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Application of Au + p nuclear reactions for proton beam monitoring up to 70 MeV

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Abstract. Experimentally measured excitation functions are presented for the ^{197}Au(p,n)^{197m}Hg, ^{197}Au(p,pn)^{196}Au, ^{197}Au(p,3n)^{195m}Hg, ^{197}Au(p,p3n)^{194}Au and ^{197}Au(p,5n)^{193m}Hg nuclear reactions up to 70 MeV. The new data are compared with published values, where available. Based on cubic spline fits through the data of the most reliable experimental data sets, recommended excitation function curves are suggested for the above nuclear processes.

1 Introduction

Gold is frequently used as a target backing material in nuclear physics experiments as well as in nuclear data measurements for radioisotope production. Irradiation of gold with protons results in several nuclear processes (forming radioactive isotopes of Au and Hg) that can be potentially useful for beam energy and intensity monitoring. As already shown in an earlier study (see ref. [1]), the reactions ^{197}Au(p,n)^{197m}Hg and ^{197}Au(p,pn)^{196}Au are useful for proton beam monitoring below 30 MeV. In this work, we further investigated the excitation functions of the Au+p reactions with the aim of extending the applicability of this monitor material up to 70 MeV.

2 Experimental

The cross sections were measured using the conventional stacked-foil method. One stack, containing 21 pieces of high-purity gold foils (99.99%, supplied by Goodfellow Metals), was irradiated with an external proton beam supplied by the separated sector cyclotron of iThemba LABS. The gold foil thickness of 4.86 μm was accurately determined by measuring the energy degradation of alpha particles emitted from a ^{228}Th source. The stack also contained high-purity Al and Cu degrader foils to obtain well spaced proton energies across the entire investigated energy region. The incident bombarding energy was 65.4 ± 0.4 MeV. The irradiation time was 3.3 h with an average beam current of 50 nA. Decay data such as half-lives, γ-ray energies, branching ratios, etc., were obtained from ref. [2]. The activities induced in the irradiated foils were measured by means of standard, off-line γ-ray spectroscopy, utilizing an accurately calibrated HPGe detector with a relative efficiency of 13% and a resolution of 1.7 keV for γ-rays of 1.3 MeV. No chemical separations were performed. The combined uncertainty of the measured cross sections varied between 12% and 15%.

3 Results and discussion

The investigated nuclear reactions suggested for monitor purposes are ordered according to increasing mass number of the residual nuclei. "Recommended" excitation function curves were calculated using the cubic spline fitting method in those cases where an acceptable database of a given reaction could be compiled (for details of the methods, see ref. [3]). The recommended values, tabulated in 2 MeV increments, are presented in table 1.

3.1 The ^{197}Au(p,5n)^{193m}Hg reaction

The (p,5n) reaction populates an isomer of ^{193}Hg which is longer-lived than the ground state: $T_{1/2}$(metastable) = 11.8 h and $T_{1/2}$(ground) = 3.8 h. The metastable state decays predominantly to ^{193}Au (92%) by electron capture. Unfortunately, it also populates the ground state of ^{193}Hg (8%) by internal transition (IT). From a beam monitoring point of view, therefore, the ground state is not useful as it is continually fed from the decay of the longer-lived metastable state. In contrast, the metastable state has potential for this purpose. The activity of ^{193m}Hg can be measured easily through its γ-lines at 499 keV (Iγ = 4.7%) and 537 keV (Iγ = 26%). As a further advantage, this monitor reaction can be employed without any cooling time above 33 MeV. Prior to our study, no cross-section data were reported in the literature. Our measured values (16 data points) are presented in figure 1. The fitted excitation function curve for this reaction peaks at about 48.6 MeV with a maximum of 308 mb.

3.2 The ^{197}Au(p,p3n)^{194}Au reaction

In addition to the ground state, the m1 and m2 metastable states of ^{194}Au are also populated. However, these states are quite short-lived ($T_{1/2}$(m1) = 420 ms and $T_{1/2}$(m2) = 600 ms). They both decay rapidly to the ground state ($T_{1/2}$(ground) = 1.6 d) by IT, therefore the cumulative ^{194}Au ground-state cross sections can be used for practical purposes. It should be noted that the decay of the ^{194}Hg co-produced during the activation of the gold foils above 24 MeV (^{197}Au(p,4n), E_{thr} = 24.07 MeV), also populates the ground state of ^{194}Au. However, due to the very long half-life ($T_{1/2}$ = 520 y) of ^{194}Hg, its contribution is negligible. There is another contributing process, namely the ^{197}Au(n,4n) reaction, which also

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Table 1. Recommended fitted cross sections for the ^{197}Au(p,5n)^{193m}Hg, ^{197}Au(p,p3n)^{194}Au, ^{197}Au(p,3n)^{195m}Hg, ^{197}Au(p,pn)^{196}Au and ^{197}Au(p,n)^{197m}Hg nuclear reactions up to 70 MeV.

