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Isotope separation of ^{176}Lu a precursor to ^{177}Lu medical isotope using broadband lasers

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A new photoionization scheme accessible by Rhodamine dye lasers is proposed for the isotope separation of ^{176}Lu .

Optimum conditions for the isotope separation have been derived and compared with the previously reported work. The enrichment of $\sim 49\%$ can be obtained with $> 12\text{ mg / hour}$ production rates even when broadband lasers with bandwidth of 500 MHz employed for the two step excitation. The simplified system requirements for the photoionization scheme with a high production rate is expected reduce the global shortage of ^{176}Lu for medical applications.

Chemistry which is a science of elements, compounds and their separation has immensely helped in the development of mankind. However, separation of isotopes remained a formidable challenge as it is technology intensive and therefore it is largely limited only to the technologically advanced nations. Some of the isotopes of the low Z-elements could be separated relatively easily by the chemical methods by exploiting the difference in the kinetic isotope effect¹. Since the kinetic isotope effect for mid-Z and high-Z elements will be gradually diminishing with increase in Z, it is therefore required to switchover to physical methods for the efficient separation of isotopes. Isotope separation technology largely limited to the developed nations has been applied to the separation of actinides (particularly Uranium) due to its applications in the defence and nuclear industry². However the real potential of isotope separation for the separation of rest of the isotopes remains

unexploited due to limited access to this technology to the most of the nations and the complexity of the systems involved.

Among the several methods of the separation of isotopes, gas centrifugation method has evolved as the most commercially viable method, however, apart from the technological challenges, the primary limitation, remains being highly capital intensive. On the other hand, Atomic Vapor Laser Isotope Separation (AVLIS) has evolved as an alternative technique to the gas centrifugation method. AVLIS has significant advantages over gas centrifugation such as compact, less capital intensive and high separation factor. If appropriately employed, these systems open up excellent opportunities for the separation of several isotopes other than Uranium as well.

In daily life, several radioisotopes are being used for medical applications in particular for cancer diagnosis and therapy³. While many of them are produced as fission products in nuclear reactors, some can be obtained by irradiation of their precursor isotopes. The medical fraternity desires to use nuclear medicine in highest possible radio-isotopic purity. This demand is normally not met for all the isotopes of nuclear medicine due to challenges in producing the precursor isotopes with a high level of isotopic purity (degree of enrichment) followed by full conversion into the medical isotopes upon irradiation. Among the several isotopes for nuclear medicine, ^{177}Lu isotope has gained lots of attention due to its application in targeted radionuclide therapy⁴ (TRT). However, currently, there is a global shortage of ^{177}Lu isotope primarily due to the shortage of its precursor which is enriched ^{176}Lu isotope. Therefore, there is a need to enhance the production of this isotope to meet the global demand.

Separation of ^{176}Lu (natural abundance = 2.59%) from the other natural major isotope ^{175}Lu (natural abundance = 97.41%) through gas centrifugation is not possible as it has no volatile compounds⁵. Seemingly, AVLIS remains as the only available option for the separation of ^{176}Lu . Kurchatov Institute⁵⁻¹⁰, Russia has extensively worked on the isotope separation of Lu isotopes. The experimental facilities required are technologically most demanding particularly with respect to the bandwidth of the excitation lasers which has to be limited to < 100 MHz level. Therefore, such

systems cannot be developed by the nations which are not technologically advanced, thus hampering large scale production of enriched ^{176}Lu for medical applications. Therefore, search for suitable photoionization schemes with less stringent requirements and ability to produce at higher production rates is currently on.

In the present work, a new photoionization pathway is proposed for the isotope separation of ^{176}Lu from natural Lu. The proposed photoionization scheme has been theoretically evaluated by computing ionization efficiency and isotope selectivity under various experimental conditions using density matrix formalism¹¹.

Results & Discussion.

The primary limitation in direct irradiation of natural Lu for the production of ^{177}Lu is the low natural abundance of its precursor ^{176}Lu (2.59 %). Upon direct irradiation in a nuclear reactor, the content ^{177}Lu varies between 0.3 – 1.2 % (Fig. 1a) in case of medium to high flux reactors, while irradiation of ^{176}Lu enriched to 50% produces ^{177}Lu whose content varies between 6 - 24 % (Fig. 1b). Further enrichment of ^{176}Lu does not result in significant improvement in ^{177}Lu content upon irradiation. For example, ^{176}Lu enriched to 100% produces ^{177}Lu whose content varies between 12 - 47 % (Fig. 1c). Therefore, enrichment of ^{176}Lu to the degree of ~ 50% can be considered as optimum.

