DETERMINATION OF THE ELEMENTAL COMPOSITION AND THE AGE OF GEOLOGICAL SAMPLES COLLECTED AT THE KAILASH MOUNTAIN (TIBET) BY X-RAY AND NUCLEAR ANALYTICAL METHODS

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Abstract. X-ray fluorescence (XRF), gamma-activation (GAA) and neutron activation (NAA) methods were applied for the elemental analysis of geological samples taken in the Western part of the Tibetan plateau (near the mount Kailash). The geological classification of the samples and the estimation of their ages were made based on analytical results. The sample ages were evaluated by strontium method based on the radioactive decay of the isotope Rb-87 and its turning into Sr-87. The content of Sr-87 in minerals was determined using GAA. Data on the total contents of Rb and Sr in the samples were obtained using XRFA and checked by NAA. The age of volcanic rocks is estimated at 24.6 \pm 0.5 Ma and the age of the youngest rocks is about 10.1 \pm 1.4 Ma. The dating is well fitted to the estimation of other sources.

Introduction

Kailash mountain in South Tibet of China is famous as a sacrificial center for Hindu, Buddhist, and Bonist pilgrims. In the last decades, there has been the growing of interests in the tectonic evolution pattern of the continental collision of the Indian and Asian plates with the closing and subduction of the Tethyan Ocean located between them in the Eocene. Consequently, events followed to the evolution as the Himalaya formation; as well as the post-collisional complex process of deformation, metamorphism, plutonism, thrusting, northsouth compression, extension, and uplift of mountain ranges have been investigating more and more. Kailash mountain is located to the northern barrier of the Indus-Tsangpo (or Zangbu). Suture that is a thrust fault representing the Eocene boundary of the Indian and Asian plates, with subsequent right lateral strike-slip displacement. The high average elevation of this area (about 17,000 feet) is typical of the western Tibetan Plateau and results from the ongoing penetration of India into Asia. Although there has been some scattering of data, but many geochronologic and geologic data have been supporting this evolution.

By the purpose of examining the famous place, this work was carried out to determine trace-element composition of geological samples taken at the Kailash mountain, and to estimate their age using X ray and nuclear analytical techniques.

Experimental

13 geological samples of six rock types collected by geologists during the expedition in 2013-2014 at many sites on different faces of the Kailash mountain and at the height of 5390 to 5800 meters over the sea level.

Elemental and isotopic concentrations of these samples were determined using three analytical methods – the X-ray Fluorescence Analysis (XRFA), the Gamma Activation Analysis (GAA) and Instrumental Neutron Activation Analysis (INAA) carried out in The Flerov Laboratory of Nuclear Reactions (FLNR) and in The Frank Laboratory of Neutron Physics (FLNP), respectively.

Before the analysis all the samples were dried at a temperature of 105°C, milled and passed through a sieve 1 mm in diameter.

Name	Samp. number	Collecting site
K1	2	on the left of the east face base , at 5570 M
K2	2	on the left of the top stage of the western face, at 5390 M
K3	2	in the middle of the northern face base
K4	2	inner crust on the western ridge
K5	3	inner layer under the conglomerate layer of 13 feet of the southern face, at 5800 м
K6	2	outer layer, on the right of eastern face

Table 1. Collecting sites

X-ray fluorescence analysis of samples (XRFA): The geological samples and standards were placed into polyethylene cylindrical cassette 35 mm in diameter and 5 mm in height and were closed on the ends with a lavsan film 6–10 μ m thick. Determination of stable elements were carried out on a standard Canberra spectrometer with the semi-conductor Si(Li) detector with full width at half maximum (FWHM) resolution 145 eV for a line of Fe (6.4 keV). Standard ring-shaped radioisotope sources of ¹⁰⁹Cd (E = 22.16 keV, T_{1/2} = 453 days) and ²⁴¹Am (E=59.6 keV, T_{1/2}=458 years) with an activity of 20 mCi for excitation of X-ray radiation were used. The Canberra company analysis software such as WinAxil and WinFund were used for elemental contents of the samples. The united standard curve was used for analysis and calculating the elemental content of the samples.

