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Impurities in a ^{28}Si -enriched Single Crystal Produced for the Realisation of the Redefined Kilogram

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ABSTRACT: The practical realization of the unit of mass is possible by manufacturing a perfect one-kilogram sphere from a ^{28}Si -enriched single crystal. The mass of the sphere can be determined in terms of a fixed value of the Planck constant by counting the number of silicon atoms in the core of the single crystal. To reach the target 2.0×10^{-8} relative standard uncertainty, the mass of the surface layer and the mass deficit due to point defects such as impurities and vacancies must be investigated and corrected for. A sample of a ^{28}Si -enriched single crystal produced to test the possibility of obtaining material at a scale useful to the dissemination of mass standards was measured by instrumental neutron activation analysis to check the purity with respect to a large number of possible contaminant elements. The results collected in a neutron activation experiment performed with the high thermal neutron flux available at the 20 MW OPAL research reactor are described. The data collected in this study showed that the produced material has a purity level never achieved with silicon used to manufacture previous one-kilogram spheres.

The x-ray-crystal-density (XRCD) method is one of the possible ways to realize the unit of mass in terms of a fixed value of the Planck constant, h .¹

In fact, since this method succeeded in determining the Avogadro constant, N_A , with 2.0×10^{-8} relative standard uncertainty,² it can be reversely used to prepare a ^{28}Si -enriched kilogram artifact in terms of a fixed h value, linked to N_A with negligible uncertainty via the molar Planck constant, $N_A h$.

Recently, three silicon single crystal ingots were manufactured in the framework of a project financed by the Physikalisch-Technische Bundesanstalt (PTB) and aimed to demonstrate the possibility of production of material on the quantity and with the quality required for realizing and disseminating mass standards.

The high ^{28}Si -enrichment and the nonexistence of point defects play a key role to achieve the 2.0×10^{-8} relative standard uncertainty required for the realization of the kilogram.

The isotopic composition of the new material has been investigated starting from the gas phase by Isotope Ratio Mass Spectrometry (IRMS) to the ^{28}Si poly-crystal by Multi-Collector Inductively Coupled Plasma Mass Spectrometry (MC-ICP-MS).³ In addition, an independent measurement of the ^{30}Si mole fraction in the ^{28}Si single crystal has been performed using Instrumental Neutron Activation Analysis (INAA).⁴ The measured 10^{-6} mol mol⁻¹ and 10^{-7} mol mol⁻¹ levels for the ^{29}Si and ^{30}Si mole fractions are confirming the expected high enrichment of the produced material.

With respect to point defects, previous Avogadro silicon materials were measured using positron lifetime and copper decoration to determine vacancies.^{5,6} In addition, infrared spectroscopy was used to quantify carbon, oxygen, boron and nitrogen,⁵ and INAA to probe a large number of the remaining elements.^{7,8,9}

With the aim of collecting experimental data on the contamination of the new material, a sample of one of the ^{28}Si -enriched single crystal ingots was allocated to be investigated by INAA.

As a continuation of the collaboration between the Istituto Nazionale di Ricerca Metrologica (INRIM) and the Australian Nuclear Science and Technology Organisation (ANSTO), we report here the result of a neutron activation experiment concerning sixty-four elements of the periodic table.

Instrumental Neutron Activation Analysis. INAA has been frequently adopted in semiconductor technology to detect contaminant elements. Silicon is an excellent material for trace element analysis via neutron activation and detection of delayed γ -emissions of the produced radionuclides because the interference due to the activation of the matrix is limited to somewhat short-lived radionuclides, i.e. ^{28}Al , ^{29}Al , ^{31}Si , and in negligible amounts, to ^{24}Na and ^{28}Mg .

In this study, the k_0 -method of standardisation was applied to determine the largest possible number of elements using a mono-elemental standard. A full description of the fundamental principles of the k_0 -method can be found elsewhere,¹⁰ while the measurement model we adopted to estimate the mass fraction of a target isotope is the equation 4 reported in a earlier work together with details on the experimental parameters.⁹

EXPERIMENTAL SECTION

The neutron irradiation and γ -spectrometry measurements were performed at the ANSTO laboratory to take advantage of the high thermal neutron flux available at the 20 MW OPAL research reactor.

