

http://www.scirp.org/journal/gep ISSN Online: 2327-4344

ISSN Print: 2327-4336

C-H-O Stable Isotope, Elements and Fluid Geochemistry of Uraniferous Leucogranites in Gaudeanmus Area, Southern Central Zone, Damara Orogen, Namibia

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How to cite this paper: Chen, J.Y., Fan, H.H., Wang, S.Y. and Gu, D.Z. (2017) C-H-O Stable Isotope, Elements and Fluid Geochemistry of Uraniferous Leucogranites in Gaudeanmus Area, Southern Central Zone, Damara Orogen, Namibia. *Journal of Geoscience and Environment Protection*, 5, 1-18

https://doi.org/10.4236/gep.2017.52001

Received: September 16, 2016 **Accepted:** February 7, 2017 **Published:** February 14, 2017

Abstract

This paper focuses on the effect of the later hydrotherm on uraniferous leucogranites and the stages of uranium mineralization. Here, we review C-H-O stable isotope, elements and fluid geochemistry of uraniferous leucogranites in Gaudeanmus, Namibia. The results show that there is significant increasing amount of rare earth element from non-mineralized to uraniferous leucogranites, indicating the synchronization of REE enrichment and uranium mineralization. Uranium enrichment may have close relations with Pb, Th, Co, Ni, REE in this region, so REE and U evidently exist homology. There are at least two stages of uranium mineralization by later hydrothermal alteration: firstly, due to magnatic residual high temperature and low salinity fluid, the temperature of main metallogenetic epoch ranges from 470°C to 530°C, salinity ranges from 3.55% to 9.60% NaCleq, and C, H, O stable isotope is -23% --13.6%, -53.3% - -46.4%, 7.71% - 8.81%, respectively. Secondly, due to superimposed hydrothermal fluid, the temperature, salinity, and C, H, O stable isotope is 150°C - 220°C, 4.65% - 19.05% NaCleq, -20.3% - -3.7%, -64.7% - -53.6%, 1.49% - 1.99%, respectively. The fluid for reformation is derived from postmagmatic fluid, mixed with a number of meteoric water.

Keywords

Geochemistry, C-H-O Stable Isotope, Uranium Mineralization, Leucogranites, Gaudeanmus, Damara Belt

1. Introduction

Namibia is located in southwest Africa and very rich in mineral resources. At

DOI: <u>10.4236/gep.2017.52001</u> February 14, 2017

present, there are mainly four types of uranium deposit: intrusive, Surficial and sandstone-hosted. Rössing uranium deposit is the famous leucogranite-type uranium deposits [1]. With the geological work progression, leucogranite-type uranium deposits were discovered in the surrounding areas, including Valencia, Rössing South, Ida Dome, Goanikontes, Gaudeanmus [1]-[11]. The geochronology of monazite U-Pb has shown that the U-Pb age of leucogranite is about 509 ± 1 Ma, consistent with the age of native uraninite (508 ± 2 Ma) [12] [13]. However, there are disagreements on the mechanism of leucogranite-type uranium deposit. Nex et al. concluded that leucogranite-type uranium deposits were merely related to the magmatic differentiation, didn't experience hydrothermal alteration [13]; Herd found later fluids in the leucogranites, but only related to the formation of secondary uranium uranophane [14]; while Chen et al. found that the pitchblende which was in the uraniferous leucogranites, indicated that leucogranite-type uranium deposits formed by the original magmatic crystallization differentiation and later superimposed hydrothermal alteration [10] [11]. So the opinion may relate to the later hydrothermal alteration, which has not yet reached agreement.

REE, fluid inclusion and C-H-O stable isotope is an important geochemical tracer, a powerful tool for source and the forming mechanism of mineralization [15] [16] [17]. This paper presented the C-H-O stable isotope, elements and fluid geochemistry of the uraniferous leucogranites, discussed which specific trace elements have close relation with uranium mineralization and whether postmineralized hydrotherm has superimposed transformation to the uraniferous leucogranites, and determined the stages of uranium mineralization as well. An essential evidence for metallogenic mechanism was proposed and new geochemical information to research on the metallogenic model and prognosis was provided.

2. Geological Setting

The Damara Orogen is a part of the Late Precambrian (Neoproterozoic) and Early Paleozoic (650 - 460 Ma) system of the Pan-African mobile belt, which transverses the African continent. In Namibia, the Damara Orogen has been divided into a north-south trending coastal branch and a northeast trending inland branch which has been interpreted as a result of the collision between the Congo, Kalahari and Rio de la Plata cratons [18]. The inland branch has been divided into Northern Platform, Northern Zone, northern Certral Zone, southern Certral Zone, Okahandja Lineament Zone, Southern Zone, Southern Margin Zone and Southern Foreland by the lithostratigraphical, structural and metamorphic criteria [19] [20]. The boundaries between the zones are formed by major faults or linear structures with an aeromagnetic expression [21]. Uranium-bearing leucogranites in Namibia occur within the Central Zone of the Damara Orogen [22]. The Gaudeanmus locates in the southern Certral Zone (Figure 1). The distinguishing characteristics of the southern Central Zone are a number of granitoid intrusions and low-pressure high temperature metamor-

