Constraints on the $^{176}$Lu Cosmochronometer

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Received 1987 July 15; accepted 1987 December 12

Abstract. In an endeavour to resolve reported discrepancies in the value of the branching ratio of $^{176}$Lu at astrophysical energies, a new determination of the $^{175}$Lu ($n\gamma$) $^{176}$mLu capture cross section has been measured as $958 \pm 58$ mb. This gives a value of the branching ratio of $0.21 \pm 0.05$. This result indicates that some reequilibration of the ground and isomeric states of $^{176}$Lu occurs in stellar environments undergoing s-process nucleosynthesis, and confirms that $^{176}$Lu is not a reliable cosmochronometer. However the very existence of $^{176}$Lu in the solar system implies that the ground state of $^{176}$Lu was not completely depopulated, and provides the possibility of using this nuclide as a sensitive thermometer for stellar processes.

Key words: s-process nucleosynthesis—$^{176}$Lu cosmochronometer—branching ratio—nuclear reactions

1. Introduction

Neutron capture processes play an important role in astrophysics. The major processes involved in heavy element nucleosynthesis are the slow ($s$) and rapid ($r$) neutron capture processes (Burbidge et al. 1957), although the proton ($p$) capture process also plays a role. The site of s-process nucleosynthesis is believed to be the red giant phase of stars at temperatures of $2-4 \times 10^8$ K, corresponding to a Maxwellian temperature $kT=30$ keV. The nuclei are synthesized in a weak neutron flux over a relatively long period of time so that $\beta$-decay can occur between successive neutron captures. The r-process follows a path on the neutron-rich side of the valley of $\beta$-stability, successive neutron captures occurring before the resulting nuclide $\beta$-decays.

Of particular significance is the branch in the s-process path at $^{176}$Lu, because this nuclide is one of the few naturally-occurring radioactive species that has survived from the period of galactic nucleosynthesis. Audouze, Fowler & Schramm (1972) and Arnould (1973) independently suggested that $^{176}$Lu could be developed as a cosmochronometer, because the ground state $\beta$-decays to $^{176}$Hf with a half-life of $3.57 \times 10^{10}$ y. This unique s-process chronometer is therefore of potential value in determining the mean age of nucleosynthesis.

Figure 1 shows the mass region in the vicinity of lutetium, together with the neutron capture and proton capture processes involved in synthesising the various nuclides.
Figure 1. s-process path in the rare earth element mass region. s-only process nuclides $^{170}\text{Yb}$, $^{176}\text{Lu}$ and $^{176}\text{Hf}$ are shielded from r-process contributions by $^{170}\text{Er}$ and $^{176}\text{Yb}$ respectively. The s-process branches at $^{176}\text{Lu}$ if a significant population of the 3.68 h isomeric state occurs.


\[1^{76}\text{Lu} \text{ cosmochronometer}\]

\(^{176}\text{Lu}\) and \(^{176}\text{Hf}\) are shielded from the r-process by the isobaric nuclide \(^{176}\text{Yb}\) and are shown as cross-hatched squares in Fig. 1. The other numbered squares represent stable isotopes. The Lu/Hf chronometer provides the opportunity of using the well-established s-process systematics to determine the production ratios, rather than having to rely on theoretical models, as is the case for r-process cosmochronometers. The advantages of the \(^{176}\text{Lu}\) chronometer have been discussed by McCulloch, De Laeter & Rosman (1976) and Beer et al. (1981).

2. The \(^{176}\text{Lu}\) chronometer

Schrødamm & Wasserburg (1970) developed a nucleochronological formalism for long-lived radionuclides which enables the mean age of nucleosynthesis \(\Delta_{\text{max}}\), to be determined. Thus a long-lived nuclide, such as \(^{176}\text{Lu}\), is independent of the model of galactic evolution, since it effectively integrates any short-term irregularities which may occur.