Density matrix formalism provides most accurate description of the laser-atom interaction. Lineshapes arising in double-resonance¹² and triple resonance ionization¹³ of atoms have been investigated in detail. Recently, investigation of the following photoionization scheme has been carried out for the ionization efficiency and degree of enrichment of ^{177}Lu under various conditions.

540 – 535 Scheme.

The results obtained were in good agreement with the experimental results¹⁴. The optimum conditions for the isotope selective photoionization of ^{176}Lu have been derived. The primary limitation of the scheme for the production of enriched ^{176}Lu is the utilization of narrowband lasers

with bandwidth < 100 MHz. Moreover, incorporation of additional apertures along the atomic beam axis is required to limit the Doppler broadening of the atomic beam. This also causes reduction in the throughput of the enriched isotope.

From the energy levels of Lu, it is possible to formulate several photoionization pathways, among them, the following photoionization pathway seems to be particularly promising.

573 – 609 Scheme.

The photoionization pathway has a number of advantages over the photoionization scheme reported previously⁵⁻¹⁰. They are,

- 1) The wavelengths of the excitation lasers are easily accessible by high conversion efficiency Rhodamine dye lasers.
- 2) Large separation between hyperfine components of the Lu isotopes than previously reported
- 3) Possibility of employing relatively a broadband lasers for isotope separation

The hyperfine structure constants of Lu isotopes were measured by Brenner¹⁵ et al and Kuhnert¹⁶ et al for the 0.0 cm⁻¹ and 17427.28 cm⁻¹ levels. Vergès and Wyart¹⁷ have measured hyperfine structure constants of ¹⁷⁵Lu isotope for the 33831.46 cm⁻¹ level using Fourier Transform Infrared Spectrometry, whereas the same for the ¹⁷⁶Lu have not been reported so far. In the absence of hyperfine anomalies^{15,18}, the relationship between the hyperfine structure constants can be expressed as

$$\frac{A_{176}}{A_{175}} = \frac{\mu_{176} \times I_{175}}{\mu_{175} \times I_{176}} = \frac{3.1692 \times 3.5}{2.2325 \times 7} = 0.71 \quad (1)$$

$$\frac{B_{176}}{B_{175}} = \frac{Q_{176}}{Q_{175}} = \frac{4.92}{3.49} = 1.41 \quad (2)$$

Where A, B are the magnetic dipole and electric quadrupole constants; μ is the magnetic moment, I is the nuclear spin and Q is the Quadrupole moment. The hyperfine structure constants for the ¹⁷⁶Lu were calculated to be A = 1498.4 MHz and B = 2322.2 MHz for the 33831.46 cm⁻¹ level (Table 1).

Based on the spectroscopic selection rules, a total of 26 hyperfine pathways are possible in step wise excitation scheme each denoted by a serial number are tabulated in Table 2 along with their resonance frequencies. Among them, the hyperfine pathway 17/2-17/2-17/2 of ^{176}Lu (denoted by serial no 2) lies far away from the hyperfine pathways of the interfering ^{175}Lu isotope. Isotope separation of ^{176}Lu can be accomplished by ionization through 17/2-17/2-17/2 hyperfine pathway.

In an AVLIS process, when the first and second excitation lasers are tuned to the frequencies ν_1 and ν_2 respectively, isotope selectivity is defined as

$$\text{Isotope Selectivity } S(\nu_1, \nu_2) = \left(\frac{\eta_{176\text{Lu}}}{\eta_{175\text{Lu}}} \right) \quad (3)$$

Where η is the ionization efficiency of the isotope

For the case, wherein the abundance of the isotopes is not equal, one needs to normalize the selectivity with the abundance of the constituent isotopes; thus, isotope ratio enhancement factor is defined as

$$\text{Isotope Ratio Enhancement Factor IRE}(\nu_1, \nu_2) = \left(\frac{\eta_{176\text{Lu}}}{\eta_{175\text{Lu}}} \right) * \left(\frac{A_{176\text{Lu}}}{A_{175\text{Lu}}} \right) \quad (4)$$