Materials. The sample measured in this study was cut by PTB and coded Si28-23Pr11 part Q.4.1. The dimensions, 20 mm diameter and 19 mm length, and mass, 14 g, were established according to the maximum sample size and mass allowed for the irradiation. An external layer, about 70 μm thickness and 0.5 g mass, was removed by deep etching (10:1 of nitric acid, HNO_3 , assay 67-69%, and hydrofluoric acid, HF, assay 47-51%) to remove the surface contamination due to machining.

One sample, 70 mm length and 38 mg mass, of Al-0.1%Co wire (IRMM-527R, 0.5 mm diameter) and three samples, 2 mm length and 4 mg mass, of Al-0.1% Au wire (IRMM-530R, 1 mm diameter) were used as Co and Au mono-elemental standards for the k_0 -method, respectively.

High purity Al foils (Sigma-Aldrich #356859, 50 μm thickness) and polyethylene (PE) vials were selected to envelop and contain the sample during the irradiation.

The tools used to handle the sample were cleaned in an ultrasonic bath with 10% nitric acid. The water was purified using a Millipore system ($\rho > 18 \text{ M}\Omega \text{ cm}$) and the etching chemicals were of ultrapure grade.

The mass of the silicon sample and of the mono-elemental standards were measured with a digital balance calibrated with SI-traceable weights.

Neutron irradiation. The experiment concerned short and long-lived radionuclides produced in two separated irradiations, hereafter called short and long-term irradiation, lasting 300.0(6) s and 116.869(5) h, performed in the NAA-SRT facility (nominal thermal neutron flux, $\Phi_{\text{th}} = 2.42 \times 10^{13} \text{ cm}^{-2} \text{ s}^{-1}$, thermal to epithermal neutron flux ratio, $f = 3250$, $\alpha = 0.15$) and in the LE6-1C channel ($\Phi_{\text{th}} = 6.05 \times 10^{13} \text{ cm}^{-2} \text{ s}^{-1}$, $f = 262.6$, $\alpha = 0.143$), respectively.

The silicon sample was sealed in a PE vial and surrounded by the three Al-0.1% Au wires located at 120° with respect to the axis during the short-term irradiation, while it was enveloped in the Al foils with the Al-0.1%Co wire spiral wrapped around during the long-term irradiation.

γ -Spectrometry Measurements. The γ -spectrometry measurements of the silicon sample were performed with a high purity germanium (HPGe) detector, ORTEC GEM35-70-PL (58 mm crystal diameter, 39 % relative efficiency, 1.69 keV FWHM resolution at 1332 keV) inside a low-background graded lead shield. The sample was located with its axis vertical and as close as possible (i.e. at about 10 mm) to the end-cap of the detector.

Two and six γ -spectra were sequentially collected after the short and long-term irradiation, respectively. To eliminate the effect of any possible external contamination during handling, the sample was slightly etched after each irradiation.

The analysis of first two γ -spectra following the long-term irradiation showed (unexpected) significant amounts of Co and Ni.

Table 1. Quantified or detection limit mass fractions, w_E , and number densities, N_E , of the impurity elements in the ^{28}Si -enriched sample determined in the neutron activation experiment. The neutron capture reaction, the half-life of the produced radionuclide, the natural mole fraction of the target isotope, the detected γ -peak and the irradiation term from which the result was obtained are also stated. Data collected from the first two γ -spectra following the long-term irradiation are marked with *. The detection limits are evaluated according to Currie's method and the uncertainties ($k = 1$) in parentheses apply to the last respective digits.

