Characteristic manifestations of uranium enrichment in certain Pan-African granitoids of NW Karbi Hills, Assam (NE India)

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Abstract-*The northwestern edge of Karbi Hills witnessed massive granitoids formation during Cambro-Ordovician time forming a part of late Pan-African magmato-orogeny. The medium to fine-grained, leucocratic, non-porphyritic granitoids contain mafic xenoliths, aplities, microgranite, and quartz veins, invariably saturated with either REE or U and Th, found in Parkop Pahar (PKP), Gufa Pahar (GP), Pulibagan (PULI), and Udmarigaon (UDG) granites. Thin section petrography, supported by powder XRD, reveals a distinct assemblage of U and Th bearing accessory minerals such as uraninite, brannerite, coffinite, thorite, and others, with U and Th concentrations of up to 159 and 121 ppm, respectively. The presence of pitted surfaces in plagioclase and quartz with occasional disseminated alpha tracks, pleochroic haloes in biotites, and/or apatite inclusions in biotites is a miniature scale-model of the regional hydrothermal alteration types and patterns representing uranium mineralization/mobilization. Plagioclase feldspar is the most vulnerable to radiation damage and damage-controlled fluid-assisted alteration, which may redistribute metals, including actinides, in which an alteration sequence Na+—> K+—> H+ has been proposed, integrating U and REE redistribution. The alteration process also included interaction of the hydrothermal fluid with primary U-bearing minerals, inclusions resulting in uraninite resorption, redistribution of elements, including U and Pb, and resetting of isotopic make-up, resulting in the characteristic background ionizing radiation (BIR) profile. The record of aquatic BIR (0.016-0.059) mR/h, total dissolved solids (TDS) values (900-6700) mg/l, and marginal aquatic uranium concentration (<1ppb) establishes the reducing condition of U^{6+} and formation of low soluble U^{4+}minerals with an increase in daughter element concentration of U decay series supported by low pH of water (5.46-5.76).*

Key words: **Karbi Hills, non-porphyritic granitoids, BIR, TDS, pH.**

1. INTRODUCTION

Karbi Hills is unexplored for uranium reserves, although it is reported from the Meghalaya part of the Shillong plateau. The A2 type NW Karbi Hill's granites were formed during post- collisional rifting episode, often rich in REE and/or U-Th bearing accessory minerals. Geochemical attribute suggests metaluminous to peraluminous chemistry, A2 type, within plate character (Majumdar and Dutta, 2016[1]). The mineral composition is dominated by quartz, feldspar, plagioclase, and biotite, with a significant amount of accessory phases such as apatite, zircon, xenotime, monazite, and allanite. The petrography of the studied granites reveals radioactivity damages such as pitted quartz and feldspar surfaces, pleochroic haloes in biotites, the presence of metamict allanite, and alpha tracks on mineral surfaces. The U and Th bearing accessory minerals such as uraninite, brannerite, coffinite, thorite and their alteration products promotes an overall increase in U and/or Th concentration up to 45 ppm (Majumdar and Dutta, 2016). There are reports on a related study on outdoor and indoor background ionizing radiation (BIR) (10.52 mBq/kg/h to 56.29 mBq/kg/h approx.) as well as the determination of radon and uranium counts in residual soil and natural water in Karbi Anglong (Kakati and Bhattacharjee, 2011[2]). According to reports, radioactivity caused by thorium and uranium has been noted in granitoids terrain of Bargaon, Donkamokam, and Teragaon (Karbi- Hills). (GSI Report, 2009). Radionuclides, which act as contaminants, are mobilised by ground water or natural water sources found in granite outcrops (Ahmed et al., 2018[3]). According to Kozowska et al., (2005) [4], high TDS bearing natural water is being polluted by dissolved radio nuclides, which are progenies of Th and U decay series derived from various geological strata. The pH of water is said to control uranium mobility. Hexavalent uranium predominates at low pH (Himri et al., 2000[5]). Natural waters with a pH less than 7 are considered acidic due to a lack of or low concentration of calcite and its close relationship with granitic rocks (Robins, 2002[6]). The interaction of water and acidic (granite) catchment is the primary cause of the low pH level of the natural water.

