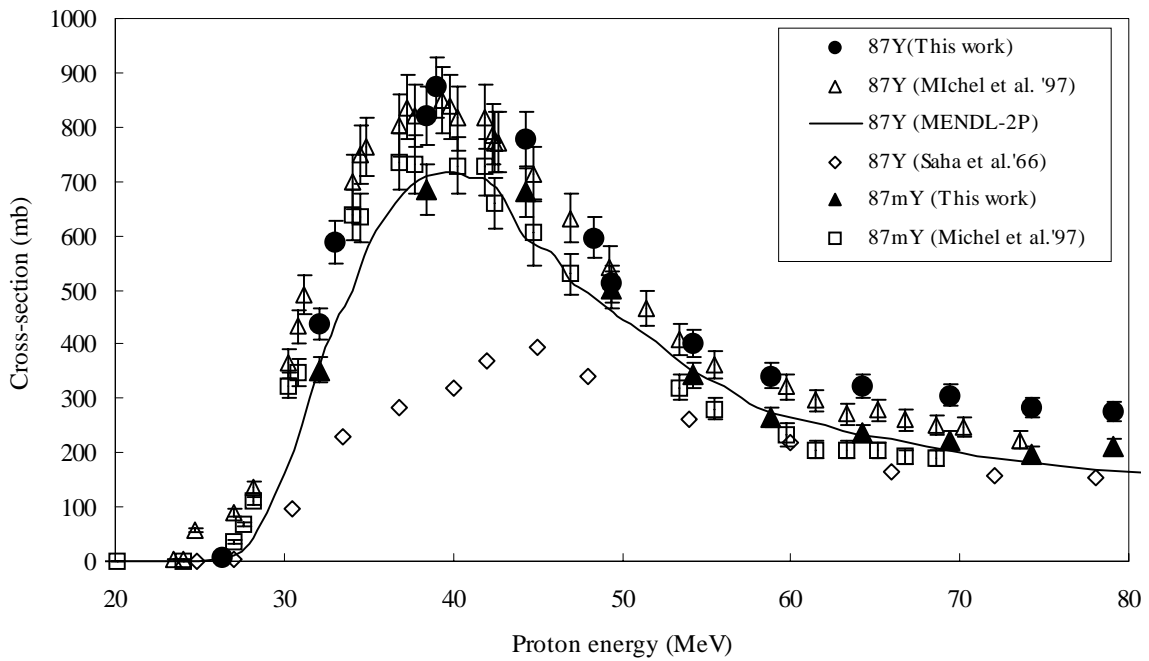


Fig.5 Excitation functions of the  $^{89}\text{Y}(p,x)^{87,87m}\text{Y}$  reaction.

### 3.4 $^{89}\text{Y}(p,x)^{84,83}\text{Rb}$

The  $^{84}\text{Rb}$  production cross-sections shown in Fig.9 were measured after the complete decay of  $^{84\text{m}}\text{Rb}$  ( $T_{1/2} = 20.26$  min), i.e. the obtained results are the sum of the ground and the isomeric state cross-sections.  $^{84}\text{Rb}$  is produced by  $^{89}\text{Y}(p,\alpha\text{pn})$  reaction. The probability of this reaction occurring is largely inhibited by the Coulomb barrier in both the entrance and exit channels. The results of this work are consistent with Michel et al. A large discrepancy is found between the measured and calculated values.

This work presents the cumulative cross-section for the production of  $^{83}\text{Rb}$  shown in Fig.9, which contains contribution from the decay of  $^{83}\text{Sr}$ . The sharply increase in excitation function above 35 MeV is probably due to more independent particle emission. Only one earlier data reported by Michel et al. are also shown in Fig.9. The results of this work are in about 97% agreement with Michel et al. The ALICE-IPPE calculation underestimates the measured values.

## 4. Conclusion

The present work gives new cross-section data for all of the investigated radionuclides. In some cases, our data are consistent with the results of Michel et al. The data in MENDL-2P calculated with the theoretical code ALICE-IPPE are consistent in shape with the measured values, but disagreement in magnitudes. Two stacks were irradiated in different conditions and observed good agreement between the obtained results in the overlapping energy range. The reliability of the data is guaranteed by using the relative method. The results of this work will play an important role to test model calculations. The cross-sections obtained in this study useful to estimate the optimal production yields and impurities in the energy range from threshold up to 80 MeV.

It seems to us the production of  $^{85}\text{Sr}$  will be effective by proton-induced activation on yttrium using accelerator having 80 MeV proton beam. It is also possible to give production of  $^{83}\text{Sr}$  through  $^{89}\text{Y}(p,x)$  reactions. From the point of view of the production yields the Y+p route is much more efficient to give productions of  $^{88}\text{Zr}$  and  $^{88}\text{Y}$  than  $^{\text{nat}}\text{Mo}+p$ ,  $^{\text{nat}}\text{Nb}+p$  and  $^{\text{nat}}\text{Zr}+p$  processes.

The data base is promising to prepare recommended data sets for practical TLA applications. Among all investigated products the  $^{87}\text{Y}$ ,  $^{88}\text{Y}$ ,  $^{88}\text{Zr}$  and  $^{89}\text{Zr}$  radionuclides are the most suitable for thin layer activation analysis, but the  $^{87}\text{Y}$  and  $^{89}\text{Zr}$  only for investigations of short processes. It seems to us the proton-induced activation on yttrium can be effectively used for wear studies in alloys and ceramics by using TLA.