| Reaction | $t_{1/2}^{15}$ | $x(^E)^{15}$ | γ -peak ¹⁵ / keV | Irradiation | $w_E / \text{g g}^{-1}$ | N_E / cm^{-3} |
|--|----------------|--------------|------------------------------------|-------------|----------------------------|---------------------------|
| $^{23}\text{Na}(n,\gamma)^{24}\text{Na}$ | 14.96 h | 1.000 | 2754 | Short | $\leq 9.7 \times 10^{-12}$ | $\leq 5.9 \times 10^{11}$ |
| $^{26}\text{Mg}(n,\gamma)^{27}\text{Mg}$ | 9.462 min | 0.11 | 843.8 | Short | $\leq 3.0 \times 10^{-7}$ | $\leq 1.7 \times 10^{16}$ |
| $^{36}\text{S}(n,\gamma)^{37}\text{S}$ | 5.05 min | 0.0002 | 3103.4 | Short | $\leq 7.0 \times 10^{-3}$ | $\leq 3.1 \times 10^{20}$ |

Thus, a deep etching was repeated before starting the collection of the third γ -spectrum.

The 1332.5 keV ^{60}Co γ -emission of the Al-0.1%Co wire was measured with the ORTEC GEM35-70-PL at 350 mm from the end-cap while the 411.8 keV ^{198}Au γ -emission of the three Al-0.1% Au wires was measured with an ORTEC GEM25P4-PLUS (59 mm crystal diameter, 32 % relative efficiency, 1.67 keV FWHM resolution at 1332 keV) at 216 mm from the end-cap.

The detectors were connected to ORTEC DSPEC-Pro digital spectrometers, and the data were collected using a personal computer running the ORTEC Maestro software.¹¹

RESULTS AND DISCUSSION

The number of counts, $N_{p,\gamma}$ corresponding to the area of the full-energy γ -peak or to detection limit according to the Currie's method¹² was evaluated from the recorded γ -spectra using the Hyperlab software.¹³

The output peak table files were input into the Kayzero for Windows software¹⁴ to estimate the mass fraction, w_{iE} , of the target isotope, iE . In the case of a non-quantified element, the lowest of the detection limits obtained with different γ -spectra was taken as a final result.

The effects due to neutron self-shielding and burn-up during irradiation and pulse losses, self-absorption and geometry during γ -spectrometry measurements were evaluated and, where necessary, corrected for.

Amounts of ^{50}Cr , ^{59}Co , ^{63}Cu , ^{71}Ga , ^{81}Br , ^{139}La , ^{186}W and ^{197}Au were quantified from γ -spectra collected after the long-term irradiation while amount of ^{75}As from γ -spectra collected after the short-term irradiation.

It must be noticed that the hypothesis of external contamination during the long-term irradiation was confirmed by a significant decrease of ^{59}Co and the disappearance of ^{64}Ni after the deep etching. Thus, since ^{63}Cu , ^{71}Ga , ^{81}Br , ^{139}La , ^{186}W were quantified with the first two γ -spectra following the long-term irradiation, the concentration of these elements in the crystal might be significantly lower.

Counting statistics were the main contributors to the uncertainty of the results. In addition, a ± 1 mm positioning tolerance of the silicon sample was assigned and gave a contribution of about 4% to the combined relative uncertainty.

A summary of the data obtained in this study is given in table 1 under the reasonable assumption of natural isotopic abundance of the impurities. The mass fraction, w_E , and number density, N_E , are reported with the neutron capture reaction, the half-life of the produced radionuclide, the natural mole fraction of the target isotope and the detected γ -peak.