This paper discusses the issue of high background ionizing radiation (BIR), total dissolved solids (TDS), and aquatic uranium concentration, pH dependence on U and Th concentration, and geochemical basis in the granitic domain of Pan-African magmato- orogenic granites in the NW Karbi Hills, Assam.

2. METHODOLOGY

In the northwest of the Karbi Hills, both terrestrial and aquatic radiations were recorded using Gamma dosimeter Radex RD1503+. This extremely accurate and trustworthy dosimeter can measure in-situ gamma radiation. The device measures the BIR values in units of Sv/y or mR/hr. Until the icon square in the digital display has completed four cycles of four minutes and sixty seconds, the BIR is measured. This radioactive monitor, Radex RD1503+, calculates and evaluates the amount of dose rate of gamma radiation received outside or by a person exposed to radiation in the environment while also taking into account the contamination of objects by sources of beta particles or X-ray emissions. Aquatic radiation recordings are measured primarily at discharging locations of natural water.

A portable MODENNA digital pH meter was used to measure the pH level of the natural water sources in the study area. The electrode is calibrated using buffer solutions with pH 7, 4, and 10 before being used in the field, and it is strictly required to rinse and dry the electrode with distil water or deionized water in between each different solution immersion. Thus, during field use, the pH meter electrode was routinely rinsed with distilled water, sanitised, and dried with tissue paper following each pH reading.

Total dissolve solids measurement(TDS) was done using SEMCO India's water and soil analysis kit. Following the calibration process the TDS readings were eventually recorded after removing the electrode from the distil water and dipping it into the bottles containing the collected water sample.

The laboratory study including the petrography observations and XRD were done soon after the completion of field work. Thin rectangular chips of rock measuring 420.5 cubic cm were cut out of granitoid samples using the in-house facility of the Department of Applied Geology, Dibrugarh University. These chips were being delivered to ONGC geological Laboratory in Sibsagarh, Assam, and Continental Instruments in Lucknow for treatment to a polished thin section. Thin sections were observed using the polarizing microscope Olympus BX51.

A quick analytical method called X-ray powder diffraction (XRD) is generally used to determine the phase of crystalline materials. In an XRD study, the sample to be examined is homogenised and coarsely pulverised to a mesh size of 170 ASTM before the average bulk composition is collected. The Powder X-ray Diffractometer, Rigaku Ultima IV, was used with the help of the Central Sophisticated Instrumentation Centre (CSIC), Dibrugarh University. It is equipped with a copper tube with a wavelength of 1.5418oA and a detector with a xenon filled sealed proportional counter.

3. GEOLOGICAL SETUP

The Meghalaya Craton and the Karbi Hills are separated by the Kopili lineament, often described as the "twin cratons within the Shillong Plateau" (Murlidharan and Desraj, 1983[7]; Bora and Roy, 1999[8]). Both are made up of Proterozoic basement rocks that have been penetrated by magmatic intrusions with various geneses, and it is covered with Proterozoic and Tertiary sediments. The intrusive/extrusive bodies contain at least five episodes of magmatic activity, including an early phase of basaltic composition, the second and third phases of large batholiths of porphyritic granitoids, the fourth phase of small-scale intrusions of dolerites and traps known as the Mikir Traps, and a late phase of alkaline-carbonatite formation. The study domain (Fig.1) carries evidences of evidences like flow banding, breccias with chilled margins having necessary indications of violent intrusion through colder crust in post collision extensional settings. The Pan-African episode leading to massive granitoids formation during Pan-African episode was a sudden and short lived phenomenon, causing invasion of magma through gneissic basement complex, older amphibolites and Shillong Group of supracrustals. These granitoids carry evidences of true volcanic domain, where the granitoids bear degassing vents, vesicles occasionally at their apophyses, either REE or U and Th enriched.

