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Short Communication

Insights into the origin of purely sediment-derived Himalayan leucogranites: Si–O isotopic constraints

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Granite is the dominant rock type in Earth's continental crust. The origin of granite can be directly or indirectly related to the fractional crystallization of mantle-derived basaltic melt or the reworking of pre-existing continental or oceanic crust, which contribute to the growth of continental crust. Among the various types of granites, the peraluminous leucogranites in the Himalayan orogen, which are high in SiO_2 (>73%) and low in mafic minerals (<5%), are thought to originate from the melting of pure-sediments, as inferred from comprehensive petrological, geochemical, and isotopic studies [1–3]. Formation of the Himalayan leucogranites led to the reworking of the upper continental crust by the transfer of heat and volatile elements, and shaped the highest mountains on Earth (Fig. 1). In addition, Himalayan leucogranites are economically very important [5]. Some of these leucogranites were generated from multiple crustal sources, with or without input of mantle-derived melts. Extensive crystal fractionation of the granitic magmas resulted in rare metal mineralization [4]. These observations indicate the importance of the source compositions of the purely sediment-derived Himalayan leucogranites, which remain poorly known.

Isotopic compositions are commonly used as a "fingerprint" to trace the source of granites. Traditionally, radiogenic Sr, Nd, and Pb isotopes, and stable O isotope are analyzed for granite bulk-rock samples. This strategy provides a "snapshot" of the average composition of the component mineral phases, but does not provide clues to the generation of the magma of the original composition of the crystallized melt, and is susceptible to post-magmatic alteration and weathering. The advance of modern isotopic microanalysis techniques using SIMS and LA–MC/Q–ICPMS has enabled the integrated in situ measurement of zircon U–Pb ages and Hf–O isotopes [6]. Application of these novel techniques improves our understanding of the origin and genesis of granite [7]; however, problems arise when using zircons from high-Si leucogranite, which typically have high U contents that cause the metamictization of zircon crystals due to radiation damage [8].

Silicon (Si) and oxygen (O), the two most abundant elements in the silicate Earth, constitute >85 wt% of leucogranites. Unlike high-U zircon, for which the isotopic composition is susceptible to hydrothermal alteration, quartz (SiO_2) is stable and retains the Si and O isotope ratios of the magma from which it crystallized. Thus, quartz Si and O isotopes are the most reliable isotopic fingerprint to trace the origin of leucogranites. Igneous-derived high-Si granites that have experienced extensive fractionation of mafic minerals, are enriched in ^{30}Si, as mafic minerals are rich in the lighter Si isotopes [9]. Such granites plot at the high-$\delta^{30}Si$ end of the "Igneous Array" [10]. In contrast, sediment-derived high-Si granites are low in ^{30}Si, as the clay minerals that form during weathering preferentially incorporate the lighter Si isotopes. Oxygen isotopes can be used to trace sedimentary components within granites, as sedimentary rocks are enriched in ^{18}O compared with the igneous protoliths.

To gain a better understanding of the origin of the Himalayan leucogranites, we performed high-precision quartz Si–O isotope analyses of leucogranite from the Kampa Dome of the Himalayan orogen for the first time (Fig. 1). Garnet two-mica leucogranites, which are the major lithology in the Kampa pluton, were emplaced at ~26 Ma [11]. These are characteristically high in SiO_2 (74.7 wt%–76.6 wt%). Si isotopes in quartz were measured by MC–ICPMS at the CAS Key Laboratory of Crust-Mantle Materials and Environments, University of Science and Technology of China. Quartz O isotope microanalysis was performed using a Cameca-1280 SIMS at the Institute of Geology and Geophysics, Chinese Academy of Sciences. The external precision of MC–ICPMS and SIMS measurements of Si and O isotopes was ±0.05‰ and ±0.4‰, respectively. Six quartz samples from the Kampa leucogranites yield $\delta^{30}Si_{quartz}$

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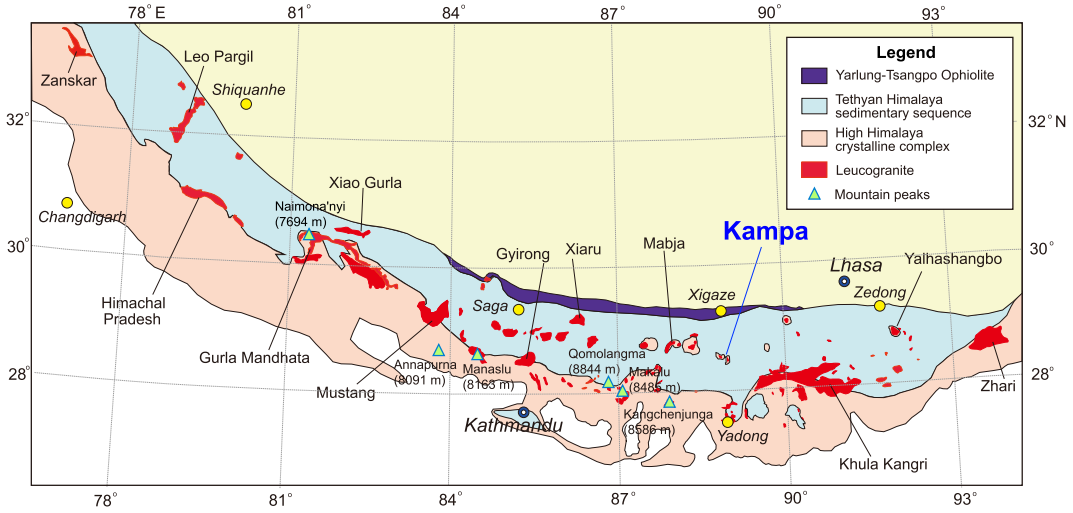