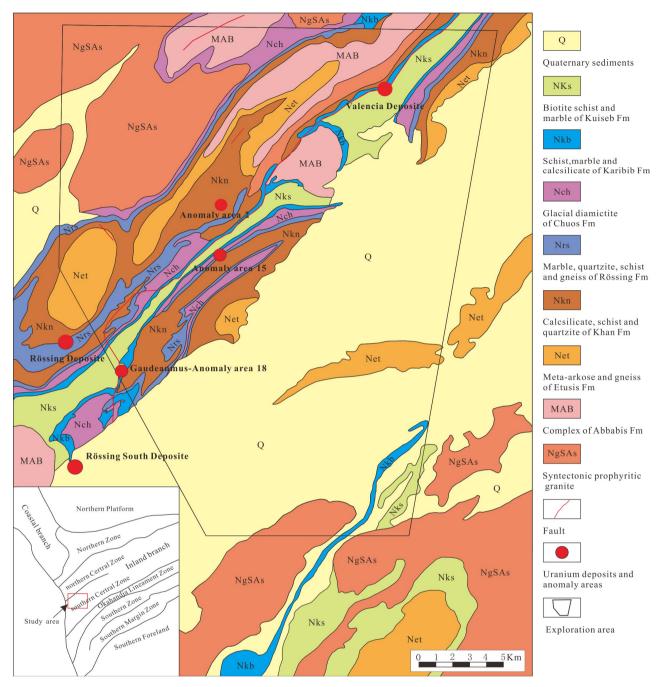


Figure 1. Geological diagram of Gaudeanmus Area, Namibia; showing the location of significant uranium anomalies.

phism at upper-amphibolite to granulite facies [23].

The strata in the Gaudeanmus have been divided into Abbabis Complex, Etusis Formation, Khan Formation, Rössing Formation, Chuos Formation, Karibib Formation, Kuiseb Formation from bottom to top. Abbabis complex is largely composed of augen gneiss and quartzofeldspathic gneiss with a small amount of argillaceous schists, quartzites, marbles, amphibolites. Geochronological studies of the gneisses have resulted in various ages, including 2093 \pm 51 Ma and 1038 \pm 58 Ma (U-Pb SHRIMP) [24]. The Etusis formation lies unconformably on the Abbabis metamorphic complex, occurs as cores to several of the prominent

domes or anticlines in the Gaudeanmus. It is mainly composed of metamorphic conglomerates, quartzites, arkoses, schists, gneisses, etc. In Damara Orogen, Rift-related acid volcanics close to the base of the Damara sequence have been dated at 746 ± 2 Ma by Hoffman et al. (1996) [25]. The Khan Formation is distinguished by the grayish-green rocks and numerous granitoid intrusions, and consists of pyroxene-garnet gneisses, schists, amphibolites. It is overlain unconformably by the Rössing Formation, which is dominated by the upper and lower sets of marble, cordierite gneisses and calcsilicates. The uranium-bearing leucogranites intrude mostly into this formation. Chuos Formation which is overlain by a major thickness of glacial diamictite containing a small quantity of calcsilicates and quartzites, and so on. These glacial deposits are interpreted as a result of the worldwide Sturtian glacial event by Hoffman, so Chuos Formation has been dated at 710 Ma [26]. Karibib is composed of thick-bedded marbles, calcsilicates, schists and gneisses, which the thick-bedded marbles developed strong joints. They are overlain by the pelitic schists, gneisses, migmatites, calcsilicates and quartzites of the Kuiseb Foramtion.

3. Petrology

In Gaudeanmus, leucogranites are common as sheeted, net-veined, and stratiform-like dykes, and vary in width from 10cm to more than 200 m across. The uraniferous leucogranites intruded into the marble of Rössing and Karibib Formations, gneiss of Khan and Kuiseb Formations. Based on detailed field observations and comparative analysis of cores, we consider that the formation of the rich ores was significantly affected by the hydrothermal alterations, which included albitization (**Figure 2(a)**), silicification, muscovitization, illitization, kaolinization, sericitization, chloritization (**Figure 2(b)**), pyritization and limonitization.

The leucogranites are texturally variable: ranging from graphic, and medium, or coarse-grained granitic to pegmatitic. They are distinguished by the smoke gray, or black quartz. The leucogranites have a simple mineralogy comprising quartz, plagioclase and K-feldspar with minor biotite and muscovite, and develop myrmekite and metasomatic edulcoration border texture (**Figure 2(c)**). Accessory minerals include zircon, apatite, monazite, epidote, rutile, sphene, pyrite (**Figure 2(d)**), galena, ilmenite, hematite and various uranium minerals. In majority of leucogranite samples, K-feldspar content is higher or slightly higher than that of plagioclase, therefore, the leucogranites are mostly Syenogranite and adamellite, a small number of alkali-feldspar granite, granodiorite and tonalite.

According to rock-mineral identification and electron microprobe analysis, In Gaudeanmus, the uranium of leucogranite-type uranium deposit mainly exists as independent uranium minerals, and partially exists in thorium minerals as isomorphic form. The uranium minerals are mainly uranium oxides, U-Ti-oxides and uranium silicates, in which uranium oxides including uraninite, pitchblende (**Figure 2(e)**, **Figure 2(f)**) and thor-uraninite. Generally, the uraninite varies from a few micrometres to 5 mm in size (**Figure 2(d)**), and often occurs