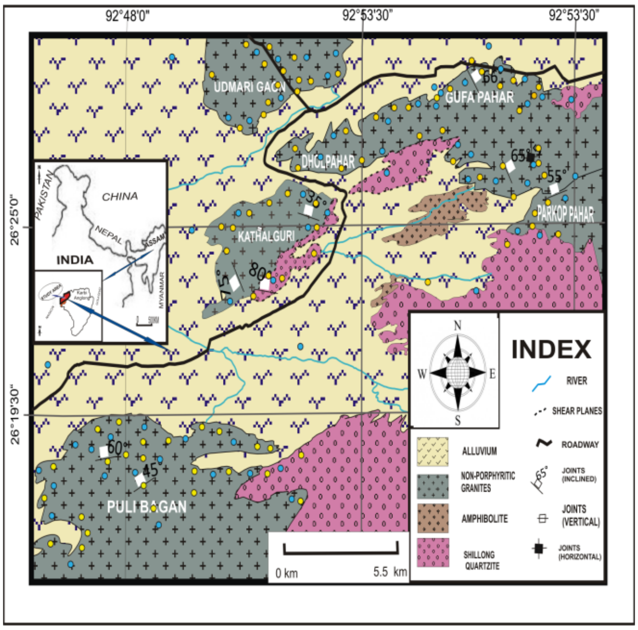

Fig-1: Geological Map of Study Area

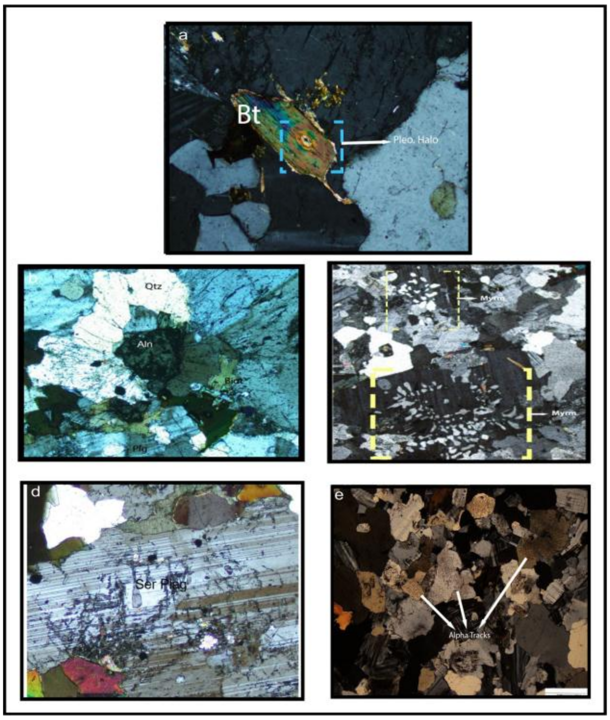

Fig-2: Photomicrographs of granites of NW Karbi Hills showing a) Pleotropic Halo in Biotite b) Metamictisation of Allanite c) Myrmekitic Texture d) Alteration texture observed upon plagioclase grains (sericitisation) e) Alpha tracks

4. RESULT AND DISCUSSION

4.1 Petrography

Presently studied granites are leucocratic, fine to medium grained, salt pepper textured, essentially composed of quartz, plagioclase, with occasional orthoclase, biotites, and microcline. Certain thin sections demonstrate the general indicator for the presence of radioactive nuclide in a granite sample is the radio halo (pleochroic halo) shown in biotite and feldspar minerals centered with euhedral to subhedral uraninite or zircon crystals or seen with empty centered due to leach out of radio nuclides (Fig. 2a), alpha tracks spotted over quartz and feldspar grains (Fig. 2e) and metamict allanites with very fine anastomosing cracks radiating out to the adjacent minerals due to emission of alpha radiations (Fig. 2b). The thin sections revealed the presence of accessory minerals supported by the XRD peaks viz., apatite (1.96), zircon (1.17, 1.80), monazite (1.97, 2.60), xenotime (1.35, 1.83). Primary uranium and thorium phases uraninite (1.60, 1.95, 1.27), thorianite (1.25, 1.14), brannerite (2.71, 3.30), and coffinite (4.93, 3.36, 1.84) peaks are significantly reflected in the XRD (Fig.3). Additionally, K-feldspar and plagioclase frequently exhibit perthite texture and with irregular, wormy blebs of quartz in plagioclase develops myrmekitic texture (Fig.2c), grows simultaneously during the late stages of crystallization in the presence of a volatile phases. Other hydrothermal alteration textures (Fig.2d) like sericitisation and kaolinization have been noted in these granites, marking the alteration sequence Na^{+}—> K^{+}—> H^{+}