The \(^{176}\text{Lu}\) chronometer is complicated in that the first excited state of the nucleus is a \(J^\pi = l^-\) level at 127 keV. This isomeric state \(^{176m}\text{Lu}\), \(\beta^-\) decays to \(^{176}\text{Hf}\) with a half-life of 3.68 h. Thus it is necessary to measure the branching ratio \(B\) which is defined as

\[
B = \frac{\langle \sigma_{175}^g \rangle}{\langle \sigma_{175} \rangle} = 1 - \frac{\langle \sigma_{175}^m \rangle}{\langle \sigma_{175} \rangle} = 1 - B^m
\]

(1)

where \(\langle \sigma_{175}^g \rangle\) and \(\langle \sigma_{175}^m \rangle\) are the 30keV \(^{175}\text{Lu}\) \((\gamma\gamma)\) cross-sections to \(^{176g}\text{Lu}\) and \(^{176m}\text{Lu}\) respectively, and \(\langle \sigma_{175} \rangle\) is the total 30 keV cross-section for \(^{175}\text{Lu}\) \((\gamma\gamma)\) \(^{176}\text{Lu}\). These cross-sections can be measured by irradiating Lu in a neutron flux obtained from the \(^7\text{Li}\) \((p, n)\) reaction for proton energies just above the reaction threshold.

Figure 2 shows the \(^{176}\text{Lu}^{\rightarrow}^{176}\text{Hf}\) decay scheme. Neutron capture on \(^{175}\text{Lu}\) leads to the population of two states—the ground state \(^{176g}\text{Lu}\) which decays to \(^{176}\text{Hf}\) with a long half-life, and the 127 keV isomeric state \(^{176m}\text{Lu}\) which decays with a half-life of 3.68 h.

The branching ratio can be determined by measuring \(\langle \sigma_{175}^m \rangle\) and \(\langle \sigma_{175} \rangle\) using neutrons covering an energy spectrum approximating that which would be found in red giant stars. Beer & Käppeler (1980) measured the capture cross-section of \(^{175}\text{Lu}\) to the isomeric state in \(^{176}\text{Lu}\) to be 809 ± 49 mb. In conjunction with the value Of \(\langle \sigma_{175} \rangle\) of 1411 ± 170 mb measured by Macklin & Gibbons (1967), this gave a branching ratio \(B = 0.43 \pm 0.05\). Allen, Lowenthal &De Laeter (1981) independently reported a value for \(B = 0.21 \pm 0.04\) based on a measurement of 1111 ± 64mb for \(\langle \sigma_{175}^m \rangle\).

A number of new determinations of \(\langle \sigma_{175} \rangle\) have now been made. Beer et al. (1981) obtained a value of 1266 ± 43 mb, and subsequently Beer et al. (1984) reported values of 1206 ± 54 mb and 1179 ± 44 mb which were determined at the Oak Ridge linear accelerator (ORELA) and the Karlsruhe 3MV pulsed Van de Graaf accelerator respectively. Since all three measurements agree within experimental errors, a mean value of 1217 mb can be adopted as the best value for \(\langle \sigma_{175} \rangle\). This value is in good agreement with earlier determinations of 1240 ± 190 mb (Lepine, Douglas & Maia 1972) and 1208 ± 60 mb (Macklin, Drake & Malanify 1978). If the revised value of 1217 mb is used to calculate \(B\), values of 0.34 ± 0.04 and 0.08 ±0.03 would be derived from the \(\langle \sigma_{175}^m \rangle\) values of Beer & Käppeler (1980), and Allen, Lowenthal & De Laeter (1981) respectively.
Figure 2. Decay scheme for neutron capture on $^{175}$Lu. The isomeric branching ratio $B^m$ is determined by measuring the 88 keV $\gamma$ yield from $^{176}$Hf.
In an endeavour to resolve the discrepancy between these two values for \( B \), we have repeated our measurement of \( \langle \sigma_{175}^{m} \rangle \). In the original experiment a Lu\(_2\)O\(_3\) powder was used. The powder was enclosed in thin aluminium foil, interleaved with gold foil and the Lu\(_2\)O\(_3\)/Au sandwich enclosed in a tight-fitting cadmium container to eliminate thermal neutrons. Although care was taken to ensure that the neutron exposures were as uniform as possible, X-ray analyses of the powder sources showed some evidence of aggregation and voiding.