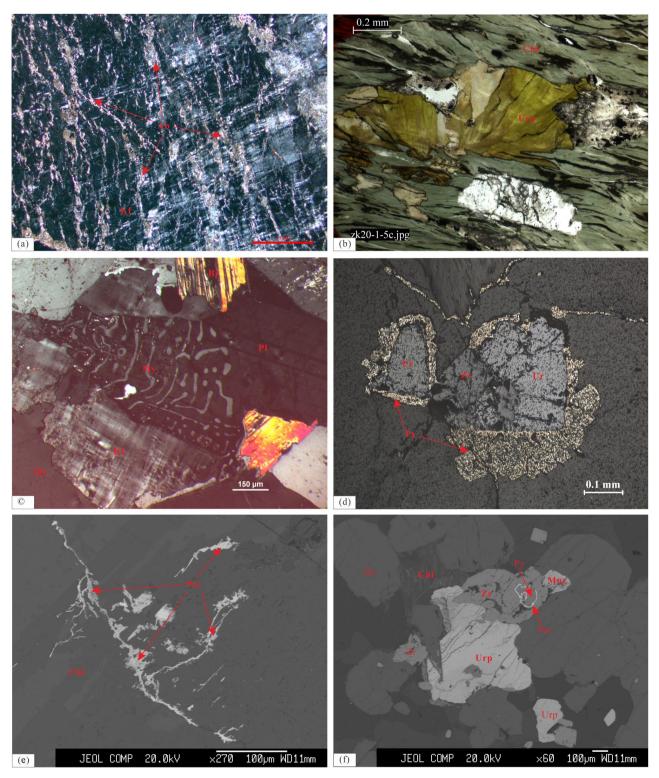


Figure 2. Petrologic and uranium mineral characteristics of leucogranites in Gaudeanmus, Namibia. (a) Feldspar(Kf) occurred albitization(Ab); (b) Biotite(Bi) occurred chloritization(Chl), with uranophane(Urp); (c) Myrmekite (My) grew in the leucogranite; (d) Uraninite(Ur) was surrounded with pyrite(Py), and coexisted with zircon(Zr); (e) Pitchblende(Ptc) filled in the fractures of chlorite (Chl); (f) Pyrite(Py) was surrounded with pitchblende(Ptc), and accompanied with uranophane(Urp), zircon(Zr), monazite(Mnz).

within plagioclase, K-feldspar and biotite. There are many radioactive halos in

the contact sites of the uraninite, biotite or feldspar. U-Ti-oxides are a complex minerals, including brannerite, betafite, and uranpyrochlore. The uranium silicates contain coffinite, uranophane (**Figure 2(b)**, **Figure 2(f)**), and uranothorite.

4. Sample Collection and Analysis

The leucogranites have been divided into six different types based on field characteristics of petrology, mineralogy and radioactivity [13]. Only the leucogranites which contains smoke gray quartz is the uranium ore. With respect to the purpose of this study, a total of 13 representative samples were collected from the drilling in Gaudeanmus, including non-mineralized leucogranites (ZK11-11-20, ZK0-11-9, ZK20-1-3), weak-mineralized leucogranites (ZK0-11-8, ZK0-11-10, ZK80-9-2), and uraniferous leucogranites (ZK76-13-5, ZK76-13-6, ZK76-13-8, ZK76-13-19, ZK80-9-4, LS-1, ZK15-2-7). The detailed sample locations are anomaly area 18 (**Figure 1**).

The concentrations of trace elements and REE in the samples were measured at the Analytical Laboratory Beijing Research Institute of Uranium Geology, using HR-ICP-MS (Element I) which manufactured by Finnigan-MAT in German, following the methods of the DZ/T0223-2001. The temperature and relative humidity of the laboratory is 20°C and 30%, respectively. The contents of trace elements and REE are greater than 10 ppm, the relative error is less than 5%, while the contents are less than 10 ppm, the relative error is less than 10%. Detailed analytical procedures for trace elements and REE are described by Qu *et al.* [27].

Doubly polished sections were prepared for fluid inclusion optical examination and microthermometric analysis. Heating and cooling experiments were carried out using a Linkam THMS 600 programmable heating-freezing stage at the Key laboratory of Uranium Resource Exploration and Evaluation Technology, CNNC. The heating rate near ice point and homogenization temperature was 0.1° C to 0.2° C/min. The reproducibility of measurements was ± 0.2 below $\pm 30^{\circ}$ C and ± 2 above 100° C, where the chips were centered in the specimen holder.

5. Elements Geochemistry

5.1. Characteristics of Trace Elements

The measured trace elements concentrations of leucogranites are listed in **Table 1**, the uranium contents of non-mineralized, weak-mineralized, uraniferous leucogranites are 13.9 - 87.8 ppm, 187 - 464 ppm, 1201 - 21,053 ppm, respectively. Rb/Sr ratios range from 1.07 to 7.53, 4.29 on average, which are significantly higher than that in the crust of eastern China (0.31) and the average of global upper crust (0.32) [28] [29]. This result indicates the leucogranites in Gaudeanmus have high maturity crustal characteristics. Nb/Ta ratios are between 8.22 and 35.82, mean 15.18; Zr/Hf ratios range 27.76 - 33.72, with an average of 30.84 (except for the sample ZK76-13-8), that are consistent with the geochemical characteristics of crust-derived type granite. Th/U ratios are between 0.07 and 1.22, mean 0.47 (the sample ZK20-1-3 may be rich in thorium

Table 1. Trace element compositions of the leucogranites in Gaudeanmus, Namibia (ppm).