Degree of enrichment after laser interaction is calculated using the expression

$$\text{Degree of enrichment (\%)} = \left\{ \frac{\eta_{176\text{Lu}} * A_{176\text{Lu}}}{(\eta_{175\text{Lu}} * A_{175\text{Lu}} + \eta_{176\text{Lu}} * A_{176\text{Lu}})} \right\} * 100 \quad (5)$$

Two dimensional contour plots of the ionization efficiency of Lu isotopes are plotted in Fig. 2. The resonance frequency positions of all the possible hyperfine pathways are numbered as per the Table 2. Horizontal ridges correspond to the first step excitation and vertical ridges correspond to the second step excitation. Strong horizontal and vertical ridges arise due to the strong transitions of the first and second excitation steps. Enhanced ionization observed at the intersection of horizontal and vertical ridges correspond either to the hyperfine pathway (numbered as per the Table 2) or crossovers. Origin of such crossovers has been discussed in detail in a recent article¹⁴. The resonance position of the 17/2-17/2-17/2 of ^{176}Lu has been shown as a “filled circle” in the ^{175}Lu two-dimensional contour plot in Fig. 2a.

The diagonal ridges correspond to the coherent two photon ionization pathway which is observed when the sum total energy of the two photons is equal to the energy of the upper level. In stepwise excitation schemes, coherent two photon ionization can be observed for the detuning (from the intermediate level) values matching the condition $\Delta_1 = -\Delta_2$. The probability of coherent two photon ionization decreases with the increase in the detuning ($|\Delta|$) of the laser frequency from its resonance transition. For the strong transitions, as in the present case, the diagonal ridge can be observed even for large detunings (Fig. 2).

Diagonal ridges corresponding to the coherent two photoionization of ^{175}Lu hyperfine pathways numbered as 2, 4, 7, 10 and 13 closely pass through the 17/2-17/2-17/2 hyperfine pathway of ^{176}Lu (Fig. 2a). The two photon sum frequency of 17/2-17/2-17/2 hyperfine pathway of ^{176}Lu is $-8198.1 \text{ MHz} + 22531.2 \text{ MHz} = 14333.1 \text{ MHz}$ (Table 2). The two photon sum frequencies of the hyperfine pathways of ^{175}Lu numbered 2, 4, 7, 10 and 13 ranges between 10532.6 MHz to 12929.3 MHz thus have significant impact on the degree of enrichment of the AVLIS process.

Computation of ionization efficiencies and isotope selectivity have been calculated varying the powers of excitation and ionization lasers for the Doppler free case and optimum conditions for the enrichment are tabulated in Table 3. From the Table 3, the limit for the bandwidth of the excitation lasers is found to be 500 MHz for obtaining the degree of enrichment $\sim 50 \%$ or higher.

Numerical computations of ionization efficiency and degree of enrichment have been carried out for various experimental parameters such as bandwidth of the excitation lasers, power of excitation and ionization lasers and half angular divergence limit to the atomic beam with a minimum limit to ionization efficiency was set to 0.2 and results are tabulated in Table 4. This corresponds to the ionization of 80% of population available at the ground hyperfine level 17/2. In order to compare the present results with the previously reported data^{5,14}, production rates have been calculated for the atomic number density of 5×10^{14} of Lu corresponding to the number density 1.3×10^{13} of ^{176}Lu . Further, the laser-atom interaction length is considered as 200 mm with a diameter of 10 mm. It can be immediately observed that for the case of lasers with bandwidth of 100 MHz, the degree of

enrichment reaches to > 98 %; while for the case of laser bandwidth of 250 MHz, the degree of enrichment reduces to > 85 % for any divergence of the atomic beam. The increase in the angular divergence has little impact on the degree of enrichment, which implies that under such conditions, larger production rates can be obtained without significant deterioration in the degree of enrichment. While in the case of lasers with bandwidth of 500 MHz, a degree of enrichment of ~ 49 % can be achieved up to the half angular divergence limit value of 20° . At higher half angular divergence values the degree of enrichment starts deteriorating. Therefore, for this case, half angular divergence of 20° can be taken as the optimum limit.