Fig. 1. Simplified geological map of the Himalayan orogen and the distribution of the Cenozoic leucogranites (modify after Wu et al. [4]).

values of −0.17‰ to −0.28‰ and $\delta^{18}O_{quartz}$ values of 11.25‰–12.28‰ (Fig. S1 online). The magnitude of Si isotope fractionation between the quartz and granitic melt ($\Delta^{30}Si_{quartz\text{-}melt}$) is 0.05‰ at ~700 °C [9]. Thus, the whole-rock $\delta_{30}Si$ value for the Kampa leucogranites can be calculated according to this fractionation factor (Table 1). Figure 2 shows a $\delta^{30}Si_{W.R}$ vs. SiO_2 diagram for the Kampa leucogranites in comparison with Si isotopes for other igneous rocks worldwide. The Si isotope data of the Kampa leucogranites are consistent with those of anatectic leucogranites and peraluminous cordierite-bearing S-type granites, but clearly lower than those of the "Igneous Array" for a given SiO_2 value. This indicates that the Kampa leucogranites were derived mainly from sedimentary rocks. Notably, the $\delta^{30}Si_{W.R.}$ values for the Kampa samples are systematically higher than those for typical anatectic peraluminous granitoids (Fig. 2). This suggests that the sources of the Kampa leucogranites were enriched in feldspar relative to other anatectic peraluminous granitoids.

The Kampa leucogranites are homogeneous in O isotopes, with measured $\delta^{18}O_{quartz}$ values falling within a range of ~1‰. For a crystallization temperature of 700 °C, the $\delta^{18}O_{zircon}$ and $\delta^{18}O_{W.R.}$ values for the Kampa leucogranites are 8.5‰–9.7‰ and 10.6‰–11.7‰, respectively, based on measured $\delta^{18}O_{quartz}$ values and equilibrium fractionation between quartz, zircon, and granitic melt [13]. These values differ from those reported for other Himalayan leucogranites [1,14]. Early whole-rock O-isotope analyses yielded $\delta^{18}O_{W.R.}$ values ranging from ~9‰ to ~13‰ for the Himalayan leucogranites [1,14]. While $\delta^{18}O_{W.R.}$ values exceeding 10‰ are typical of S-type granites, values lower than 10‰ overlap with those of the igneous-derived I-type granites. Such a large variation in $\delta^{18}O_{W.R.}$ is attributed to post-magmatic alteration. Hopkinson et al. [14] reported a wide range of $\delta^{18}O_{zircon}$ values (6.9‰–12.5‰) for leucogranites from the Bhutan Himalaya. Seven of 12 samples yielded $\delta^{18}O_{zircon}$ values above 2‰. These highly

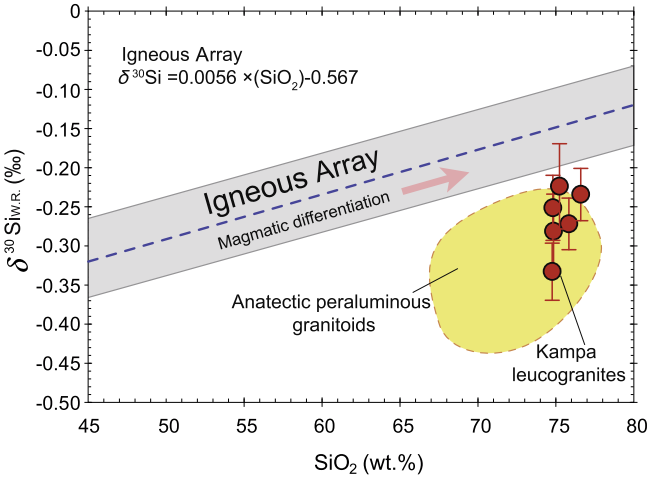

Fig. 2. $\delta^{30}Si_{W.R.}$ vs. whole-rock SiO_2 variation diagram for the Kampa leucogranites. The Si-isotope "Igneous Array" is defined by Savage et al. [10]. The yellow field of anatectic peraluminous granitoids of sedimentary origin is after Savage et al. [9] and Poitrasson [12].

variable $\delta^{18}O_{zircon}$ values most likely resulted from the "high-U effect" [8]. Our work reinforces that quartz is the most reliable of O isotope tracer for highly evolved granites such as leucogranites and alkaline granites.

