Abundances for *p*-process nucleosynthesis

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(Received 13 December 2007; revised manuscript received 10 March 2008; published 23 April 2008)

An important constraint in developing models of *p*-process nucleosynthesis is that the abundances of many of the *p*-process nuclides are not well known. A recent review of the *p*-process has identified six *p*-process nuclides that are of particular significance to *p*-process theorists [M. Arnould and S. Goriely, Phys. Rep. **384**, 1 (2003)]. These nuclides are ^{92,94}Mo, ^{96,98}Ru, ^{138}La, and ^{180}Ta^{m}. The absence of accurate abundances for these isotopes is due to the fact that the isotopic composition of the elements concerned have not been corrected for isotope fractionation induced by the thermal ionization mass spectrometric instruments used to measure them. To remedy this deficiency, a VG 354 mass spectrometer was calibrated using gravimetric mixtures of enriched isotopes to enable the absolute isotopic compositions of these elements to be obtained. Although the isotopic abundances of ^{92,94}Mo, ^{138}La, and ^{180}Ta^{m} have previously been reported, the absolute abundances of ^{96,98}Ru are reported for the first time in this article, with a significant reduction in the magnitude of the values as compared to existing abundances.

DOI: 10.1103/PhysRevC.77.045803 PACS number(s): 98.80.Ft, 26.20.−f, 26.30.−k, 27.70.+q

I. INTRODUCTION

The landmark article by Burbidge *et al.* [1], provided the basic elements of nuclear astrophysics, in that it demonstrated how the isotopes of the chemical elements could be synthesized in stars. The basic processes involved in synthesizing the isotopes of the "heavy" elements ($Z > 28$) are the slow (s)- and the rapid®-neutron capture processes and the *p*-process, the latter being largely responsible for synthesizing the relatively rare, neutron-deficient isotopes that lie on the proton-rich side of the valley of nucleosynthesis. There are 35 nuclides traditionally classified as *p*-process isotopes, yet the total abundance of these 35 nuclides amount to only 0.1 to 0.01 of the corresponding *s*- and *r*-process produced nuclides. A coherent theory of *p*-process nucleosynthesis has been a controversial topic since the proposal that *p*-process nuclides were produced in the H-rich layers of Type II supernovae, where (p, γ) and (γ, n) reactions on *s*- and *r*-process seed nuclei occurred [1]. However, Rayet *et al.* [2] argued that the *p*-process nuclides were formed in the O/Ne layers of Type II supernovae, while Wallerstein *et al.* [3] suggested that *p*-process nucleosynthesis took place in a high temperature, proton-rich environment, both by proton capture on light seed nuclei and by photonuclear reactions on heavy, stable nuclides.

A recent review of the *p*-process of heavy element nucleosynthesis has highlighted the difficulties involved in formulating a comprehensive model of the *p*-process that can explain the observed abundance distribution [4]. In particular, existing models have a significant underproduction problem in the *p*-process isotopes of Mo and Ru. Furthermore, the rare, odd-odd *p*-process nuclides ^{138}La and ^{180}Ta^{m}, which are also key nuclides in *p*-process nucleosynthesis, are underproduced in existing *p*-process nucleosynthetic models. A prerequisite for testing models of *p*-process nucleosynthesis is an accurate set of abundances for these isotopes. Although the abundances of the *p*-process nuclides ^{92,94}Mo [5], ^{138}La [6], and ^{180}Ta^{m} [7] are now based on calibrated isotopic composition measurements, the absolute isotopic composition of Ru has not previously been determined, and hence accurate values for the abundances of ^{96,98}Ru have not been available until now. The absolute isotopic composition of many polyisotopic elements have not been determined, so that the abundances of many of the *p*-process nuclides are not accurately known. Thermal Ionization Mass Spectrometry (TIMS) is the method of choice for determining the absolute isotopic composition of the *p*-process elements. The sample is loaded in the solid form in the ion source of a TIMS and evaporated by heating the filament on which the sample has been loaded. In the process of evaporation and ionization, Rayleigh distillation causes an isotope fractionation effect, which produces an enhancement in the measured abundance of the "light" isotopes with respect to the "heavy" isotopes. Thus the abundances of the "light" isotopes are overestimated in uncalibrated mass spectrometric measurements. Because the *p*-process nuclides are always the isotopes with the lowest mass in an element, it follows that *p*-process abundances, as listed in existing tables [1,8], are overestimated for noncalibrated polyisotopic elements.