The present results are compared with the previously reported^{5,14} in Table 5. For the case of lasers with bandwidth of 100 MHz, even for the maximum possible half angular divergence (90°), the degree of enrichment > 98 %, which is nearly 3 times higher for the 573-609 nm scheme than the previously reported 540–535 nm scheme by D'yachkov⁵ et al. It is not possible to enrich the ^{176}Lu isotope > 5 % using broadband lasers having a width of 500 MHz through 540–535 scheme. However, lasers with bandwidth of 500 MHz can be used in the present 573 – 609 scheme and a degree of enrichment of ~ 49 % can be achieved with production rate > 12 mg / hour which is > 3 times higher than previously reported rate. Thus the proposed scheme enables production of ^{176}Lu with high production rate with relatively broad band lasers.

Effect of unknown parameters. In atomic vapor laser isotope separation process, all the atomic parameters such as isotope shifts, hyperfine structures, lifetimes, branching ratios, decay to trapped, autoionization states and their cross-sections; laser parameters such as power, pulse width, bandwidth, repetition frequency; source parameters as temperature, angular divergence all influence overall degree of enrichment and the production rate. In case of 573 – 609 scheme, some of the parameters have not been measured thus not known so far. The effect of these parameters on degree of enrichment and production rates is discussed below.

a) Isotope shift for the 609 nm transition. Isotope shift for the $5d6s6p\ ^4F_{3/2}^o(17427.28\text{ cm}^{-1}) \xrightarrow{609.6007\text{ nm}} 6s6p^2\ ^4P_{3/2}(33831.46\text{ cm}^{-1})$ transition has not been

reported so far. Isotope shift of a transition comprises of field shift and mass shift can be expressed as¹⁹

$$\delta\gamma \approx F \cdot \delta\langle r^2 \rangle^{175,176} + M \cdot \left(\frac{M^{175} - M^{176}}{M^{175} \cdot M^{176}} \right)$$

Where F, M are the field shift and mass shift parameters, $\delta\langle r^2 \rangle^{175,176}$ is the difference in the mean square nuclear charge radii.

In case of Lutetium isotopes, the difference in the mean square nuclear charge radii $\delta\langle r^2 \rangle^{175,176} = 0.041 \text{ fm}^2$ is small¹⁹, resulting in small field shift. The mass shift is also small owing to the small value of the factor $\left(\frac{M^{175} - M^{176}}{M^{175} \cdot M^{176}} \right)$. As a result, the isotope shift is small *i.e.*, about $\sim 100 \text{ MHz}$. Large isotope selectivity of the present scheme is due to the large spread of hyperfine spectrum of ^{176}Lu , therefore, the influence of the variation isotope shift is not of much significance.

b) Einstein's coefficient for the

$5d6s6p \ ^4F_{3/2}(17427.28 \text{ cm}^{-1}) \xrightarrow{609.6007 \text{ nm}} 6s6p^2 \ ^4P_{3/2}(33831.46 \text{ cm}^{-1})$ transition.

Einstein's A coefficient for the transition has not reported so far. However, the typical values for reported the fine structure 4P level were reported by in the range of 3×10^6 to 2×10^7 . A mean value of 1.3×10^7 has been taken for the computations. Due this uncertainty in the value of Einstein's A coefficient, the optimum power of the second excitation transition may vary between 26 – 97 W/cm^2 . Due to the large spread of the hyperfine spectrum ^{176}Lu and the separation of its 17/2-17/2-17/2 hyperfine pathway from the ^{175}Lu spectrum, the power broadening resulting in the variation of the power, varies the ionization efficiency rather than selectivity.

c) Autoionization states and cross-sections.

The availability of the data of autoionization states and their ionization cross-sections is the primary impediment to the proposed 573 – 609 scheme. Most of the work^{20,21} carried out corresponding to the Rydberg series and autoionization states originating from the $5d6s6p \ ^2D_{3/2,5/2}^o$, $5d6s6p \ ^4D_{3/2,5/2}^o$ and $6s^2nd \ ^2D_{3/2,5/2}$, $6s^2ns \ ^2S_{1/2}$. D'yachkov^{5,10} et al have found a strong autoionization level connecting

to the upper level $5d6s7s\ ^4D_{3/2}$ of the 540-535 scheme. Further work on search for suitable autoionization states connecting to the upper level $6s6p^2\ ^4P_{3/2}$ of the 573-609 scheme is necessary.