Although essentially the same procedures were used in this experiment as those described by Allen, Lowenthal & De Laeter (1981), Lu metal foil replaced the Lu\(_2\)O\(_3\) powder. The Lu/Au sandwich was irradiated in a neutron flux with an energy range up to 105 keV, produced by the Li\((p, n)\) reaction in the Australian Atomic Energy Commission’s 3MV accelerator. The 12.7 mm diameter, 25–60 mg cm\(^{-2}\) gold foils were counted in a 4\(\pi\), \(\beta\)-\(\gamma\) coincidence chamber. The 88 keV \(\gamma\) rays from the \(^{176}\)Lu activation were measured in a well-calibrated Ge(Li) spectrometer at source-detector distances of 4.5 cm and 9.5 cm. Correction factors and uncertainties are the same as those reported by Allen, Lowenthal & De Laeter (1981). Four separate irradiations were made at a proton energy of 25 keV above the Li\((p, n)\) threshold. The flux-weighted ratio of the Lu isomer and Au capture cross-sections was found to be \( \langle \sigma_{175}^{m} \rangle / \langle \sigma_{197} \rangle = 1.654 \pm 0.076 \). A preliminary report of this result was given by Allen et al. (1981).

Using the data of Macklin, Halperin & Winters (1975) for the 30 keV Maxwellian average cross section \( (E_n=0.5–300\text{ keV}) \) of \( \langle \sigma_{197} \rangle_{kT=30} = 624 \pm 25\text{mb} \), the average cross-section at 30±10 keV is 579 ± 23 mb. The isomeric cross-section \( \langle \sigma_{175}^{m} \rangle_{kT-30} \pm 10 \) can be calculated to be 958 ±58 mb and the branching ratio is therefore \( B = 0.21 \pm 0.05 \) (using the revised value of 1217 mb for \( \langle \sigma_{175} \rangle \)). This value is in much closer agreement with the corresponding value of 0.34 ± 0.04 derived from the work of Beer & Käppeler (1980), than our previous value.

Audouze, Fowler & Schramm (1972) derived a simple formalism for determining the mean age of nucleosynthesis \( \Delta_{\text{max}} \), which can be expressed as

\[
\Delta_{\text{max}} = \frac{1}{\lambda_{176}} \ln \left[ \frac{B \langle N_i \sigma_{176} \rangle_{176}}{N_{176} \langle \sigma_{176} \rangle_{176}} \right]
\]

where \( \lambda_{176} \) is the decay constant of \(^{176}\)Lu; \( \langle N_i \sigma_{176} \rangle_{176} \) is the average value of the product of \( s \)-process abundance and 30 keV average capture cross-section evaluated at mass 176; \( N_{176} \) is the abundance of \(^{176}\)Lu at the time of formation of the solar system; \( \langle \sigma_{176} \rangle \) is the 30 keV \(^{176}\)Lu \((\eta\gamma)\) \(^{177}\)Lu cross section and \( B \) is the branching ratio.

One of the characteristic features of \( s \)-process systematics is that the product of the abundances of the \( s \)-process nuclides \( (N_i) \) and their corresponding capture cross-sections measured at astrophysical energies \( \langle \sigma \rangle \) is a smooth, slowly varying function of mass number \( A \), except for those regions where closed shell effects are dominant (Clayton et al. 1961). \(^{176}\)Lu is not in a region of this curve where nuclear shell effects occur, and experimental measurements have confirmed that the ‘local approximation’ \( N_i \sigma \) = constant, is valid over a limited mass region (Allen, Gibbons & Macklin 1971).