Energy	Cross-section (mb)				
(MeV)	(p,5n)	(p,p3n)	(p,3n)	(p,pn)	(p,n)
6					0.50
8					2.02
10					23.8
12					42.5
14				0.99	25.7
16				4.27	17.8
18				16.5	14.0
20			120	38.5	12.1
22			304	65.0	11.1
24			485	90.6	10.2
26		1.54	643	113	9.82
28		2.98	733	131	9.46
30		4.61	625	144	9.06
32		6.85	420	153	8.61
34		10.8	252	159	8.39
36	11.9	18.8	177	163	8.00
38	49.3	31.1	154	165	7.60
40	110	46.3	143	165	7.21
42	180	62.5	135	166	7.00
44	242	78.2	125	166	6.75
46	286	92.6	115	165	6.49
48	307	105	106	164	6.11
50	301	116	99.1	164	5.99
52	271	124	93.4	164	5.65
54	228	131	89.1	163	5.45
56	185	136	85.4	162	5.35
58	151	140	81.3	161	5.25
60	125	143	76.7	160	5.05
62	108	144	72.0	159	4.95
64	96.2	146	68.0	157	4.85
66		146	64.0	155	4.65
68		146	60.8	153	4.49
70		145	57.5	151	4.35

forms the residual nucleus ^{194}Au. Neutrons come from the experimental set-up (e.g., the other gold foils, degrader foils, collimators, etc.), and their influence to the ^{194}Au production could cause problems, especially close to the threshold energy region of the ^{197}Au(p,p3n)^{194}Au reaction. To evaluate the magnitude of this 'interference', it is suggested that some gold foils should be exposed separately to the neutron background of an experiment. However, based on our previous experience [4], this interference is generally small (< 0.1%). Similar to the case of the ^{197}Au(p,5n)^{193m}Hg reaction, this monitor reaction can also be used without cooling time. The ^{194}Au activity can be measured through two strong γ-lines at 294 keV (Iγ = 10.4%) and 328 keV (Iγ = 61%). Up to 2007, only two groups [5,6] reported total cross-section values in the energy region up to 70 MeV. The different experimental data sets, including our 21 new data points, show acceptable agreement with each other. All available experimental results are shown in figure 2, together with the fitted excitation function curve. This excitation function shows a maximum of 146 mb at about 67.2 MeV.

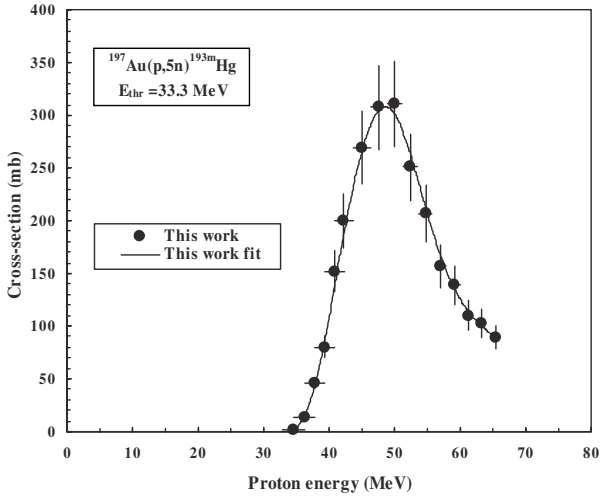

Fig. 1. Cross-sections of the ^{197}Au(p,5n)^{193m}Hg nuclear reaction.