Conclusion. A new photoionization pathway has been proposed for the isotope separation of ^{176}Lu . The optimum conditions for the isotope separation of ^{176}Lu have been obtained through density matrix formalism. It has been shown that it is possible to obtain degree of enrichment $> 98\%$ with high production rates ($> 20\text{ mg / hour}$) when lasers with bandwidth of 100 MHz are employed. Even when broadband Rhodamine lasers of bandwidth of 500 MHz are used, the degree of enrichment $\sim 49\%$ can be obtained with a production rate of $> 12\text{ mg / hour}$. This significantly eases the restrictions on bandwidth on excitation lasers and Doppler broadening enabling high production rates than previously reported, thus expected to minimise the global shortage of the ^{176}Lu precursor for the production of the ^{177}Lu medical isotope.

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Table 1. HFS Coupling Constants of Lutetium energy levels

Energy Level	¹⁷⁵ Lu		¹⁷⁶ Lu		Reference
	A (MHz)	B (MHz)	A (MHz)	B (MHz)	
5d6s ²² D _{3/2} (0.0 cm ⁻¹)	194.331617	1511.398650	137.920537	2132.296936	[15]
5d6s6p ⁴ F _{3/2} ⁰ (17427.28 cm ⁻¹)	-924.82	1766.8	-651.47	2494.2	[16]
6s6p ²⁴ P _{3/2} (33831.46 cm ⁻¹)	70.33 [#] / 70.94 [#] / 70.80 [#] (Average: 70.69 [#]) 2119.2	54.35 [#] / 55.92 [#] / 54.39 [#] (Average: 54.89 [#]) 1645.5	1498.4 (Calculated)	2322.2 (Calculated)	[7]

- in mK

Table 2. A table of hyperfine excitation pathways of ^{175}Lu and ^{176}Lu of the photoionization. (The frequency scale is in reference with ^{176}Lu centre of gravity)