Gamma activation analysis (GAA): The samples were irradiated during 2 - 4 hours by bremsstrahlung gamma ray produced by stopping electrons with $E_e = 25$ MeV in the MT-25 microtron. The electron current was 15 mA. Gamma-ray spectra of the irradiated samples after gamma-activation by the MT-25 microtron were measured using HPGe detector with FWHM resolution of 1.5 keV for the 1332.5 keV photons of ⁶⁰Co and the measuring time was chosen from 600 to 3600 s.

Instrumental neutron-activation analysis (NAA) was performed to refine of the total content of rubidium and strontium in the samples, the calculations based on the reactions of ⁸⁵Rb (n, γ) ⁸⁶Rb and ⁸⁴Sr (n, γ) ⁸⁵Sr. To determine concentration of the long-lived isotopes ⁸⁶Rb (half-life: 18.7 days) and ⁸⁵Sr (half-life: 64.8 days), about 0.1g of dry weight of every sample was packed in aluminum cup; the sample cups were irradiated for 2.5 days in the Cd-screened channel 1 of the pulsed fast reactor IBR-2, FLNP, JINR. Then they were re-packed after 3 days of decay. Sample gamma spectra were obtained by measuring with a Ge (Li) detector with resolution of 2.5-3 keV for the ⁶⁰Co 1332 keV line or HPGe detector with resolution of 1.9 keV for the ⁶⁰Co 1332 keV line.

Data processing was performed using the software developed in FLNP JINR, and element concentrations were determined on the basic of certified reference materials [1]. To provide quality control, contents of elements yielding short – lived and long – lived isotopes were determined using a group of certified reference materials served by International Atomic Energy Agency (IAEA) and the United States National Institute of Standards and Technology (US NIST).

Rubidium-Strontium dating method was used to estimate the sample age. The method is based on the radioactive decay of the isotope ⁸⁷Rb and its transformation into ⁸⁷Sr. The basic equation of decay [2] is used for calculations

 ${}^{87}\text{Sr} = {}^{87}\text{Sr}_{1} + {}^{87}\text{Rb}(e^{\lambda t} - 1)$ (i: initial) (1).

The radiogenic ⁸⁷Sr isotope content in the sample depends on the time. In practice, the ratio of isotopes in the sample of rock or a mineral is considered. Therefore the equation (1) is divided through by the number of ⁸⁶Sr atoms which is constant during the time ⁸⁶Sr = ⁸⁶Sr_i. This gives us the equation

 ${}^{87}\text{Sr}/{}^{86}\text{Sr} = ({}^{87}\text{Sr}/{}^{86}\text{Sr})_i + {}^{87}\text{Rb}/{}^{86}\text{Sr} (e^{\lambda t} - 1)$ (2).

The half-life of ⁸⁷Rb is $T_{1/2} = 4.89 \cdot 10^{10}$ and $\lambda = \ln 2/T_{1/2}$, so that λ is very small, then $(e^{\lambda t} - 1)$ reduces to λt , and the decay equation is

This equation has the form of a straight line with the slope $a = \lambda t$, called isochrone line.

Thus, to determine the age need to know the content of specific isotopes of rubidium $({}^{87}\text{Rb})$ and strontium $({}^{87}\text{Sr}, {}^{86}\text{Sr})$.

The rubidium-strontium dating method has usually carried out using Mass Spectrometry (for example Inductively coupled plasma mass spectrometry (ICP-MS), Thermal ionization mass spectrometry (TIMS), Secondary ion mass spectrometer (SIMS)) [3], but this work shows that this dating method could be realized using nuclear analytical techniques.

shows that this dating method could be realized using nuclear analytical techniques. We used special condition to obtain ^{87m}Sr, because the isomeric state of ^{87m}Sr can be obtained using gamma irradiation by two reactions, such as ⁸⁷Sr(γ , γ ')^{87m}Sr (E = 388 keV, T_{1/2} = 2.81h) (useful) and ⁸⁸Sr (γ , n) ^{87m}Sr (unuseful). In order to obtain only one optimal useful process, energy of irradiating beam should be near E_{γ} = 10 MeV [4]. Fig.1 shows that a threshold of the reaction ⁸⁸Sr (γ , n) ^{87m}Sr is E_{γ} =11.2 MeV [4].