The data base for the distribution curve has been continually refined. New abundance data by Anders & Ebihara (1982) and Beer et al. (1984), and new capture cross-sections by Käppeler et al. (1982) have shown that the empirical data is in excellent
agreement with the traditional theoretical model, which assumes a steady neutron flux and an exponential distribution of neutron irradiations. Thus it is possible to estimate a value for \( \langle N_s \sigma \rangle_{176} \), based on the assumption that in this region \( N_s \sigma \) is a smooth, nearly constant function of mass number.

The values necessary to calculate the mean age of nucleosynthesis using Equation (2), are given in Table 1. A number of determinations for \( \lambda_{176} \), \( N_{176} \) and \( \langle \sigma_{176} \rangle \) have been reported, and these parameters no longer represent a limitation in calculating \( \Delta_{\text{max}} \), although their accuracy needs to be determined to about 1 per cent if a reliable age is to be achieved.

If the value of \( B = 0.34 \) is used, a mean age of \( 7.6 \times 10^9 \) y is derived. On the other hand if \( B = 0.21 \) is used, the logarithmic term is \( < 1 \) and \( \Delta_{\text{max}} \) cannot be calculated. It is therefore apparent that the limiting parameter in obtaining a good assessment of \( \Delta_{\text{max}} \) is the branching ratio.

The essential difference between the values of the branching ratio between this work and that of Beer & Käppeler (1980) arises from the experimental ratio of the energy-averaged neutron capture cross-sections of \(^{175m}\text{Lu}\) and \(^{197}\text{Au}\). Other factors relating to the choice of cross-section values and treatment of the neutron energy spectrum may differ, but are not the source of the disagreement. The cross-section ratio is

\[
\frac{\langle \sigma_{175m} \rangle}{\langle \sigma_{197} \rangle} = \frac{k_{176m}}{k_{198}} \frac{n_{197}}{n_{175}}
\]

where \( k \) and \( n \) are expressed per mg of target material in the area defined by the gold foil, and \( k \) is the end point abundance of the product nuclei. Our value for this ratio is \( 1.654 \pm 0.076 \), where the error represents the standard deviation of four separate measurements. Both the Karlsruhe measurements and those at Lucas Heights were measured at \( E_p = 25 \) keV above threshold. The difference between the two results may be in foil composition, detector calibrations or thickness corrections, but in the light of

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Reference</th>
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<tbody>
<tr>
<td>( \langle \sigma_{175} \rangle )</td>
<td>809 ± 49 mb</td>
<td>Beer &amp; Käppeler (1980)</td>
</tr>
<tr>
<td></td>
<td>958 ± 58 mb</td>
<td>This work</td>
</tr>
<tr>
<td>( \langle \sigma_{175} \rangle )</td>
<td>1266 ± 43 mb</td>
<td>Beer et al. (1981)</td>
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<td></td>
<td>1206 ± 54 mb</td>
<td>Beer et al. (1984)</td>
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<td></td>
<td>1179 ± 44 mb</td>
<td>Beer et al. (1984)</td>
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<tr>
<td>mean</td>
<td>1217 mb</td>
<td></td>
</tr>
<tr>
<td>( B )</td>
<td>0.34 ± 0.04 mb</td>
<td>Beer &amp; Käppeler (1980)</td>
</tr>
<tr>
<td></td>
<td>0.21 ± 0.05 mb</td>
<td>This work</td>
</tr>
<tr>
<td>( \lambda_{176} )</td>
<td>( 1.94 \times 10^{-11} ) y(^{-1} )</td>
<td>Patchett (1983)</td>
</tr>
<tr>
<td>( \langle N_s \sigma \rangle_{176} )</td>
<td>5.47 mb (Si = ( 10^6 ) atoms)</td>
<td>Beer (1983)</td>
</tr>
<tr>
<td>( N_{176} )</td>
<td>0.00106</td>
<td>Anders &amp; Ebihara (1982)</td>
</tr>
<tr>
<td>( \langle \sigma_{176} \rangle )</td>
<td>1526 ± 69 mb</td>
<td>Beer et al. (1984)</td>
</tr>
<tr>
<td></td>
<td>1514 ± 56 mb</td>
<td>Beer et al. (1984)</td>
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<td>mean</td>
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the discrepancy and the importance of the measurement to astrophysics, we would urge that another independent determination of the branching ratio of $^{176}\text{Lu}$ be undertaken as soon as possible.