S No	^{175}Lu						^{176}Lu					
	F1-F2-F3	Frequency Position of the first hyperfine transition (MHz)	Normalised Intensity	Frequency Position of the second hyperfine transition (MHz)	Normalised Intensity	Sum two photon frequency position (MHz)	F1-F2-F3	Frequency Position of the first hyperfine transition (MHz)	Normalised Intensity	Frequency Position of the second hyperfine transition (MHz)	Normalised Intensity	Sum two photon frequency position (MHz)
1	5-5-4	-5418.3	100.0	4179.4	38.9	-1238.9	17/2-17/2-15/2	-8198.1	100.0	8384.4	49.1	186.3
2	5-5-5	-5418.3	100.0	15950.9	100.0	10532.6	17/2-17/2-17/2	-8198.1	100.0	22531.2	100.0	14333.1
3	4-5-4	-3367.1	38.9	4179.4	38.9	812.3	15/2-17/2-15/2	-5731.2	49.1	8384.4	49.1	2653.2
4	4-5-5	-3367.1	38.9	15950.9	100.0	12583.8	15/2-17/2-17/2	-5731.2	49.1	22531.2	100.0	16800
5	5-4-3	-2056.2	38.9	-7189.5	50.5	-9245.7	17/2-15/2-13/2	-4175.0	49.1	-6590.0	65.1	-10765
6	5-4-4	-2056.2	38.9	817.3	24.2	-1238.9	17/2-15/2-15/2	-4175.0	49.1	4361.3	18.3	186.3
7	5-4-5	-2056.2	38.9	12588.8	38.9	10532.6	17/2-15/2-17/2	-4175.0	49.1	18508.0	49.1	14333
8	4-4-3	-5.0	24.2	-7189.5	50.5	-7194.5	15/2-15/2-13/2	-1708.0	18.3	-6590.0	65.1	-8298
9	4-4-4	-5.0	24.2	817.3	24.2	812.3	15/2-15/2-15/2	-1708.0	18.3	4361.3	18.3	2653.3
10	4-4-5	-5.0	24.2	12588.8	38.9	12583.8	15/2-15/2-17/2	-1708.0	18.3	18508.0	49.1	16800
11	3-4-3	340.5	50.5	-7189.5	50.5	-6849	13/2-15/2-13/2	-937.2	65.1	-6590.0	65.1	-7527.2
12	3-4-4	340.5	50.5	817.3	24.2	1157.8	13/2-15/2-15/2	-937.2	65.1	4361.3	18.3	3424.1
13	3-4-5	340.5	50.5	12588.8	38.9	12929.3	13/2-15/2-17/2	-937.2	65.1	18508.0	49.1	17570.8
14	2-3-2	4048.0	37.9	-16575.9	37.9	-12527.9	15/2-13/2-11/2	3486.3	65.1	-20114.5	48.9	-16628.2
15	2-3-3	4048.0	37.9	-11393.5	0.0	-7345.5	15/2-13/2-13/2	3486.3	65.1	-11784.4	1.9	-8298.1
16	2-3-4	4048.0	37.9	-3386.7	50.5	661.3	15/2-13/2-15/2	3486.3	65.1	-833.1	65.1	2653.2
17	4-3-2	4199.1	50.5	-16575.9	37.9	-12376.8	11/2-13/2-11/2	3859.0	48.9	-20114.5	48.9	-16255.5
18	4-3-3	4199.1	50.5	-11393.5	0.0	-7194.4	11/2-13/2-13/2	3859.0	48.9	-11784.4	1.9	-7925.4
19	4-3-4	4199.1	50.5	-3386.7	50.5	812.4	11/2-13/2-15/2	3859.0	48.9	-833.1	65.1	3025.9
20	3-3-2	4544.6	0.0	-16575.9	37.9	-12031.3	13/2-13/2-11/2	4257.1	1.9	-20114.5	48.9	-15857.4
21	3-3-3	4544.6	0.0	-11393.5	0.0	-6848.9	13/2-13/2-13/2	4257.1	1.9	-11784.4	1.9	-7527.3
22	3-3-4	4544.6	0.0	-3386.7	50.5	1157.9	13/2-13/2-15/2	4257.1	1.9	-833.1	65.1	3424
23	2-2-2	8084.5	25.3	-20612.4	25.3	-12527.9	11/2-11/2-11/2	9607.9	50.5	-25863.4	50.5	-16255.5
24	2-2-3	8084.5	25.3	-15430.0	37.9	-7345.5	11/2-11/2-13/2	9607.9	50.5	-17533.3	48.9	-7925.4
25	3-2-2	8581.1	37.9	-20612.4	25.3	-12031.3	13/2-11/2-11/2	10006.0	48.9	-25863.4	50.5	-15857.4
26	3-2-3	8581.1	37.9	-15430.0	37.9	-6848.9	13/2-11/2-13/2	10006.0	48.9	-17533.3	48.9	-7527.3

Table 3. A table of Selectivity, Isotope Ratio Enhancement (IRE) factor and degree of enrichment of ^{176}Lu for the Doppler free case.

Laser Bandwidth (MHz)	Laser Power (W/cm^2)			η_{176}	Selectivity	IRE (natural Lu)	Degree of enrichment %
	573.8130 nm	609.0007 nm	Ionization laser				
100	160	60	28000	0.40	423.15	11.25	91.84
250	60	20	26000	0.200	345.26	9.18	90.18
500	100	40	19000	0.200	36.89	0.98	49.52

Table 4. A table of degree of enrichment and production rates of ^{176}Lu for various experimental parameters.