Our integrated analysis of quartz Si–O isotopes indicates that the Kampa leucogranites were generated by the melting of pure sediments without appreciable mantle- or igneous-derived melts. Despite the limited range of measured quartz Si and O isotopes, the Kampa leucogranites still show detectable variations in Si (range of ~0.1‰) and O (~1‰) isotopes, with quartz Si and O

Table 1
Whole-rock SiO_2 and quartz Si–O isotope data for the Kampa leucogranites.

Sample	SiO_2 (%)	$\delta^{18}O_{quartz}$ (‰)	±2S.D.	$\delta^{30}Si_{quartz}$ (‰)	±2S.D.	$\delta^{30}Si_{W.R.}$ (‰)[#]
13 KB01	74.83	12.28	0.50	−0.23	0.05	−0.28
13 KB03	74.74	12.41	0.52	−0.28	0.04	−0.33
GBY02	74.79	11.78	0.42	−0.20	0.04	−0.25
GBY37	75.81	11.67	0.45	−0.22	0.03	−0.27
GBY39	75.21	11.32	0.42	−0.17	0.05	−0.22
GBY41	76.57	11.25	0.49	−0.19	0.03	−0.24

[#] $\delta^{30}Si_{W.R.} = \delta^{30}Si_{quartz} - 0.05$ (‰) (after Savage et al. [9]).

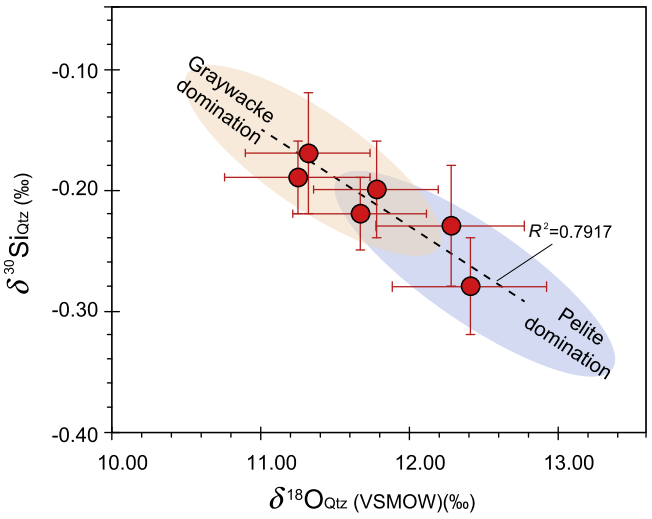

Fig. 3. Quartz $\delta^{18}O$ vs. $\delta^{30}Si$ variation diagram for the Kampa leucogranites.

isotopes displaying a strong negative correlation (Fig. 3). Two possible mechanisms can explain this correlation: (1) the fractional crystallization of granitic magma, and (2) the inheritance of a granite source. Liu et al. [11] reported that the formation of the Kampa leucogranites involved varying degrees of fractional crystallization of K-feldspar and other minerals. Si isotope fractionation between K-feldspar and a granitic melt ($\Delta^{30}Si_{K\text{-feldspar-melt}}$) is 0.02‰–0.06‰ [10]; thus, fractionation of K-feldspar has a small influence on the Si isotopes of the residual melts, as even extensive K-feldspar fractionation results in only a slight decrease in $\delta^{30}Si$ with increasing SiO_2. This subtle relationship between $\delta^{30}Si$ and SiO_2 is not observed for the Kampa leucogranites (Fig. 2), indicating that fractional crystallization of K-feldspar was not the major cause of the observed co-variation of Si–O isotopes. Thus, the measured quartz Si and O isotopes in the Kampa leucogranites mostly likely reflect their sources. The strong correlation between Si and O isotopes indicates the contribution of two end-members in their sedimentary sources [15]: one characterized by low ^{30}Si ($\delta^{30}Si_{W.R.} \leq -0.35‰$) and high ^{18}O ($\delta^{18}O_{W.R.} \geq 12‰$) values, indicating a pelite-dominated sediment, and one characterized by relatively high ^{30}Si ($\delta^{30}Si_{W.R.} \geq -0.25‰$) and low ^{18}O ($\delta^{18}O_{W.R.} \leq 10.5‰$) values, suggesting a feldspar-rich sediment such as metagraywackes.

In summary, our new Si–O isotope data reinforce the view that the Kampa leucogranites were generated by the melting of pure sedimentary rocks. The negative correlation between Si and O isotopes indicates that the "pure sediment" protolith comprised two main lithologies.

Conflict of interest

The authors declare that they have no conflict of interest.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at https://doi.org/10.1016/j.scib.2018.09.001.

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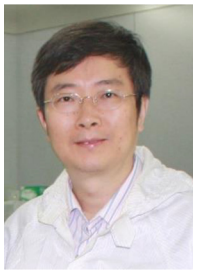

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