Lithology	Non-mineralized leucogranites			Weak-mineralized leucogranites			Uraniferous leucogranites						
Sample No.	ZK11-11 -20	ZK0-11 -9	ZK20-1 -3	ZK0-11 -8	ZK0-11 -10	ZK80-9 -2	ZK76-13 -5	ZK76-13 -6	ZK76-13 -8	ZK76-13 -19	ZK80-9 -4	LS-1	ZK15-2 -7
Sc	4.74	4.31	1.54	5.42	3.92	17.90	3.44	5.76	14.10	8.97	19.60	13.00	20.70
Cr	10.30	4.65	2.14	2.69	2.52	10.20	4.47	5.34	5.25	4.38	18.30	21.20	31.70
Co	1.23	0.96	1.31	1.26	0.97	5.36	0.56	1.16	6.77	1.51	6.56	2.74	4.63
Ni	1.01	0.50	1.96	1.00	0.60	3.79	0.90	1.19	4.99	2.22	8.04	1.58	4.38
Cu	1.35	1.23	2.47	1.35	0.79	11.00	2.16	6.52	78.40	15.90	13.10	12.10	5.49
Zn	21.50	13.50	7.41	15.60	14.00	79.60	8.59	12.20	45.50	10.10	49.20	20.70	97.00
Rb	296.00	335.00	141.00	288.00	296.00	408.00	496.00	398.00	155.00	421.00	339.00	157.00	211.00
Sr	59.30	44.50	122.00	39.80	47.20	381.00	102.00	130.00	60.20	73.50	148.00	32.50	50.80
Y	22.30	14.40	17.40	30.50	31.20	140.00	84.20	65.90	974.00	215.00	892.00	371.00	120.00
Nb	30.00	20.10	12.70	20.80	17.50	91.50	11.10	20.20	7.98	39.10	96.00	72.70	39.90
Mo	0.53	1.87	0.32	6.56	1.35	173.00	4.51	2.45	4.33	13.80	442.00	294.00	740.00
Sb	0.04	0.05	0.06	0.06	0.04	0.26	0.55	0.26	0.04	0.34	0.20	0.16	0.05
Cs	3.25	6.02	1.09	5.21	5.34	8.97	9.67	4.84	3.27	8.64	12.10	2.28	3.20
Ba	119.00	139.00	76.60	115.00	122.00	159.00	471.00	310.00	128.00	324.00	171.00	118.00	321.00
Ta	2.98	2.14	1.05	1.42	1.68	4.70	1.35	1.48	0.28	3.98	2.68	6.08	2.96
W	2.12	1.02	1.22	1.24	1.04	3.07	2.10	1.52	4.74	9.88	4.12	7.11	7.82
Pb	36.60	34.60	29.70	41.80	41.40	58.40	132.00	115.00	651.00	189.00	1405.00	648.00	406.00
Th	107.00	76.50	831.00	183.00	207.00	357.00	132.00	90.80	1693.00	242.00	2805.00	709.00	382.00
U	87.80	65.00	13.90	291.00	187.00	464.00	1182.00	1201.00	10034.00	2551.00	21053.00	9581.00	4525.00
Zr	87.50	37.20	291.00	276.00	170.00	258.00	640.00	761.00	615.00	2183.00	2860.00	769.00	381.00
Hf	3.03	1.34	9.69	9.13	5.81	8.60	19.60	23.00	3.41	67.60	89.40	25.50	11.30
Nb/Ta	10.07	9.39	12.10	14.65	10.42	19.47	8.22	13.65	28.30	9.82	35.82	11.96	13.48
Zr/Hf	28.88	27.76	30.03	30.23	29.26	30.00	32.65	33.09	180.35	32.29	31.99	30.16	33.72
Th/U	1.22	1.18	59.78	0.63	1.11	0.77	0.11	0.08	0.17	0.09	0.13	0.07	0.08
Rb/Sr	4.99	7.53	1.16	7.24	6.27	1.07	4.86	3.06	2.57	5.73	2.29	4.83	4.15
Ba/Rb	0.40	0.41	0.54	0.40	0.41	0.39	0.95	0.78	0.83	0.77	0.50	0.75	1.52

minerals and is excluded), showing the features of magmatogenic granite. And in the non-mineralized and weak-mineralized leucogranites, Ba/Rb ratios range from 0.39 to 0.54, with an average of 0.43, while the Ba/Rb ratios of uraniferous leucogranites are between 0.5 and 1.52, with an average of 0.87, consequently it can be used as the indicator to differentiate between the mineralized and non-mineralized leucogranites.

In the primitive mantle-normalized spider diagram of trace elements (**Figure 3**), all the leucogranites show enrichment in large ion lithophile elements (LILE), especially Rb, Th, U, Pb, and relative depletion in high field strength elements such as Ti, P. The significant negative anomalies of Sr may be associated with magmatic differentiation or low calcium content in magma, indicating the

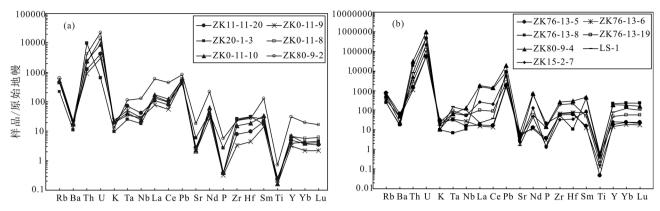


Figure 3. The primitive mantle-normalized spider diagram of trace element in Gaudeanmus, Namibia (data of primitive mantle are from [30]). (a) the diagram of non-mineralized and weak-mineralized leucogranites; (b) the diagram of uraniferous leucogranites.

evolution of partial melting or crystal fractionation of magma might involve the fractionation of a few of plagioclases. The strong negative Ba anomalies are the characteristics of anorgenic granite, and Ta enrichment also reflects the presence of strong magmatic crystallization differentiation. The depletion of Ti probably results from the fractionation of Fe-Ti oxides, indicating that the leucogranites have obviously experienced a high degree of crystallization differentiation.