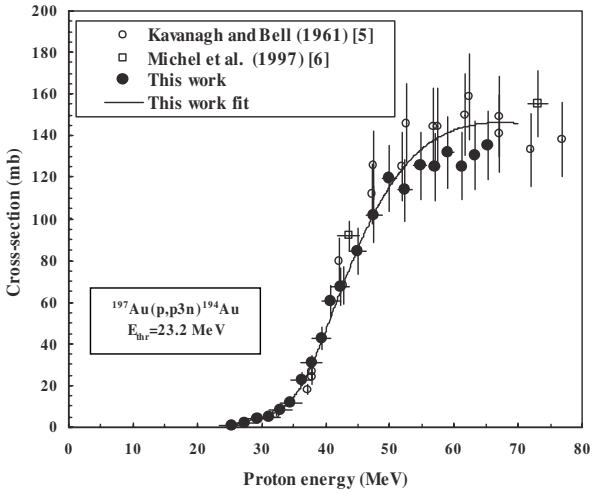

Fig. 2. Cross-sections of the ^{197}Au(p,p3n)^{194}Au nuclear reaction.

3.3 The ^{197}Au(p,3n)^{195m}Hg reaction

Proton bombardment of gold above 17 MeV populates two relatively long-lived states of ^{195}Hg. Unfortunately, the metastable state ($T_{1/2}$(metastable) = 1.73 d) has a longer half-life than the ground state ($T_{1/2}$(ground) = 9.5 h) and it decays partly to the ground state (IT = 54%). Therefore, only the metastable state can be used for beam monitoring purposes. It has two relative strong γ-lines, 261 keV (Iγ = 30.9%) and 560 keV (Iγ = 7%), which can be employed for immediate activity measurement. Our systematic study has yielded 20 new cross section values up to 70 MeV. By comparing them with the available experimental data of four different groups (refs. [6–9]), it can be concluded that, in general, the database of this reaction seems to be acceptable. However, the values of Nagame et al. [9] are systematically lower than those of other authors above 40 MeV. The fitted excitation function curve of the ^{197}Au(p,3n)^{195m}Hg reaction has a peak at about 28.0 MeV with a maximum of 730 mb. The results are plotted in figure 3.

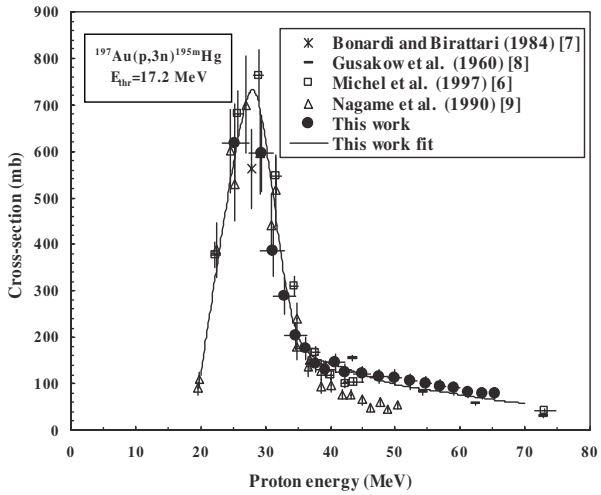

Fig. 3. Cross-sections of the ^{197}Au(p,3n)^{195m}Hg nuclear reaction.

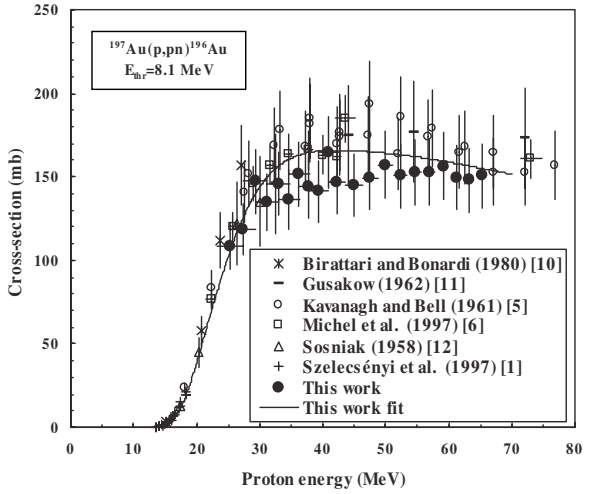

Fig. 4. Cross-sections of the ^{197}Au(p,pn)^{196}Au nuclear reaction.