In recognition of the fact that most *p*-process nuclides were not known with optimum accuracy, and in response to the review by Arnould and Goriely [4] that ^{92,94}Mo, ^{96,98}Ru, ^{138}La, and ^{180}Ta^{m} were key nuclides in *p*-process theories, this laboratory embarked on a mass spectrometric program to measure the absolute isotopic composition of these elements. We have recently measured the absolute isotopic composition of Mo [5], La [6], Yb [9], and Ta [7] to enable accurate abundances of five *p*-process nuclides to be determined. The laboratory standards of these elements were obtained from Johnson-Matthey Chemicals Ltd. The purity of these elements are given by the Johnson-Matthey Chemicals and are reported in individual articles [5–7,9]. We now report the absolute isotopic composition of Ru, which enables the isotope

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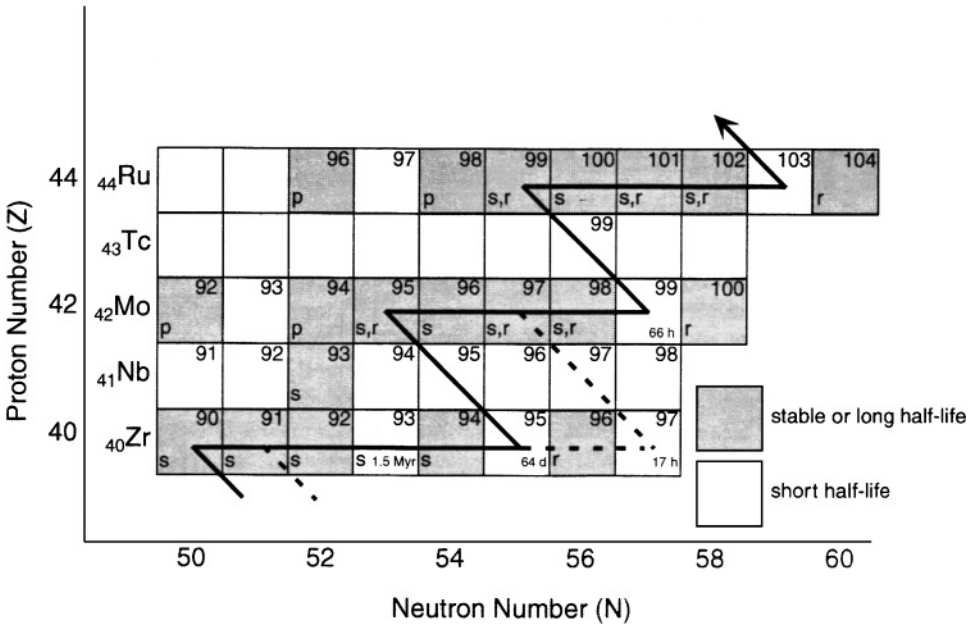

FIG. 1. The chart of the nuclides in the mass region of Mo and Ru showing the isotope abundances and the nucleosynthetic origin of the various isotopes of Mo and Ru. The "zig-zag" *s*-process neutron-capture path is shown, together with the four *p*-process isotopes ^{92,94}Mo and ^{96,98}Ru that are bypassed by neutron-capture processes.

abundances of ^{96,98}Ru to be determined with greater accuracy than the presently accepted values.

II. RESULTS AND DISCUSSION

Molybdenum and Ru each possess two *p*-process isotopes that constitute a significant fraction of the elemental abundance of these two elements. These four isotopes are underproduced in existing theories of *p*-process nucleosynthesis [4]. To determine the absolute isotopic composition of Mo and Ru, it is necessary to determine the magnitude of isotope fractionation in this mass region of the Periodic Table. A VG 354 mass spectrometer was used to measure the absolute isotopic composition of Mo by calibrating the mass spectrometer with gravimetric mixtures of two highly enriched isotopes —^{92}Mo and ^{98}Mo. Details of this experiment are described in Ref. [5]. A fractionation correction factor of 0.35% per mass unit was derived in this experiment. This enabled the isotope abundances of the two *p*-process isotopes ^{92,94}Mo to be determined to be 14.5246 ± 0.0015 and $9.1514 \pm; 0.0074\%$, respectively, as shown in Table I.

The absolute isotopic composition of Ru has not been experimentally determined, but because it is very close to the Mo mass region and is similar to Mo both in terms of the number of isotopes and the relative mass range (see Fig. 1), it is possible to use the fractionation correction factor measured for Mo [5] to determine the absolute isotopic composition of Ru and, hence, obtain the abundances of ^{96}Ru and ^{98}Ru. The presently accepted, but noncalibrated, isotope abundances of Ru as reported in Ref. [10] have been corrected by an isotope fractionation factor of 0.35% per mass unit, as measured in Ref. [5]. The net result is to decrease the abundances of the lighter isotopes of Ru and correspondingly enhance the abundances of the heavier isotopes. The absolute isotope abundances of Ru and its associated atomic weight $A_r = 101.080 \pm 0.001$ are listed in Table I. The isotope abundances of ^{96,98}Ru are 5.4423 ± 0.0002 and $1.8483 \pm 0.003\%$, respectively. The net effect of correcting the Ru isotopic composition for isotope fractionation is to decrease the abundances of ^{96,98}Ru by 3.1 and 3.9%, respectively. This reduction is consistent with the reduction in the abundances of the other *p*-process nuclides listed in Tables I and II, as a result of correcting for isotope fractionation effects.