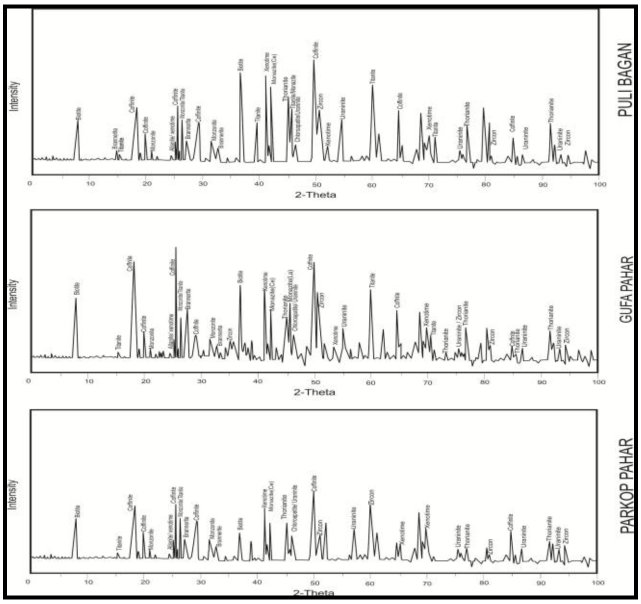

Fig-3: Representative XRD patterns of the constituent mineral phases representing individual plutons a) Puli Bagan b) Gufa Pahar c) Parkop Pahar

4.2 Geochemistry

The bulk geochemistry of the granite samples is indicated in the table (Table1). The granites samples have high SiO_2 wt% that range from 66.76 to 73.95 weight percentages, making them acidic rocks supported by their low MnO (0.06 wt%) and TiO_2 (0.35 wt%) compositions. Al_2O_3 concentrations range from 13.44 to 14.79 wt%, with 14.01 wt% being the norm. The samples' average total alkali concentration (Na_2O + K_2O) is 3.83%, with K_2O being more abundant than Na_2O. The high SiO_2 level of the granite samples indicates that they are substantially fractionated.

Uranium concentrations in the examined samples reach up to 159 ppm, whereas, Th concentrations ranges (0.37- 76.21) ppm and the Th/U ratio is shows an average of 0.59 ppm(Table2). The (Fig.4a) granite samples drops below the worldwide border line ratio Th/U (=4) demonstrates post magmatically enrichment of uranium amid secondary processes (Rogers et al., 1969[9]). The positive interdependence of U and Th demonstrates the control of magmatic processes in the distribution of U and Th (Fawzy, 2017). The uranium mobilisation is represented by an equation U-(Th/3.5). Using the equation, the estimated values for the samples shows greater than zero value (positive value), except a Parkop Pahar granite (PAKP 3) showing negative value -6.94 indicating uranium enrichment.

Major Oxide(wt%) concentrations of NW Karbi Hill Granites														
Sample	**GP1**	**GP2**	**GP3**	**GP4**	**GP5**	**GP6**	**GP7**	**PAKP1**	**PAKP2**	**PAKP3**	**PULI1**	**PULI2**	**PULI3**	**PULI4**
SiO_2	73.95	72.51	66.76	73.64	73.74	73.26	73.79	70.79	70.85	71.46	69.68	70.22	67.05	70.55
Al_2O_3	14.72	13.95	13.44	13.88	13.73	14.48	14.32	14.19	13.8	13.75	13.92	13.9	13.33	14.79
$Fe_2O_3(t)$	1.8	2.44	4.3	1.65	1.71	2.68	2.18	2.94	2.82	2.94	3.23	3.84	4.32	2.3
MnO	0.05	0.06	0.11	0.07	0.06	0.07	0.05	0.05	0.05	0.05	0.06	0.06	0.08	0.06
MgO	0.33	0.39	1.91	0.33	0.25	0.25	0.14	0.34	0.33	0.3	0.48	0.67	1.66	0.41
CaO	1.32	1.21	3.31	1.35	0.88	0.97	0.91	1.14	2.17	2.09	2.03	1.59	3.5	1.36
Na_2O	2.72	3.13	2.42	2.17	2.92	2.08	3.37	4.35	3.31	3.06	3.73	3.49	2.47	3.71
K_2O	4.25	4.38	4.57	4.99	4.75	4.25	4.42	5.19	5.02	4.34	4.94	4.02	4.14	5.23
P_2O_5	0.05	0.07	0.65	0.04	0.03	0.03	0.02	0.05	0.06	0.05	0.19	0.15	0.59	0.09
TiO_2	0.19	0.31	0.73	0.18	0.19	0.2	0.14	0.23	0.23	0.24	0.56	0.56	0.87	0.32