5.2. Characteristics of REE

As listed in **Table 2**, total rare earth elements (REE) contents of the non-mineralized, weak-mineralized, uraniferous leucogranites are 222.02 - 377.26 ppm (average 302.59 ppm), 385.06 - 763.00 ppm (average 878.07 ppm), 109.19 - 5994.28 ppm (average 1925.79 ppm), respectively. The results show there is an obvious growth trend from non-mineralized to uraniferous leucogranites, which might be related with uranium minerals enriched in REE. There are two types of chondrite-normalized REE patterns of the leucogranites (**Figure 4**): one is the rightinclining type with obvious Eu negative abnormity and enrichment of LREE (**Figure 4(a)**); the other is leftinclining type with obvious Eu negative abnormity and enrichment of HREE (**Figure 4(b)**). The strong Eu depletion (δ Eu = 0.03 - 0.47, average 0.22), indicating that the magma experienced strong crystallization differentiation.

The samples of rightinclining type include all non-mineralized, weak-mineralized and some uraniferous leucogranites (ZK76-13-8, ZK80-9-4 and ZK15-2-7), LREE/HREE = 6.63 - 25.11, mean 13.60, which gradually decreases from non-mineralized to uraniferous leucogranites. $\delta Eu = 0.03 - 0.39$, mean 0.18, reflecting the magma of leucogranites is originated from the partial melting of superficial part of the crust. These samples are relatively enriched in LREE with significant fractionations of HREE and LREE ((La/Yb)_N = 6.95 - 43.79, mean 25.15), and possess weak Ce anomalies ($\delta Ce = 0.91 - 1.01$, mean 2.87 - 1.01), mean 2.87 - 1.01 mean 2.

Table 2. REE concentrations of the leucogranites in Gaudeanmus, Namibia (ppm).

Lithology	Non-mineralized leucogranites		Weak-mineralized leucogranites			Uraniferous leucogranites							
Sample No.	ZK11-11 -20	ZK0-11 -9	ZK20-1 -3	ZK0-11 -8	ZK0-11 -10	ZK80-9 -2	ZK76-13 -5	ZK76-13 -6	ZK76-13 -8	ZK76-13 -19	ZK80-9 -4	LS-1	ZK15-2 -7
La	73.50	53.50	103.00	90.60	116.00	405.00	12.00	10.90	1115.00	72.20	1277.00	15.90	180.00
Ce	139.00	101.00	176.00	170.00	216.00	796.00	29.60	25.50	2244.00	161.00	2558.00	68.20	353.00
Pr	15.20	11.10	18.40	20.10	25.40	85.80	3.93	3.48	243.00	20.00	279.00	11.20	48.40
Nd	51.50	37.10	56.90	65.80	84.50	294.00	17.30	15.20	831.00	78.90	962.00	61.20	177.00
Sm	9.60	6.39	7.83	11.80	15.20	57.20	6.83	5.82	182.00	23.50	210.00	32.50	40.30
Eu	0.81	0.73	0.68	0.70	0.85	1.62	1.16	0.86	2.86	1.83	2.18	1.66	1.12
Gd	7.10	4.66	5.42	9.06	11.40	46.00	8.48	7.22	163.00	26.80	187.00	38.90	32.70
Tb	1.11	0.75	0.71	1.49	1.71	7.73	2.49	2.03	32.90	6.84	37.50	13.30	6.76
Dy	5.28	3.47	3.35	7.38	8.42	36.80	17.90	14.30	196.00	46.00	214.00	101.00	34.60
Но	0.82	0.54	0.57	1.21	1.21	6.16	3.76	2.95	39.40	9.34	40.80	21.50	5.35
Er	2.15	1.37	1.71	3.23	2.92	14.00	11.50	8.92	114.00	27.60	109.00	65.30	14.00
Tm	0.33	0.19	0.28	0.48	0.36	1.90	1.97	1.52	19.50	4.70	16.70	11.00	2.12
Yb	1.85	1.05	2.07	2.76	1.90	9.54	12.20	9.13	115.00	28.90	89.20	61.20	12.20
Lu	0.25	0.16	0.34	0.46	0.30	1.25	1.80	1.36	17.00	4.37	11.90	7.46	1.60
Y	22.30	14.40	17.40	30.50	31.20	140.00	84.20	65.90	974.00	215.00	892.00	371.00	120.00
Σ REE	308.50	222.02	377.26	385.06	486.16	1763.00	130.92	109.19	5314.66	511.98	5994.28	510.32	909.15
LREE	289.61	209.82	362.81	359.00	457.95	1639.62	70.82	61.76	4617.86	357.43	5288.18	190.66	799.82
HREE	18.89	12.20	14.45	26.07	28.21	123.38	60.10	47.43	696.80	154.55	706.10	319.66	109.33
LREE/ HREE	15.33	17.20	25.11	13.77	16.23	13.29	1.18	1.30	6.63	2.31	7.49	0.60	7.32
$(La/Yb)_N$	28.50	36.55	35.69	23.55	43.79	30.45	0.71	0.86	6.95	1.79	10.27	0.19	10.58
$(La/Sm)_N$	4.94	5.41	8.49	4.96	4.93	4.57	1.13	1.21	3.96	1.98	3.93	0.32	2.88
$(Gd/Yb)_N$	3.17	3.67	2.17	2.72	4.96	3.99	0.58	0.65	1.17	0.77	1.73	0.53	2.22
δEu	0.29	0.39	0.30	0.20	0.19	0.09	0.47	0.40	0.05	0.22	0.03	0.14	0.09
δCe	0.97	0.96	0.92	0.94	0.93	1.00	1.05	1.01	1.01	1.02	1.00	1.20	0.91

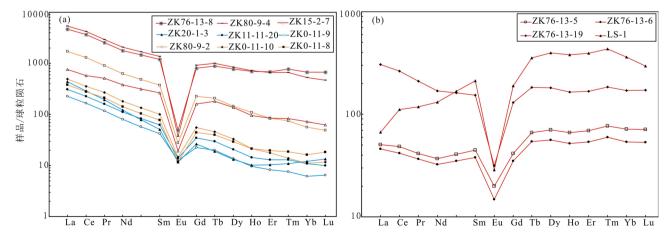


Figure 4. Chondrite-normalized REE distribution patterns of the leucogranites in Gaudeanmus, Namibia (data of chondrite are from [30]). (a) rightinclining type with enrichment of LREE; (b) leftinclining type with enrichment of HREE.

magmatic crystallization differentiation.