| | | | | | | |
|---|-----------|----------|--------|-------|------------------------------|-----------------------------|
| $^{37}\text{Cl}(n,\gamma)^{38}\text{Cl}$ | 37.24 min | 0.2423 | 2167.4 | Short | $\leq 2.2 \times 10^{-10}$ | $\leq 8.8 \times 10^{12}$ |
| $^{40}\text{Ar}(n,\gamma)^{41}\text{Ar}$ | 1.822 h | 0.996 | 1293.6 | Short | $\leq 8.9 \times 10^{-12}$ | $\leq 3.1 \times 10^{11}$ |
| $^{41}\text{K}(n,\gamma)^{42}\text{K}$ | 12.36 h | 0.0673 | 1524.7 | Short | $\leq 1.6 \times 10^{-11}$ | $\leq 5.6 \times 10^{11}$ |
| $^{46}\text{Ca}(n,\gamma)^{47}\text{Sc}$ | 3.349 d | 0.000035 | 159.4 | Long | $\leq 6.3 \times 10^{-10}$ | $\leq 2.2 \times 10^{13}$ |
| $^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$ | 83.83 d | 1.0000 | 889.3 | Long | $\leq 2.5 \times 10^{-15}$ | $\leq 7.9 \times 10^7$ |
| $^{50}\text{Ti}(n,\gamma)^{51}\text{Ti}$ | 5.76 min | 0.052 | 320.1 | Short | $\leq 4.5 \times 10^{-8}$ | $\leq 1.3 \times 10^{15}$ |
| $^{51}\text{V}(n,\gamma)^{52}\text{V}$ | 3.75 min | 0.9975 | 1434.1 | Short | $\leq 1.8 \times 10^{-9}$ | $\leq 5.0 \times 10^{13}$ |
| $^{50}\text{Cr}(n,\gamma)^{51}\text{Cr}$ | 27.7 d | 0.0435 | 320.1 | Long | $1.40(14) \times 10^{-12}$ | $3.76(38) \times 10^{10}$ |
| $^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$ | 2.579 h | 1.0000 | 846.8 | Short | $\leq 7.1 \times 10^{-13}$ | $\leq 1.8 \times 10^{10}$ |
| $^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$ | 44.5 d | 0.0028 | 1099.3 | Long | $\leq 2.6 \times 10^{-11}$ | $\leq 6.4 \times 10^{11}$ |
| $^{59}\text{Co}(n,\gamma)^{60}\text{Co}$ | 1925.3 d | 1.0000 | 1332.5 | Long | $7.6(1.5) \times 10^{-14}$ | $1.81(34) \times 10^9$ |
| $^{64}\text{Ni}(n,\gamma)^{65}\text{Ni}$ | 2.517 h | 0.0091 | 1115.5 | Short | $\leq 2.8 \times 10^{-9}$ | $\leq 6.6 \times 10^{13}$ |
| $^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$ | 12.7 h | 0.6917 | 1345.8 | Long | $6.9(1.2) \times 10^{-10} *$ | $1.52(27) \times 10^{13} *$ |
| $^{64}\text{Zn}(n,\gamma)^{65}\text{Zn}$ | 244.3 d | 0.486 | 1115.5 | Long | $\leq 1.4 \times 10^{-12}$ | $\leq 3.0 \times 10^{10}$ |
| $^{71}\text{Ga}(n,\gamma)^{72}\text{Ga}$ | 14.1 h | 0.399 | 834 | Long | $3.1(1.0) \times 10^{-13} *$ | $6.2(2.0) \times 10^9 *$ |
| $^{76}\text{Ge}(n,\gamma)^{77}\text{Ge}$ | 11.3 h | 0.078 | 264.4 | Long | $\leq 3.2 \times 10^{-10} *$ | $\leq 6.2 \times 10^{12} *$ |
| $^{75}\text{As}(n,\gamma)^{76}\text{As}$ | 26.24 h | 1.0000 | 559.2 | Short | $1.28(47) \times 10^{-12}$ | $2.39(87) \times 10^{10}$ |
| $^{74}\text{Se}(n,\gamma)^{75}\text{Se}$ | 119.781 d | 0.0089 | 136 | Long | $\leq 2.6 \times 10^{-13}$ | $\leq 4.6 \times 10^9$ |
| $^{81}\text{Br}(n,\gamma)^{82}\text{Br}$ | 35.3 h | 0.4931 | 554.3 | Long | $1.80(30) \times 10^{-13} *$ | $3.15(52) \times 10^9 *$ |