Table-1: Major Oxide (wt%) concentrations of NW Karbi Hill Granites

Elemental concentrations (ppm) of U and Th in terms of studied granites of parts of field areas														
SAMPLE	**GP1**	**GP2**	**GP3**	**GP4**	**GP5**	**GP6**	**GP7**	**PAKP1**	**PAKP2**	**PAKP3**	**PULI1**	**PULI2**	**PULI3**	**PULI4**
U	28.83	22.43	3.95	43.86	55.53	159.10	42.28	20.96	30.72	14.82	3.62	5.75	5.26	16.39
Th	2.72	2.81	0.44	10.02	9.21	54.98	46.69	6.21	4.29	76.21	0.37	1.02	0.50	1.54
Th/U	0.09	0.13	0.11	0.23	0.17	0.35	1.10	0.30	0.14	5.14	0.10	0.18	0.10	0.09
U-(Th/3.5)	28.05	21.62	3.82	3.82	52.89	143.39	28.94	19.19	29.5	-6.95	3.52	5.46	5.11	15.95

Table-2: Elemental concentrations (ppm) of U and Th in terms of studied granites of parts of field areas

Among the rock samples the highest value 143.39, shown by Gufa Pahar (GP6). In the mobility diagram (Fig.4c) the rock samples shows positive value (3.51-143.39) greater than zero specifying uranium enrichment or addition to the rock while if the equation equals to zero it represents absence of any uranium mobilisation. The relations in the diagram indicates a lowering trend of Th/U ratio with uranium mobilisation and post magmatic redistribution in the studied granites which could lead to a significant economic criterion in the north east A2 type Karbi Hill granites (Boyle, 1982[10]) disregarding the lone granite sample, PAKP3.

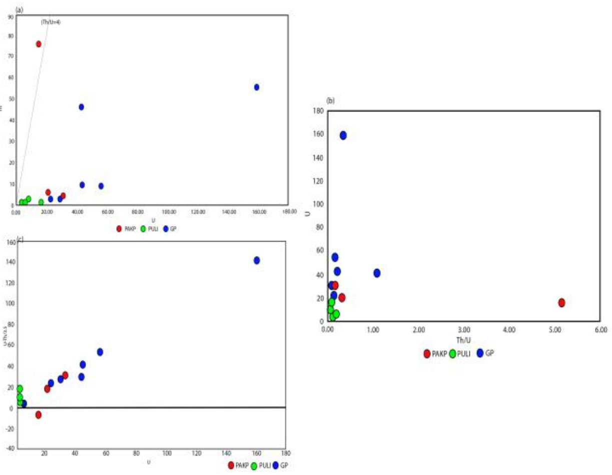

Fig-4: (a) Plotting of NW Karbi Hill granites on the U versus Th variation plot. (b)The U versus Th/U ratio variation plot. (c) The U content versus uranium mobilization equation U-(Th/3.5) plot.