3. Discussion

Allen, Lowenthal & De Laeter (1981) concluded that the negative value derived for the mean age of nucleosynthesis for $^{176}\text{Lu}$ could be explained on the basis that thermal equilibrium of the ground and isomeric states occurs in stellar interiors on a timescale less than the 3.68 h half-life of $^{176}\text{mLu}$. The application of the Schramm-Wasserburg formalism to $^{176}\text{Lu}$ assumes that the observed $s$-process branch at $^{176}\text{Lu}$ is determined solely by the isomeric branching ratio $B^m$. Allen, Lowenthal & De Laeter (1981) discussed a number of possible mechanisms to explain why this assumption must be invalid.

Clayton (1963, personal communication) was the first to suggest that thermalization of the $^{176}\text{Lu}$ ground and isomeric state populations may occur in the stellar environment. The effective half-life of $^{176}\text{Lu}$ is obtained from the Boltzmann equation and reduces to

$$t_\gamma = t_{1/2} \left[ \frac{2J_0 + 1}{2J_1 + 1} \right] \exp \left[ \frac{E_1 - E_0}{kT} \right] = 0.14 \text{ yr}$$

(3)

for $J_0 = 7$, $J_1 = 1$, $E_1 = 127$ keV, $t_{1/2} = 3.68$ h and $kT=30$ keV. The changed half-life results from the weak population of the isomeric state (0.3 per cent at 30 keV) in thermal equilibrium with the stellar environment.

The branching ratio for $\beta^-$ decay from $^{176}\text{Lu}$ depends on the effective $\beta^-$ half-life and the neutron flux. The observed branching ratio ($f_\gamma$) is obtained from the $s$-process local approximation model and can be defined as

$$f_\gamma = 1 - \frac{N_{176} \langle \sigma_{176} \rangle}{\langle N_\sigma \rangle_{176}}$$

(4)

$$= 0.69 \pm 0.07.$$  

This value is less than our isomeric branching ratio $B^m$ of 0.79 ± 0.09. Consequently, the population of the isomeric state must be reduced significantly in less than 3.68 h to reduce the isomeric decay rate. It is unlikely that this effect could be achieved directly by photo-de-excitation ($\Delta J = 6$), but photon inelastic scattering and Coulomb excitation processes could establish thermal equilibrium by electromagnetic linkage to higher excited states.

Shaw & Clayton (1967) examined the effect of particle-induced electromagnetic de-excitation of nuclei in stellar matter, whilst Ward (1981) found that enhanced decay rates could occur from Coulomb collisions with the ion plasma in the stellar environment. From analytic approximations the resulting de-excitation rates are strongly dependent on the mass and charge numbers of the target and projectile, stellar temperature, multipole order and type, and energy of the transition. For $^{176}\text{Lu}$, Ward (1981) estimates that enhancement could occur above $4 \times 10^8$ K for helium burning and above $10^9$ K for carbon burning. Coulomb enhancement of indirect
transitions, linked through higher-lying states, may prove to be more significant than
direct transitions in determining the thermal equilibrium rate.