| $^{85}\text{Rb}(n,\gamma)^{86}\text{Rb}$ | 18.63 d | 0.7217 | 1077 | Long | $\leq 1.7 \times 10^{-12}$ | $\leq 2.8 \times 10^{10}$ |
| $^{84}\text{Sr}(n,\gamma)^{85}\text{Sr}$ | 64.84 d | 0.0056 | 514 | Long | $\leq 1.8 \times 10^{-11}$ | $\leq 2.9 \times 10^{11}$ |
| $^{89}\text{Y}(n,\gamma)^{90m}\text{Y}$ | 3.19 h | 1.0000 | 479.5 | Short | $\leq 1.5 \times 10^{-8}$ | $\leq 2.3 \times 10^{14}$ |
| $^{94}\text{Zr}(n,\gamma)^{95}\text{Zr}$ | 62.02 d | 0.1728 | 756.7 | Long | $\leq 1.9 \times 10^{-11}$ | $\leq 2.9 \times 10^{11}$ |
| $^{93}\text{Nb}(n,\gamma)^{94m}\text{Nb}$ | 6.26 min | 1.0000 | 871 | Short | $\leq 2.3 \times 10^{-5}$ | $\leq 3.5 \times 10^{17}$ |
| $^{98}\text{Mo}(n,\gamma)^{99m}\text{Tc}$ | 6.01 h | 0.2413 | 140.5 | Long | $\leq 5.2 \times 10^{-12}$ | $\leq 7.6 \times 10^{10}$ |
| $^{102}\text{Ru}(n,\gamma)^{103}\text{Ru}$ | 39.35 d | 0.316 | 497.1 | Long | $\leq 1.8 \times 10^{-13}$ | $\leq 2.4 \times 10^9$ |
| $^{108}\text{Pd}(n,\gamma)^{109}\text{Pd}$ | 13.36 h | 0.2646 | 309.1 | Long | $\leq 8.0 \times 10^{-12} *$ | $\leq 1.1 \times 10^{11} *$ |
| $^{109}\text{Ag}(n,\gamma)^{110m}\text{Ag}$ | 249.8 d | 0.4817 | 657.8 | Long | $\leq 2.2 \times 10^{-13}$ | $\leq 2.8 \times 10^9$ |
| $^{114}\text{Cd}(n,\gamma)^{115}\text{Cd}$ | 53.46 h | 0.2872 | 527.9 | Long | $\leq 1.2 \times 10^{-12} *$ | $\leq 1.5 \times 10^{10} *$ |
| $^{115}\text{In}(n,\gamma)^{116}\text{In}$ | 54.41 min | 0.957 | 1293.5 | Short | $\leq 1.8 \times 10^{-13}$ | $\leq 2.2 \times 10^9$ |
| $^{116}\text{Sn}(n,\gamma)^{117m}\text{Sn}$ | 13.6 d | 0.147 | 158.5 | Long | $\leq 3.5 \times 10^{-11}$ | $\leq 4.2 \times 10^{11}$ |
| $^{123}\text{Sb}(n,\gamma)^{124}\text{Sb}$ | 60.2 d | 0.427 | 602.7 | Long | $\leq 6.3 \times 10^{-14}$ | $\leq 7.2 \times 10^8$ |
| $^{130}\text{Te}(n,\gamma)^{131}\text{I}$ | 8.021 d | 0.338 | 364.5 | Long | $\leq 9.9 \times 10^{-13}$ | $\leq 1.1 \times 10^{10}$ |
| $^{127}\text{I}(n,\gamma)^{128}\text{I}$ | 24.99 min | 1.0000 | 442.9 | Short | $\leq 9.5 \times 10^{-11}$ | $\leq 1.1 \times 10^{12}$ |
| $^{133}\text{Cs}(n,\gamma)^{134}\text{Cs}$ | 754.2 d | 1.0000 | 604.7 | Long | $\leq 4.1 \times 10^{-14}$ | $\leq 4.4 \times 10^8$ |
| $^{130}\text{Ba}(n,\gamma)^{131}\text{Ba}$ | 11.5 d | 0.00106 | 496.3 | Long | $\leq 1.5 \times 10^{-11} *$ | $\leq 1.5 \times 10^{11} *$ |
| $^{139}\text{La}(n,\gamma)^{140}\text{La}$ | 1.678 d | 0.9991 | 1596.2 | Long | $7.14(71) \times 10^{-14} *$ | $7.18(71) \times 10^8 *$ |
| $^{140}\text{Ce}(n,\gamma)^{141}\text{Ce}$ | 32.51 d | 0.8848 | 145.4 | Long | $\leq 1.7 \times 10^{-13}$ | $\leq 1.7 \times 10^9$ |
| $^{141}\text{Pr}(n,\gamma)^{142}\text{Pr}$ | 19.12 h | 1.0000 | 1575.6 | Long | $\leq 7.3 \times 10^{-13} *$ | $\leq 7.2 \times 10^9 *$ |
| $^{146}\text{Nd}(n,\gamma)^{147}\text{Nd}$ | 10.98 d | 0.1719 | 91.1 | Long | $\leq 6.5 \times 10^{-13}$ | $\leq 6.3 \times 10^9$ |