The REE patterns of some uraniferous leucogranites (ZK76-13-5, ZK76-13-6, ZK76-13-19, LS-1) present leftinclining type, LREE/HREE ranged from 0.6 to 2.31, average 1.35, (La/Yb) $_{\rm N}$ = 0.19 - 1.79, average 0.89, δ Eu = 0.14 - 0.47, average 0.31, δ Ce = 1.01 - 1.20, average 1.07, indicating that these samples are relatively enriched in HREE, without fractionations of REE. (La/Sm) $_{\rm N}$ = 0.32 - 1.98, average 1.16, and (Gd/Yb) $_{\rm N}$ = 0.53 - 0.77, average 0.63 respectively, showing that there is no obvious LREE and HREE internal fractionation. The results suggest uranium mineralization is controlled by later hydrothermal.

6. Ore Forming Fluid Geochemistry

A systematic research on the ore-forming fluid, can judge the uranium mineralization stages and different uranium mineralization [31]. Therefore, a detailed study of the quartz inclusion in mineralization granite and quartz veins, found that there are two periods of ore-forming fluid. The primary inclusions of main metallogenic epoch are isolated or clustered, homogenization temperature concentrated in 470°C - 530°C (**Figure 5**), salinity is 3.55% - 9.60% NaCleq, mean 6.14% NaCleq. It belongs to magnatic residual high temperature and low salinity fluid. Another inclusions is secondary, distributed as linear or banded, homogenization temperature ranges from 150°C to 220°C (**Figure 5**), salinity is 4.65% - 19.05% NaCleq, average 11.5% NaCleq. Obviously, the late hydrothermal fluids for reformation is mid-low temperature, low salinity fluid [32].

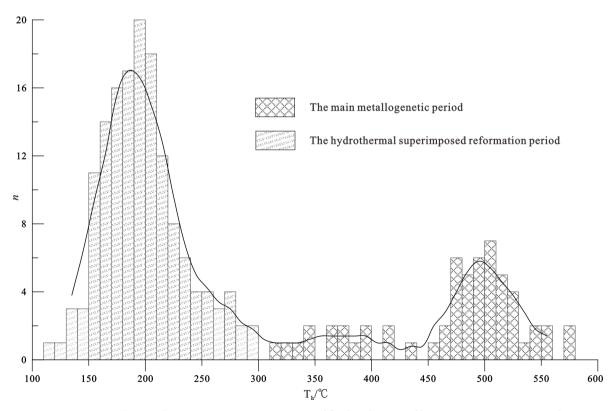


Figure 5. Histogram showing homogenization temperatures of fluid inclusions of leucogranite-type uranium deposit in Gaudeanmus area, Namibia.

7. Discussion

7.1. Correlation between Trace Elements and Uranium Mineralization

In order to find out the trace elements which are closely related with U, correlation and R-type clustering analyses have been made to the non-mineralized, weak-mineralized, uraniferous leucogranites by SPSS statistical software (**Table 3**, **Figure 6**). The results show the relationship between trace elements and U is not consistent from non-mineralized to uraniferous leucogranites. Therefore, this study focuses on the uraniferous leucogranites to investigate the relationship between trace elements and U.

Table 3. Correlation coefficients of U and trace elements of leucogranites in Gaudeanmus, Namibia.

Element	Sc	Cr	Co	Ni	Cu	Zn	Rb	Sr	Nb	Mo
Non-mineralized leucogranite	0.984	0.906	-0.424	-0.792	-0.923	0.957	0.879	-0.884	0.955	0.336
Weak-mineralized leucogranite	0.960	0.935	0.949	0.965	0.945	0.936	0.905	0.921	0.942	0.938
Uraniferous leucogranite	0.685	0.339	0.793	0.855	0.260	0.318	-0.391	0.246	0.754	0.385
Element	Sb	Cs	Ва	Ta	W	Pb	Th	Zr	Hf	ΣREE
Non-mineralized leucogranite	-0.976	0.621	0.811	0.990	0.612	1.000	-0.942	-0.880	-0.878	-0.625
Weak-mineralized leucogranite	0.956	0.917	0.863	0.900	0.958	0.936	0.874	0.678	0.685	0.902
Uraniferous leucogranite	-0.427	0.293	-0.717	0.117	0.002	0.997	0.963	0.587	0.527	0.849

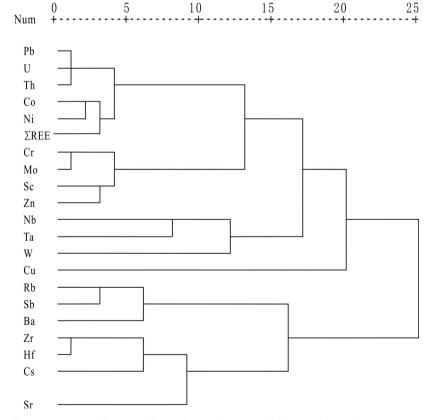


Figure 6. R-type cluster analysis of trace elements of the uraniferous leucogranites in Gaudeanmus, Namibia.