Watanabe, Mukoyama & Katano (1981) have examined the implications of nuclear
excitation of $^{176}\text{Lu}$ by positron annihilation by irradiating Lu with positrons from
$^{64}\text{Cu}$. Characteristic 88.35 keV $\gamma$ rays from $^{176m}\text{Lu}$ were observed, which enabled
the cross-section for the annihilation-excitation process to be established at $9.0 \pm
3.2 \times 10^{-22}$ cm$^2$. More recently Norman et al. (1985) have examined the effects
of photoexcitation and positron annihilation-excitation of $^{176g}\text{Lu}$ to $^{176m}\text{Lu}$ and found
that these two processes alone are capable of establishing thermal equilibrium
between these two states at temperatures $>3.5 \times 10^8$ K. Norman et al. (1985) point
out that a number of other processes (such as Coulomb excitation and inelastic
neutron scattering) will tend to produce equilibration at temperatures below
$3.5 \times 10^8$ K.

It is therefore apparent that the final abundance of $^{176g}\text{Lu}$ which survives the s-
process stellar environment will be a very sensitive function of the thermal history that
it has experienced. A detailed treatment of the temperature dependence of the Lu
branching ratio is given by Beer et al. (1981). Using theoretical estimates for the $\gamma$-
deexcitation branching ratios of a number of known energy levels these authors found
evidence for the effective reduction of the $^{176}\text{Lu}$ half-life for thermal energies above
16keV. Consequently the $^{176}\text{Lu}$ chronometer can only keep time for temperatures
somewhat lower than those envisaged for the s-process.

Allen, Lowenthal & De Laeter (1981), on the basis of competition between $\beta^-$ decay
and neutron capture rates, calculated that the neutron flux must be in the range
$2 \times 10^{15}$ to $10^{17}$ cm$^{-2}$ s$^{-1}$. However if thermal equilibrium between the ground and
isomeric state is maintained after the neutron source is exhausted, all of the $^{176}\text{Lu}$ will
decay to $^{176}\text{Hf}$ in a year or so. The temperature dependence of the neutron flux and
the $^{176}\text{Lu}$ effective half-life could then result in a ‘freeze-out’ of abundances on the
termination of the neutron exposure. Thus the observed $f_\gamma$ value may not result from
the branching ratio $B_m$ nor from the effects of thermal equilibrium, but from the
freezing out of abundances when the s-process neutron exposure terminates.

4. Conclusions

Although considerable progress has been made in elucidating the various parameters
involved in the $^{176}\text{Lu}$ chronometer since Audouze, Fowler & Schramm (1972) first
proposed it as a unique s-process chronometer, uncertainty still exists with respect to
the correct value for the branching ratio of $^{176}\text{Lu}$. The value of $0.21 \pm 0.05$ reported in
this paper is at variance with an earlier value of $0.34 \pm 0.04$ by Beer & Käppeler (1980),
and there is therefore an urgent need for other investigators to redetermine this
parameter, which is of crucial importance in deciphering the $^{176}\text{Lu}/^{176}\text{Hf}$ nuclear
system.

The implication of the lower value for the branching ratio is that significant
equilibration has occurred between the ground and isomeric states in $^{176}\text{Lu}$. A survey
of possible mechanisms such as particle-induced electromagnetic de-excitation,
Coulomb collisions with the ion plasma, photo-excitation and positron annihilation-
excitation imply that the final abundance of the ground state of $^{176}\text{Lu}$ which survives
the s-process environment in stellar interiors, is a sensitive function of the thermal
history that it has experienced. Our conclusion is that although $^{176}$Lu is not a reliable $s$-process cosmochronometer, it can reveal details of the dynamics of the $s$-process. However in order to delineate details of its time and temperature dependence, a more accurate measurement of the isomeric capture cross-section, and a detailed understanding of intra-nuclear excitation and decay processes are required.

Acknowledgements

This work has been supported by the Australian Research Grants Scheme and the Australian Institute of Nuclear Science and Engineering. Mrs P. R. Harris typed the manuscript with care and patience.

References