| $^{152}\text{Sm}(n,\gamma)^{153}\text{Sm}$ | 46.5 h | 0.266 | 103.2 | Long | $\leq 5.7 \times 10^{-15} *$ | $\leq 5.3 \times 10^7 *$ |
| $^{151}\text{Eu}(n,\gamma)^{152}\text{Eu}$ | 4945.4 d | 0.4786 | 121.8 | Long | $\leq 5.1 \times 10^{-15}$ | $\leq 4.7 \times 10^7$ |
| $^{152}\text{Gd}(n,\gamma)^{153}\text{Gd}$ | 240.4 d | 0.002 | 97.4 | Long | $\leq 5.8 \times 10^{-13}$ | $\leq 5.2 \times 10^9$ |
| $^{159}\text{Tb}(n,\gamma)^{160}\text{Tb}$ | 72.3 d | 1.0000 | 298.6 | Long | $\leq 1.8 \times 10^{-14}$ | $\leq 1.6 \times 10^8$ |
| $^{164}\text{Dy}(n,\gamma)^{165}\text{Dy}$ | 2.334 h | 0.281 | 94.7 | Short | $\leq 1.3 \times 10^{-12}$ | $\leq 1.2 \times 10^{10}$ |
| $^{165}\text{Ho}(n,\gamma)^{166}\text{Ho}$ | 26.83 h | 1.0000 | 80.6 | Long | $\leq 6.1 \times 10^{-14} *$ | $\leq 5.2 \times 10^8 *$ |
| $^{170}\text{Er}(n,\gamma)^{171}\text{Er}$ | 7.516 h | 0.149 | 308.3 | Long | $\leq 1.1 \times 10^{-11} *$ | $\leq 9.3 \times 10^{10} *$ |
| $^{169}\text{Tm}(n,\gamma)^{170}\text{Tm}$ | 128.6 d | 1.0000 | 84.3 | Long | $\leq 5.8 \times 10^{-15}$ | $\leq 4.8 \times 10^7$ |
| $^{174}\text{Yb}(n,\gamma)^{175}\text{Yb}$ | 4.185 d | 0.3183 | 396.3 | Long | $\leq 2.6 \times 10^{-14} *$ | $\leq 2.1 \times 10^8 *$ |
| $^{176}\text{Lu}(n,\gamma)^{177}\text{Lu}$ | 6.73 d | 0.0259 | 208.4 | Long | $\leq 1.3 \times 10^{-14}$ | $\leq 1.1 \times 10^8$ |
| $^{180}\text{Hf}(n,\gamma)^{181}\text{Hf}$ | 42.39 d | 0.3522 | 482.2 | Long | $\leq 2.9 \times 10^{-14}$ | $\leq 2.3 \times 10^8$ |
| $^{181}\text{Ta}(n,\gamma)^{182}\text{Ta}$ | 114.4 d | 0.9999 | 67.8 | Long | $\leq 3.6 \times 10^{-14}$ | $\leq 2.8 \times 10^8$ |
| $^{186}\text{W}(n,\gamma)^{187}\text{W}$ | 23.72 h | 0.2864 | 685.7 | Long | $6.87(42) \times 10^{-12} *$ | $5.22(32) \times 10^{10} *$ |
| $^{185}\text{Re}(n,\gamma)^{186}\text{Re}$ | 3.718 d | 0.374 | 137.2 | Long | $\leq 2.7 \times 10^{-14} *$ | $\leq 2.0 \times 10^8 *$ |
| $^{190}\text{Os}(n,\gamma)^{191}\text{Os}$ | 15.4 d | 0.264 | 129.4 | Long | $\leq 5.2 \times 10^{-14}$ | $\leq 3.8 \times 10^8$ |
| $^{191}\text{Ir}(n,\gamma)^{192}\text{Ir}$ | 73.827 d | 0.373 | 316.5 | Long | $\leq 4.5 \times 10^{-16}$ | $\leq 3.3 \times 10^6$ |
| $^{198}\text{Pt}(n,\gamma)^{199}\text{Au}$ | 3.139 d | 0.072 | 158.4 | Long | $\leq 1.2 \times 10^{-12} *$ | $\leq 8.5 \times 10^9 *$ |
| $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ | 2.695 d | 1.0000 | 411.8 | Long | $3.02(38) \times 10^{-14}$ | $2.14(27) \times 10^8$ |
| $^{202}\text{Hg}(n,\gamma)^{203}\text{Hg}$ | 46.61 d | 0.297 | 279.2 | Long | $\leq 9.4 \times 10^{-14}$ | $\leq 6.5 \times 10^8$ |
| $^{232}\text{Th}(n,\gamma)^{233}\text{Pa}$ | 26.97 d | 1.0000 | 311.9 | Long | $\leq 3.1 \times 10^{-14}$ | $\leq 1.8 \times 10^8$ |
| $^{238}\text{U}(n,\gamma)^{239}\text{Np}$ | 2.357 d | 0.9928 | 106.1 | Long | $\leq 1.6 \times 10^{-13} *$ | $\leq 9.5 \times 10^8 *$ |