According to the correlation between trace elements and uranium mineralization (**Table 3**, **Figure 6**), in the uraniferous leucogranites, U is positively correlated to Sc, Cr, Co, Ni, Cu, Zn, Sr, Nb, Mo, Cs, Ta, W, Pb, Th, Zr, Hf, and REE, especially Pb, Th, Co, Ni, and REE (the correlation coefficients range from 0.793 to 0.997), and negatively to Rb, Sb, Ba. The correlation analysis suggests that Pb, Th, REE and U exist homology evidently.

7.2. Stages of Uranium Mineralization

According to the spider diagram of trace elements and REE patterns, in Gaudeanmus, the leucogranite-type uranium deposits are principally derivatives of magmatic crystallization differentiation. In this stage, uranium minerals contain uraninite, thor-uraninite, coffinite, uranothorite, brannerite and betafite, which are primary uranium minerals.

Geochemistry of large ion lithophile elements are active, they are useful in determining the extent of element mobility and fluid-rock interaction associated with granites [33]. The nonlinear arrays on LILE diagrams (Rb, Ba, Sr and K) reveal that hydrothermal or post-magmatic variation has been a significant factor in the leucogranites in Gaudeanmus (**Figure 7**). Particularly, in the Ba vs Sr

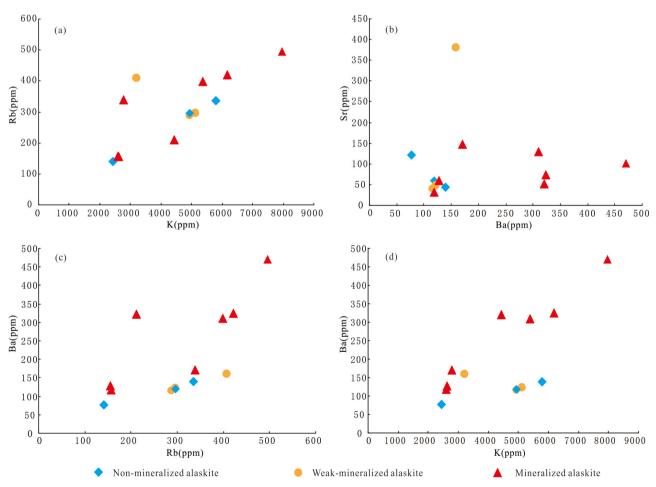


Figure 7. Graphs of LILE variation for the leucogranites in Gaudeanmus, Namibia: (a) K vs Rb; (b) Ba vs Sr; (c) Rb vs Ba; (d) K vs Ba.

diagram (**Figure 7(b)**), the mineralised leucogranites have been obviously affected by later hydrothermal reformation. This result is consistent with the phenomenon which mineralised leucogranites occurred kaolinization, sericitization, chloritization, pyritization and hematitization.

Later hydrothermal reformation leads to uranium mineralization again, this opinion also can be confirmed by the leftinclining type REE patterns. It is a common phenomenon that there is an increase in the contents of REE and HREE in the process of hydrothermal uranium mineralization, especially HREE [34] [35] [36]. The principal reasons are as follows: 1) Affected by the "lanthanide contraction", the decrease in ionic radius of HREE and increase in ionization potential, thus relatively enhance the coordination ability of HREE, which makes sense intuitively as the HREE are stronger in mobility with the complexes than LREE, finally causes the fractionation between LREE and HREE of the hydrotherm; 2) REE and U can move together with the analogous complexes in the hydrotherm. A large number of studies have indicated that metallogenic hydrothermal uranium mainly migrate as the form of uranyl carbonate ligands in $UO_2(CO_3)_2^{2-}$ and $UO_2(CO_3)_3^{4-}$ [37] [38] [39], CO_2 of metallogenic hydrotherm promotes the mobilization of uranium. Research on the fluid inclusions in uraniferous leucogranites, indicates that there are a great quantity of CO2 in the ore-forming fluid. Therefore, it can be inferred that the hydrothermal uranium in this stage move also in the form of $UO_2(CO_3)_2^{2-}$ and $UO_2(CO_3)_3^{4-}$. In the rich HCO₃ hydrotherm, U, Th and HREE have a more similar geochemical behavior, and the ability to form complexes with HREE is far greater than LREE [39]. Then, in the process of hydrothermal uranium mineralization, HREE are much more enriched than LREE. Pitchblende and veined coffinite are the representative uranium minerals of this stage.

7.3. Origin of Mineralizing Fluid

Stable isotope is an important method of researching deposits and tracing ore-forming fluid. This study tested 6 quartzs of uraniferous leucogranites and 3 quartz veins, the analysis results are shown in **Table 4**. $\delta^{18}O_{Qtz}$ of Six quartz samples changes from 13.3% - 14.4%, average 13.65%, according to the fluid inclusion temperature mode and quartz-water system oxygen isotope equilibrium fractionation equation [40], the calculation of $\delta^{18}O_{H2O}$ varies from 7.71% to 8.81%, an average of 8.06%, δ 18D is from -53.3% to -46.4%, indicating that this period uranium mineralization is magmatic hydrothermalism. $\delta^{18}O_{Qtz}$ of Superimposed phase quartz vein samples is between 13.2% and 13.7%, an average of 13.5%, the calculated $\delta^{18}O_{H2O}$ changes in 1.49% - 1.99%, mean 1.79%, $\delta^{18}D$ varies from -64.7% to -53.6%.

In the δD - $\delta^{18}O_{H2O}$ diagram (**Figure 8**), H and O isotope values of the main metallogenic stage were distributed in the magmatic water area, reflect that the period uranium mineralization is formed by magmatic crystallization differentiation. Besides, $^{143}Nd/^{144}Nd$, $^{87}Sr/^{86}Sr$ and Sm/Nd ratios of uraniferous leucogranites are consistant with that of Abbabis complex and Etusis strata [41]. Therefore,

Table 4. C, H and O isotope composition of uraniferous leucogranite quartzs and quartz veins in Gaudeanmus Area, southern Central Zone, Damara Orogen, Namibia.