3.4 The ^{197}Au(p,pn)^{196}Au reaction

Similarly to the ^{197}Au(p,p3n)^{194}Au reaction discussed above, here also the cumulative ground-state cross sections can be used for practical purposes. Two metastable states are also prominent for this reaction: $T_{1/2}(m_1)$ = 9.7 h and $T_{1/2}(m_2)$ = 8.1 s. Since $T_{1/2}$(ground) = 6.18 d, a relatively long cooling time will be necessary for the metastable states to decay to an insignificant level (around 100 h). Luckily, the co-produced ^{196}Hg is stable, therefore it does not populate any states of ^{196}Au. The influence of the secondary neutrons is also negligible for similar reasons as discussed earlier. The activity of ^{196}Au can be determined using three major γ-lines at 33 keV (Iγ = 22.9%), 356 keV (Iγ = 87%) and 426 keV (Iγ = 7%). This reaction was studied in detail by several groups in the past [1,5,6,10–12]. The available results are reproduced in figure 4, together with our values (21 points) up to 80 MeV. The majority of the results show acceptable agreement with each other. Our values, however, are systematically somewhat lower than the others. The fitted excitation function curve reaches a maximum of 166 mb at around 42.5 MeV.

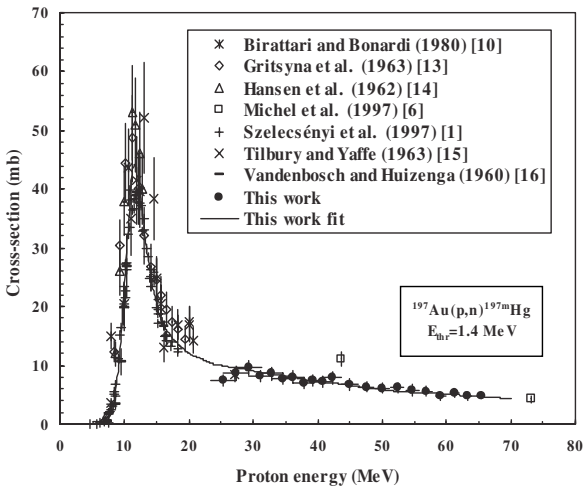

Fig. 5. Cross-sections of the ^{197}Au(p,n)^{197m}Hg nuclear reaction.

3.5 The ^{197}Au(p,n)^{197m}Hg reaction

Two relatively long-lived states of ^{197}Hg are formed via the (p,n) reaction: $T_{1/2}$(metastable) = 23.8 h and $T_{1/2}$(ground) = 2.67 d. The metastable state decays almost completely to the ground state (IT = 93%). In practice, only the metastable state is useful for proton beam energy or intensity estimation. Theoretically, the cumulative ground-state cross sections can be employed for the above purposes, however, in this case a significant cooling time would be necessary (for the metastable state to decay) before starting the measurements (around 300 h). As a result, the ground state ^{197}Hg activity of the foils would become very weak, which may lead to high experimental uncertainties. The ^{197m}Hg has only one strong γ-line at 134 keV (Iγ = 33%). Searching the literature, eight authors [1,6,10,14–16] were found who reported cross sections for this reaction below 80 MeV. Note that our present measurements added new values only to the slope part of the excitation function curve. (A compilation of the available results up to 18 MeV was already published previously by Szelecsényi et al. [1].) The majority of the results show acceptable agreement with each other. The experimental results are shown in figure 5, together with the fitted excitation function curve. This reaction shows a maximum of 44.3 mb at about 11.6 MeV.

4 Conclusions

Based on the results of the present measurements and selected literature data, the database for the ^{197}Au(p,n)^{197m}Hg, ^{197}Au(p,pn)^{196}Au, ^{197}Au(p,3n)^{195m}Hg, ^{197}Au(p,p3n)^{194}Au and ^{197}Au(p,5n)^{193m}Hg reactions can be considered to be satisfactory. The activities of ^{197m}Hg, ^{195m}Hg and ^{193m}Hg can be measured immediately after bombardment for beam monitoring purposes, as these radionuclides are directly produced. In the case of ^{196}Au and ^{194}Au, the cumulative ground state activity should be measured after the decay of their respective isomers. For ^{194}Au, the metastable states have half-lives less than 1 s, thus in practice no cooling period is required. In contrast, a cooling time of about 100 h is

required for ^{196}Au. The fitted excitation function curves can be used for beam monitoring purposes in different energy regions up to 70 MeV. We suggest, however, further cross-section measurements in the energy region of 20–40 MeV to strengthen the database in the cases of the ^{197}Au(p,3n)^{195m}Hg and ^{197}Au(p,n)^{197m}Hg reactions.

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