Fig.1. Yield of ^{87m}Sr isomeric state according to the irradiation energy; a - the yield of the isomeric state ^{87m}Sr with the contribution of the reaction ⁸⁸Sr $(\gamma,n)^{87m}$ Sr, b - the yield without the contribution of the reaction ⁸⁸Sr $(\gamma,n)^{87m}$ Sr.

Results and Discussion

Elemental concentrations of the samples (more 30 elements, such as Na, Mg, Cl, K, Ca, Sc, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Ga, As, Y, Zr, Nb, Mo, Ag, Cd, Sb, Cs, Ba, La, Ce, Nd, Ir, Hg,

Pb, Th, U including Rb and Sr) were determined by XRFA, GAA and NAA. The classification of geological samples and an estimate of their age were conducted on the basis of these results (Table 2).

Especially, the total contents of Rb and Sr were determined by XRFA and checked by NAA. Figure 2 shows the X-Ray spectrum of the K1 sample, collected near the Kailash mount.



Fig.2. The X-Ray spectrum of the K1 sample, $T_m = 600$ s.

The ⁸⁷Sr isotope content was determined by GAA using the microtron MT-25, with irradiation gamma ray of 10 MeV energy. Gamma spectrum of the K4 sample after gamma-activation with $E\gamma=10$ MeV is shown in Fig. 3.



Fig. 3. Gamma spectrum of the K4 sample after gamma-activation with $E\gamma=10$ MeV, $T_m = 300$ s.

Siberian Trap (CT-1A) with a known age was used as a standard for determining of the ⁸⁷Sr content and age of the samples. The authors of this article [5] reported that the Siberian Traps age is near 250 Ma.

As a result, if it is determined the total content of strontium (by XRFA) and radiogenic isotope content (by GAA) can be calculated all other isotopes of strontium (⁸⁴Sr, ⁸⁶Sr, ⁸⁸Sr).

The calculations used data recommended by the Sub-Commission on the chronology of the International Union of Geological Sciences [2], such as

 λ_{Rb} - the decay constant for the Rb-87 ($\lambda = 1.42 \cdot 10^{-11} \text{ y}^{-1}$);

relations: ${}^{85}\text{Rb}/{}^{87}\text{Rb}=2.59265 \rightarrow {}^{87}\text{Rb}=27.86 \%$ ${}^{86}\text{Sr}/{}^{88}\text{Sr}=0.119400$ ${}^{84}\text{Sr}/{}^{86}\text{Sr}=0.0565884$ Rubidium-Strontium dating method is useful in the case of

- measuring of the all related data is correctly;
- the samples are co-genetic and old aged;
- the rock must not have undergone any re-formation which could have disturbed the Rb-Sr system.

Also, it is important to select the correct value $({}^{87}\text{Sr}/{}^{86}\text{Sr})_i$ for the preliminary calculations. In the preliminary calculations we took $({}^{87}\text{Sr}/{}^{86}\text{Sr})_i=0.704$, like into typical of magmatic rocks.

The calculated data of an age estimate for the Kailash samples are shown in the graphic (Fig 4).



Fig. 8. The isochrone lines for Kailash samples.

They are fitted to two isochrone lines. One line relates to the age of 24.6 ± 0.5 Ma and the other to the one of 10.1 ± 1.4 Ma.

As mentioned before, the collected samples were identified, classified by geologists and they characterized the composition of Kailash conglomerates on the surface not for the underlay of the mountain. The Table 2 shows the sample specialties and their ages.

Name	Rock origin	Rock type	Age (Ma)
K1	Volcanic	Meta-andesite	24.6±0.5
K2	Plutonic	Leuco-granite	10.1±1.4
K3	Volcanic debris	Gravel of volcanic	24.6±0.5
		debris	10.1±1.4
K4	Sedimentary	Arkose sandstone of	24.6±0.5
		destructed granite	10.1±1.4
K5	Sedimentary	Sandstone	10.1 ± 1.4
K6	Plutonic	Plagio-granite	24.6±0.5

Table 2. The rock origins, rock types and the ages of the samples examined in this work

The results for the age determination of samples from the area of Mount Kailash are in good agreement with the description of the tectonic evolution of Tibet.