Sample no.	Lithology	δ ¹⁸ O _{Qtz} (‰)	δ ¹⁸ O _{H2O} (‰)	δD (‰)	$\delta C_{ ext{PDB}}\left(\% ight)$
ZK76-13-2		13.3	7.71	-52.3	-13.6
ZK76-13-8		14.4	14.4 8.81		-20.8
ZK76-13-10	Quartz in the	13.6	8.01	-50.9	-23
ZK76-13-12	uraniferous leucogranites	13.8	8.21	-46.4	-19.2
ZK76-13-16	-	13.5	7.91	-51	-19.7
ZK84-6-1		13.3	7.71	-53.3	-19
ZK76-9-1		13.2	1.49	-64.7	-14.1
13H-3	Quartz vein	13.6	1.89	-60.1	-20.3
13H-5		13.7	1.99	-53.6	-3.7

Note: the $\delta^8 O_{H2O}$ values of ore fluids were calculated using the quartz-water equilibrium fractionation equation $1000 \ln \alpha_{Qiz\cdot H2O} = 3.38 \times 10^6/T^2$ -3.40 [40], the temperatures of uraniferous leucogranite quartzs and quartz veins are $T_1 = 340^{\circ} C$ (average homogenization temperature of three-phase inclusions), $T_2 = 200^{\circ} C$ (average homogenization temperature of inclusion), respectively [32].

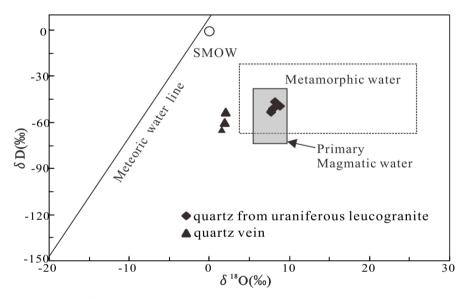


Figure 8. δ D- δ ¹⁸O_{H2O} diagram of mineralizing fluid in Gaudeanmus Area, southern Central Zone, Damara Orogen, Namibia.

uranium mainly source from Abbabis and Etusis. H and O isotope values of the superimposed reformation stage deviates significantly from magmatic water area, drifts towards the meteoric water line, indicating that the period mineralizing fluid is post-magmatic hydrothermal fluid mixed with meteoric water.

In addition, the $\delta^{13}C_{PDB}$ of quartz veins ranges from -20.3% to -3.7%, average -12.7%, is close to $\sigma^{13}C$ value of CO_2 in atmosphere or freshwater (-11% --7%) [42], and may be mixed with organic carbon, revealing that $\sigma^{13}C$ of late quartz veins is derived mostly from atmosphere or freshwater. The $\delta^{13}C_{PDB}$ of quartz in the uraniferous leucogranites is between -23% and -13.6%, mean -19.22%, basically within the $\sigma^{13}C$ range of magmatic lineage (-30% --3%)

[42], therefore, σ^{13} C of the uraniferous leucogranites stems chiefly from the magma. In conclusion, leucogranite uranium mineralization is chiefly formed by magmatic crystallization differentiation, with late hydrothermal reformation.

8. Conclusions

- 1) The contents of total rare earth elements in the non-mineralized, weak-mineralized, uraniferous leucogranites are 222.02 377.26 ppm (mean 302.59 ppm), 385.06 1763.00 ppm (mean 878.07 ppm), 109.19 5994.28 ppm (mean 1925.79 ppm), respectively, showing that there is an obvious growth trend from non-mineralized to uraniferous leucogranites, it suggests that uranium mineralization presents synchronous increase trend with REE. δ Eu = 0.03 0.47, with an average of 0.22, indicating that magma experienced strong differentiation crystallization.
- 2) In Gaudeanmus, leucogranites show enrichment in large ion lithophile elements, especially Rb, Th, U, Pb, and relative depletion in Ti, P, Sr and Ba. In the uraniferous leucogranites, there is a high correlation between U and Pb, Th, Co, Ni, REE, the correlation coefficient is 0.793 0.997, indicating that U is highly homologous with Pb, Th and REE.
- 3) The leucogranite-type uranium deposite in Gaudeanmus can be divided into two stages: one is main metallogenic epoch, the representative uranium minerals of this stage formed by magmatic crystallization differentiation, which are uraninite, thor-uraninite, coffinite, uranothorite, brannerite and betafite. And the REE pattern is characterized by rightinclining type with obvious Eu negative abnormity and enrichment of LREE. The other is secundarii metallogenic epoch, secondary uranium minerals of this stage produced by later hydrothermal reformation, representing by pitchblende and veined coffinite. And this REE pattern manifests as leftinclining type with obvious Eu negative abnormity and enrichment of HREE.
- 4) The main mineralization temperature of uraniferous leucogranites concentrates on 470°C 530°C, the salinity is 3.55% 9.60% NaCleq, with C, H and O isotopic characteristics of typical magmatic source, -23‰ -13.6‰, -53.3‰ -46.4‰, 7.71‰ 8.81‰, respectively, further revealing that the leucogranite-type uranium deposit in Gaudeanmus area was chiefly formed by magmatic crystallization differentiation. The temperature of superposed hydrothermal fluid centeres on 150°C 220°C, the salinity is 4.65% 19.05% NaCleq, C, H and O isotope is -20.3‰ -3.7‰, -64.7‰ -53.6‰, 1.49‰ 1.99‰, reflects this period hydrothermal reformation mixed with atmospheric water.

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