Laser bandwidth (MHz)	Half Angle Divergence	Laser Power (W/cm ²)			Co-propagating excitation laser beams					Counter-propagating excitation laser beams				
		573.8130 nm	609.0007 nm	Ionization laser	η_{176}	Selectivity	IRE (natural Lu)	Degree of enrichment %	Production Rate (mg / hour)	η_{176}	Selectivity	IRE (natural Lu)	Degree of enrichment %	Production Rate (mg / hour)
100	Doppler Free (0°)	40	20	16000	0.288	4095.6	108.90	99.1	-	0.288	4095.6	108.90	99.1	-
	5°	40	20	16000	0.285	4040.6	107.43	99.1	4.59	0.285	4039.1	107.39	99.1	4.59
	10°	40	20	16000	0.273	3876.7	103.08	99.0	8.72	0.274	3876.7	103.08	99.0	8.73
	20°	40	20	16000	0.237	3356.3	89.24	98.9	14.71	0.239	3371.3	89.64	98.9	14.82
	40°	40	20	16000	0.161	2219.6	59.02	98.3	19.91	0.164	2212.1	58.82	98.3	20.29
	90°	40	20	16000	0.150	2026.8	53.89	98.2	20.44	0.153	2046.6	54.42	98.2	20.86
250	Doppler Free (0°)	80	20	18000	0.213	302.1	8.03	88.9	-	0.213	302.1	8.03	88.9	-
	5°	80	20	18000	0.212	301.0	8.00	88.9	3.42	0.212	300.9	8.00	88.9	3.42
	10°	80	20	18000	0.210	297.7	7.92	88.8	6.69	0.210	297.3	7.91	88.8	6.69
	20°	80	20	18000	0.202	285.7	7.60	88.4	12.5	0.202	285.0	7.58	88.3	12.51
	40°	80	20	18000	0.172	240.2	6.39	86.5	21.29	0.174	235.8	6.27	86.3	21.46
	90°	80	20	18000	0.166	229.6	6.11	85.9	22.55	0.166	224.0	5.96	85.6	22.6
500	Doppler Free (0°)	100	40	22000	0.204	36.7	0.98	49.4	-	0.204	36.7	0.98	49.4	-
	5°	100	40	22000	0.204	36.6	0.97	49.3	3.29	0.204	36.6	0.97	49.3	3.29
	10°	100	40	22000	0.203	36.5	0.97	49.4	6.48	0.203	36.5	0.97	49.2	6.48
	20°	100	40	22000	0.201	36.1	0.96	49.0	12.47	0.201	36.0	0.96	48.9	12.47
	40°	100	40	22000	0.192	34.1	0.91	47.5	23.67	0.192	33.4	0.89	47.0	23.73
	90°	100	40	22000	0.189	33.5	0.89	47.1	25.7	0.189	32.9	0.87	46.6	25.75

Production rates are calculated for the atomic number density of 5×10^{14} of Lu.(Corresponding to the number density 1.3×10^{13} of ^{176}Lu); Length of the laser-atom interaction region 200 mm, laser beam diameter of 10 mm.

Table 5. Comparison of the photoionization schemes for the enrichment of ^{176}Lu

Bandwidth (MHz)	Half Angle Divergence (degrees)	540 nm – 535 nm Scheme				573 – 609 nm Scheme	
		D'yachkov ⁵ et al		Previous work ¹⁴		Present work	
		Production Rate (mg / hour)	Degree of Enrichment (%)	Production Rate (mg / hour)	Degree of Enrichment (%)	Production Rate (mg / hour)	Degree of Enrichment (%)
100	90 ⁰	22.55	30.7	22.55	33.8 – 34.7	20.86	98.2
	5 ⁰	3.7	68.4	3.5	66.6 – 66.8	4.59	99.1
250	90 ⁰	-	-	-	-	22.6	85.6
500	5 ⁰	-	-	-	-	3.29	49.3
	10 ⁰	-	-	-	-	6.48	49.2
	20 ⁰	-	-	-	-	12.47	48.9

Figure 1. Percentage abundance of ^{177}Lu after irradiation in reactor with different nuclear fluxes for the cases of natural Lu (a), 50 % enriched in ^{176}Lu (b) and 100% enriched in ^{176}Lu (c).

Figure 2. Two dimensional contour of the ionization efficiency of ^{175}Lu (a) and ^{176}Lu (b) for the Doppler free condition and laser power of 10 W/cm^2 . The resonance frequency positions of the hyperfine excitation pathway are numbered as per the Table 2. The resonance position of the 17/2-17/2 of the ^{176}Lu is shown as “filled circle” in figure 2a.

Figures

Figure 1

Percentage abundance of ^{177}Lu after irradiation in reactor with different nuclear fluxes for the cases of natural Lu (a), 50 % enriched in ^{176}Lu (b) and 100% enriched in ^{176}Lu (c).

Figure 2

Two dimensional contour of the ionization efficiency of ^{175}Lu (a) and ^{176}Lu (b) for the Doppler free condition and laser power of 10 W/cm². The resonance frequency positions of the hyperfine excitation pathway are numbered as per the Table 2. The resonance position of the $17/2-17/2-17/2$ of the ^{176}Lu is shown as “filled circle” in figure 2a.