4.3 Spring Water Characteristics

The results of the geochemical properties (pH, TDS, BIR and LED-fluorimetry) of sampled natural water collected from of 16 (ten) field sites were correlated (Table 3). The attempt for correlative analysis of the chemical parameters portrays the direct relation of TDS ranging from 900-6700 mg/l value with aquatic BIR ranging from 0.02 to 0.059 mR/h. The highest TDS measured of samples water of Parkop Pahar also records high aquatic BIR (0.059 mR/h) at the discharging site of spring water. The average TDS value of NW Karbi Hills calculated is 3130 ppm (mg/l). The high aquatic BIR accompanied with increase TDS of the natural water at the field sites may represent significant concentration of daughter radio isotopes of U such as radium (226Ra) that is more soluble in poor oxygen environment and fragmentation of uranium bearing minerals (Ahmed et.al., 2019; Kitto and Kim, 2005[11]).

The pH value recorded during the field indicates a significant decreasing value of pH that records considerable amount of background radiation. The measured pH values (5.51-7.83) shows an important relation with recorded aquatic BIR. Most of the pH values represents slightly acidic to neutral (5.51-6.38) condition of natural water accompanied by a consequential value of BIR. Low pH controls the U mobility, where U4+ gets leached out from the granite rocks and oxidizes to U6+ (Himri et al., 2000). Moreover, the composition of the water, or the result of water-rock interactions related to the solubility of different rock-forming minerals and the kinetics of weathering, also determines the pH condition of natural water (Lasaga et al., 1994[12]; White and Brantley 1995[13]; White et al.2001[14]; Bucher et.al., 2017[15]). Thus, low pH is caused by low $Ca2^{+}$ (calcite) formation or release, low activity acid neutralizing capacity (ANC) chemicals in water, such as HCO^{3-} ions released during cation exchange, or proton-consuming reactions of primary minerals such as K-feldspar, plagioclase, and biotite, which results in a fall in the pH condition of natural water sources. This is because the research area corresponds to an acidic rock catchment region. Presence of high concentration of dissolved radium or radon, the daughter radioisotopes of U decay series in the natural water are favored by low pH (Ahmed et al., 2019) and these leads to significant BIR instead of any uranium content (Kumar et.al., 2017[16]).

Statistical summary and correlation of geochemical properties pH and TDS with background ionising radiation of natural water of north west Karbi Hills.			
LOCATIONS	**pH**	**TDS(mg/L)/ppm**	**BIR (mR/h)**
GPS11	7.52	900	0.016
GPS13	6.9	1000	0.016
GPS14	7.63	1300	0.016
GPS4	7.1	1660	0.017
GPS7	5.7	1700	0.017
GPS1	6.85	2200	0.02
GPS3	7.85	2600	0.02
GPS5	5.76	3000	0.02
GPS6	6.21	3100	0.02
GPS2	6.44	3550	0.021
GPS10	6.21	3600	0.023
GPS12	5.51	4200	0.024
GPS9	6.38	4700	0.028
GPS8	5.5	5100	0.033
GPS15	5.51	5500	0.043
GPS16	5.46	6700	0.059

Table-3: Statistical summary and correlation of geochemical properties pH and TDS with background ionising radiation of natural water of north west Karbi Hills.

The LED-Fluorimetry analysis of the sampled water (Table?) showing uranium concentration <1ppb.In high pH condition, desorption of Uranium from aquifer materials increases with the formation uranium carbonate resulting in high uranium concentration in water. While (Cho et. al., 2019[17]) reported formation of low soluble U(IV) minerals via reduction of U (VI) in a reducing environment that results in low concentration of aquatic uranium concentration.

5. CONCLUSION

1. XRD is used to confirm the identification of primary and secondary uranium phases in thin sections. The radioactive damage symptoms seen in the A2 type Karbi granite are typical of uranium enriched granites.

2. The slightly acidic to neutral pH conditions of the natural water in the study area reflect high TDS as well as significant aquatic BIR. The low aquatic concentration of uranium in the study area indicates a reducing environment due to the reduction of U6+ and the development of U4+ minerals with low solubility.

4. Significant aquatic BIR records may show an increase in daughter radionuclide concentrations of the U decay series, such as radium or radon, supported by low pH with increasing solubility due to a low oxidation environment or reducing condition and fragmentation of U-Th bearing minerals.

5. Identification of primary and secondary uranium phases in the thin sections are being confirmed by XRD. The symptoms of radioactive damage featured in the A2 type Karbi granite typify uranium enriched granites.

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