To bring together the new isotopic data for *p*-process elements as recently determined in this laboratory, the absolute isotopic compositions and atomic weights of La, Yb, and Ta are listed in Table II. This enables the isotopic abundances of ^{138}La, ^{168}Yb, and ^{180}Ta^{m} to be determined. It should be noted that the new abundance values for the two odd-odd nuclei, ^{138}La and ^{180}Ta^{m}, may enable limits to be placed on neutrino oscillations [11]. The role of neutrino nucleosynthesis in the production of ^{138}La and ^{180}Ta^{m}, using newly derived input parameters to calculate the production of these nuclides, has recently been examined [12].

TABLE I. The absolute isotope abundances (in %) and atomic weights $\mathbf{A}_r$ of Mo and Ru as determined by calibrating a VG 354 mass spectrometer by gravimetric mixtures of highly enriched isotopes ^{92,98}Mo to obtain an isotope fractionation correction factor of 0.35% per mass unit.

	Molybdenum [5]	Ruthenium	(This work)
92	14.5246(15)	96	5.4423(2)
94	9.1514(74)	98	1.8483(3)
95	15.8375(98)	99	12.6619(7)
96	16.672(19)	100	12 5476 (5)
97	9.5991(73)	101	17.0505(11)
98	24.391(18)	102	31.6443(12)
100	9.824(50)	104	18.8053(12)
$\mathbf{A}_r$	**95.9602 (23)**		**101.080 (1)**

TABLE II. The absolute isotope abundances (in %) and atomic weights $\mathbf{A_r}$ of La, Yb, and Ta as determined by calibrating a VG 354 TIMS by gravimetric mixtures of enriched isotopes.

Isotopes of La [6]		Isotopes of ytterbium [9]		Isotopes of Ta [7]	
138	0.000888 (20)	168	0.1233 (4)	180	0.0001201 (8)
139	0.999112 (20)	170	2.982 (6)	181	0.9998799 (8)
		171	14.086 (20)		
		172	21.686 (19)		
		173	16.103 (9)		
		174	32.025 (12)		
		176	12.995 (13)		
$\mathbf{A_r}$	**138.905461 (30)**		**173.054 (1)**		**180.947878 (20)**

III. CONCLUSIONS

The observational foundation of nucleosynthesis is dependent on the determination of the elemental and isotopic abundances in a variety of cosmic objects. Despite advances in astronomical observations, the Solar System still provides much of the available abundance data. Despite the success of existing models of p-process nucleosynthesis based on SN 11 O-Ne layers in reproducing the abundances of the p-process nuclides [1], there are some major deficiencies, namely, the gross overabundances of the Mo and Ru p-process nuclides in nature and the under-production of ^{138}La and ^{180}Ta^{m}. The production of these two rare nuclides could potentially place limits on neutrino spectra and neutrino oscillations [12].

Of the absolute isotopic compositions of the five elements reported in this article, four—Mo, La, Yb, and Ta—have recently been classified by the Commission on Isotopic Abundances and Atomic Weights (CIAAW) as the best isotopic measurements from a single terrestrial source. CIAAW is the authorative source within the International Union of Pure and Applied Chemistry (IUPAC) for publishing the Table of the Isotopic Composition of the Elements [13]. The isotope abundances of the seven p-process nuclides reported in this article have been determined from measurements of the absolute isotope compositions of these elements using the same VG 354 TIMS, which has been calibrated for various elements to enable the fractionations associated with the Rayleigh distillation effect to be corrected. This new data will enable models of p-process nucleosynthesis to be tested against well-established isotope abundances in the mass region $92 \leqslant A \leqslant 180$.

Arnould and Goriely [4] have drawn attention to the necessity of more accurate astrophysical data for these key p-process nuclides. The effect of using absolute isotope abundances for ^{92,94}Mo and ^{96,98}Ru has resulted in an average reduction of 4.3% in the abundances of these four nuclides, which is in the direction required by existing p-process theory.

ACKNOWLEDGMENTS

The author acknowledges the help and advice of Professor K. Rosman, Mr. G. Burton, and an unknown referee for his suggestions. The Mass Spectrometry Laboratory at Curtin University is supported by the Australian Research Council and the Government of Western Australia through its Centre of Excellence Program.

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