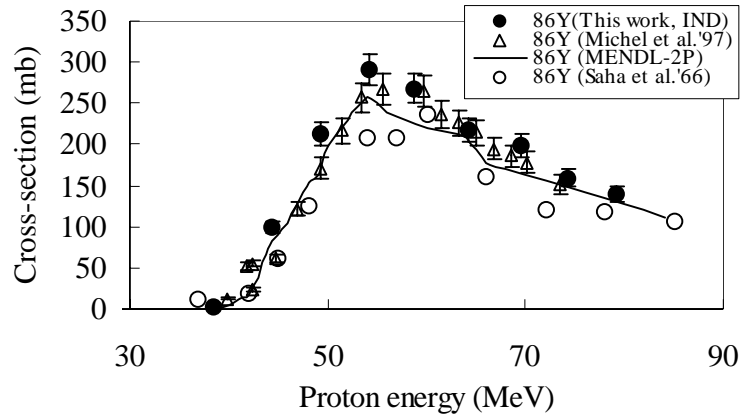


Fig.6 Excitation functions of the  $^{89}\text{Y}(p,x)^{86}\text{Y}$  reaction.

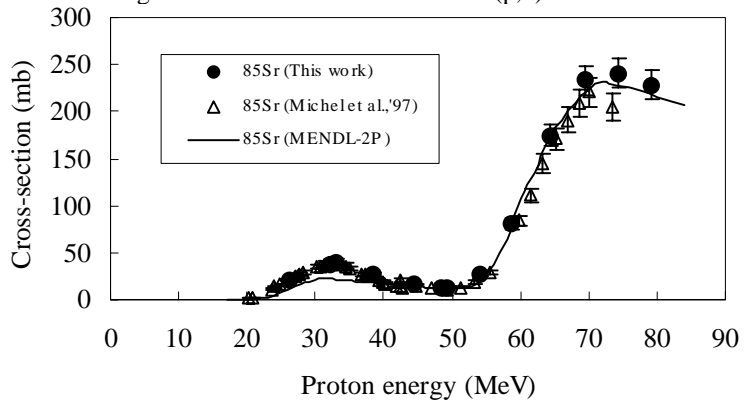


Fig.7 Excitation functions of the  $^{89}\text{Y}(p,x)^{85}\text{Sr}$  reaction.

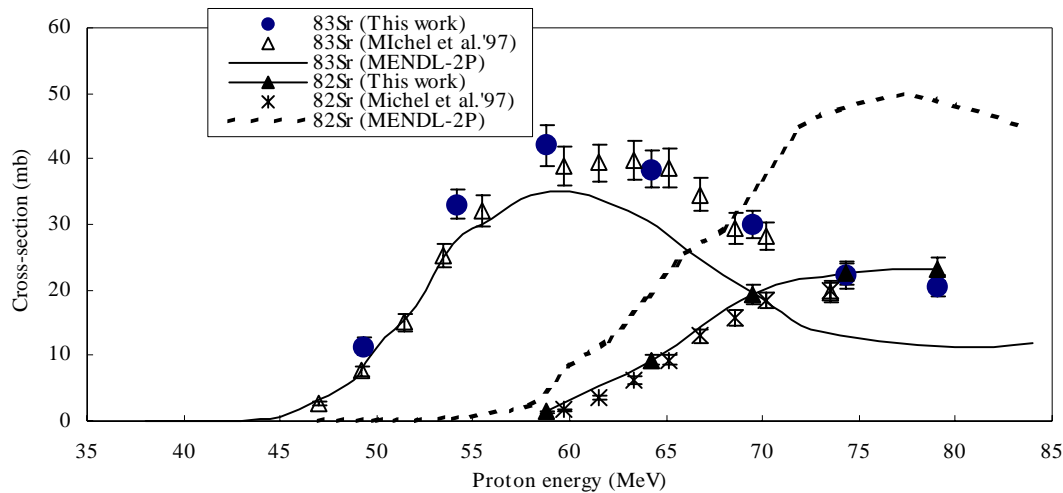


Fig.8 Excitation functions of the  $^{89}\text{Y}(p,x)^{82,83}\text{Sr}$  reaction.

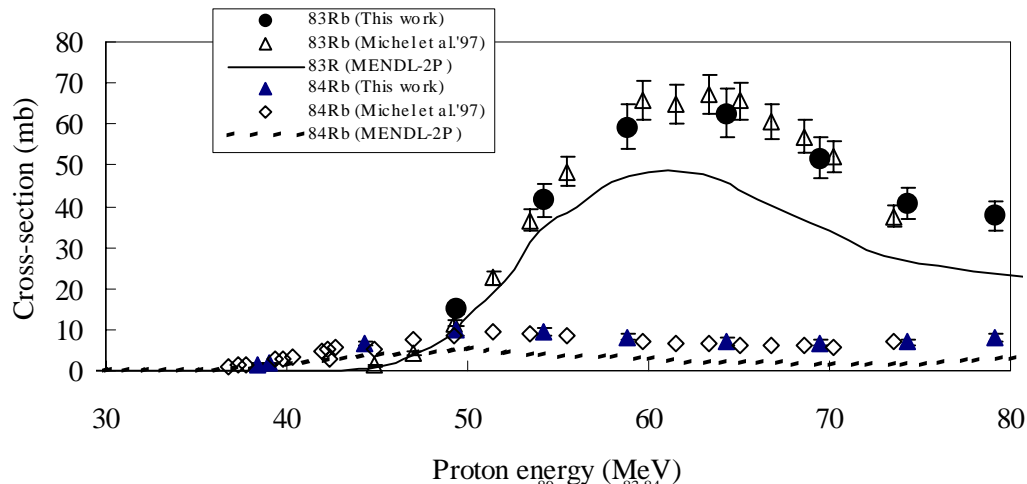


Fig.9 Excitation functions of the  $^{89}\text{Y}(p,x)^{83,84}\text{Rb}$  reaction.

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