Kailash area is situated in the Gangdese magmatic belt that consists chiefly of granodiorite granite association with wide variation in composition, texture, and well-defined transitional types. Based on original work reported by Heim and Gansser [6], Gansser [7] the Kailash mountain was named as the Kailas conglomerate; however, conglomerate forms only a fraction of the

unit. The lower part of the Kailas Formation is dominated by andesitic volcanic and granitoid rocks of the Gangdese magmatic arc. The tectonic significance of the Kailas conglomerates remains a fundamental problem in understanding the post-collisional history of the Kailash zone formation [8-10]. Kailash mountain position is shown in Fig. 5.



Fig. 5: Kailash mountain position (soure:econgeol.geoscienceworld.org).

The tectonic evolution of Himalayas depends upon multiple geological events that could occur concurrently or sequentially. The formation of southern Tibet has usually explained by three events: the uplift of the Gangdese belt, the sedimentation in Kailash basin (the flank of the southern margin of the Gangdese belt), and the thrust emplacement at the Zangbo suture zone (Indus-Yarlung Zangbu suture) [11].

Adherents to the Hindu, Buddhist, Jain, and Bön faiths regard mountain Kailas to be sacred and officially off limits; consequently all previous examination carried out in a distance from the Kailash mountain. All of these titles showed above have presented that the timing of exhumation of the Gangdese belt consisting of the mountain Kailash was during in the latest Oligocene and the early Miocene.

In addition, this work proves the existence of both old (about 24 Ma) and young age of the rocks (about 10 Ma). These results are consistent with the statement [10] that geochronologic and sedimentologic date indicate three prominent phases of uplift-denudation in the Himalaya: early Miocene (21-17 Ma), late Miocene (11-7 Ma) and Quaternary.

Besides, tectonic evolution of Tibetan Plateau studied by other method has shown that:

- Linking between uplift and initiation of the monsoonal weather system in the Tibetan Plateau shows that the uplift dating as far back as 22 Ma [12].
- The sedimentation rates deduced from seismic profiles from the South China Sea suggest an active monsoon by the early-mid Miocene (11–16 Ma) [13].

Also, Table 3 shows the different authors data about the Kailash formation timing.

Authors	Ages (Ma)	Sample collection places
[14-15]	Late Oligocene– early Miocene (30–17 Ma)	Kailas conglomerate and its equivalent along the Yalu River valley has been estimated at three locations in western, central, and eastern Tibet
[16]	22.3±0.7 to 16.9±0.2	Kailash conglomerates collected adjacent to the Yarlung Tsangpo suture zone in southern Tibet
[11]	23.4 to 16.5; 21.4 to 17.9	Conglomerates collected along the high elevation of the Gangdese belt from east of the Kailas mountain to the west of Saga at elevations ranging between 5100 m and 5750 m
This work	24.6±0.5; 10.1±1.4	Conglomerate around the Mt. Kailash at the height from 5300 m to 5800 m

 Table 3. The Kailash formation timing results of other authors

Conclusion

Nuclear analytical techniques were used to determine elemental concentrations and to estimate the ages of geological samples taken in the expeditions in 2013-2014 years, on different sites of Mount Kailash. The contents of more 30 elements, including Rb and Sr, into geological samples were determined by XRFA, GAA and NAA.

The classification of geological samples and an estimate of their age were conducted on the basis of these results. For some of the samples from the area of Mount Kailash the age was estimated about 24.6 ± 0.5 Ma, younger about 10.1 ± 1.4 Ma. The dating is well fitted to the estimation of other sources.

Until now Rubidium - Strontium dating has traditionally realized by using Mass Spectrometry; this work brings out the potentiality of using nuclear analytical technique to the Rubidium-Strontium dating. It is shown that combination electron accelerators – microtrons and XRFA may be used to determine the age of different geological rocks by Rubidium-Strontium dating method. Additionally, all requirements assure the accuracy of the Rubidium-Strontium dating method should be respected.

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