CONCLUSIONS

The neutron activation analysis of the new ^{28}Si -enriched single crystal material manufactured to test the technical feasibility of silicon mass standards dissemination did not show the presence of bulk impurities with the exception of ultra-traces of Cr, Co, Cu, Ga, As, Br, La, W and Au. Specifically, the Cu mass fraction was $6.9(1.2) \times 10^{-10} \text{ g g}^{-1}$; combined uncertainty ($k = 1$) in parentheses applies to the last respective digits. Moreover, the Cr, Co, Ga, As, Br, La, W and Au mass fractions were within the range $10^{-14} \text{ g g}^{-1}$ and $10^{-12} \text{ g g}^{-1}$. Under the hypothesis that all contaminant atoms are interstitial point defects of the lattice crystal, the corresponding mass deficit² of a one-kilogram sphere made using the new silicon material would be $-0.70(12) \mu\text{g}$.

No contamination of Na, Ar, Sc, Mn, Zn, Se, Mo, Ru, Pd, Ag, Cd, In, Sb, Te, Cs, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Tm, Yb, Lu, Hf, Ta, Re, Os, Ir, Pt, Hg, Th, and U was found to within $1.0 \times 10^{-11} \text{ g g}^{-1}$; among them, Ir showed an outstanding value of $4.5 \times 10^{-16} \text{ g g}^{-1}$. Detection limits between $1.0 \times 10^{-11} \text{ g g}^{-1}$ and $1.0 \times 10^{-7} \text{ g g}^{-1}$ were achieved for Cl, K, Ca, Ti, V, Fe, Ni, Ge, Rb, Sr, Y, Zr, Sn, I, Ba, Er. Lastly, the worst detection limits were obtained for Mg, S and Nb, i.e. $3.0 \times 10^{-7} \text{ g g}^{-1}$, $7.0 \times 10^{-3} \text{ g g}^{-1}$ and $2.3 \times 10^{-5} \text{ g g}^{-1}$, respectively.

In summary, the results obtained in this study point out that the purity level of the new material is even higher than that of the material used for the latest determination of the Avogadro constant, called AVO28.⁹ In details, with the exception of W, reduced amounts of Cr, Co, Cu, Ga, As, Br, La, Au and no traces of Sc, Mn, Fe, Zn, Sb, Ir were detected. For many of the remaining elements, the analysis achieved detection limits of one order of magnitude lower. The particularly high ^{28}Si -enrichment reached during the production might have contributed to the purification.

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Notes

The authors declare no competing financial interest.

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24 Cr quantification
42 Mo detection limit