## Magnetite and ilmenite series granitoids of Ladakh batholith, Northwest Indian Himalaya: implications on redox conditions of subduction zone magmatism

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Multiple felsic magmatic pulses north of the Indus-Tsangpo Suture Zone constitute the Ladakh batholith in the Northwest Indian Higher Himalaya, which is characterized largely by calc-alkaline metaluminous (I-type) granitoids with abundant microgranular enclave formed in a subduction setting. Mineralogical and magnetic susceptibility (MS) parameters of granitoids exposed in the northwestern, central and southeastern parts of the Ladakh batholith have been evaluated in order to understand the redox conditions (magnetite versus ilmenite series) of felsic melts with implication on the nature of the subducting source materials. Average MS values of granitoids range widely from  $0.02 \times 10^{-3}$  to  $44.54 \times 10^{-3}$  SI units, corresponding to both magnetite series (MS >  $3 \times 10^{-3}$  SI units; oxidized type) and ilmenite series (MS  $\leq 3 \times 10^{-3}$ SI units; reduced type) granites, which is broadly consistent with the observed variations in mafic to felsic minerals (low to high), colour index (leucocratic to mesocratic), modal composition (hbl-bt quartz monzodiorite-granodiorite-tonalite), biotite composition (Fe/Fe + Mg = 0.42-0.80) and whole-rock molar Al<sub>2</sub>O<sub>3</sub>/  $CaO + Na_2O + K_2O$  ratio (0.74-1.09) of granitoids of the Ladakh batholith. Although throughout the Ladakh batholith magnetite series granites dominate over the ilmenite series granites, the ratio of magnetite/ ilmenite series granites decreases gradually from the northwestern to the southeastern parts of the Ladakh batholith along its strike-length, which primarily resulted from varying amounts and types of subducting materials and contamination with mantle wedge source. At places, oxidizing condition of granitoid melts becomes elevated because of mafic (enclave) and felsic magma interaction in the open system, and granitoids are locally reduced to ilmenite series close to either intrusive or tectonic contacts.

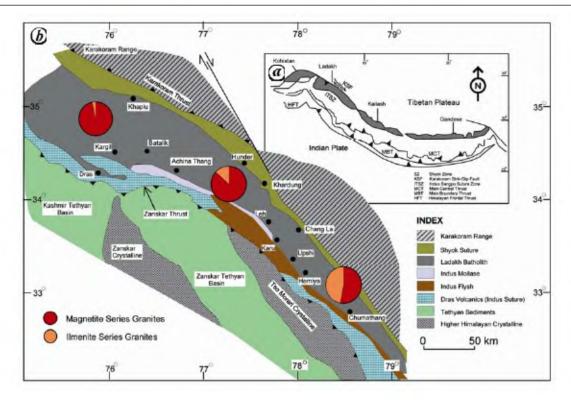
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PRIMARILY granitoids are bimodal in nature, which can be explained in petrophysical terms as magnetic or ferromagnetic, and weakly magnetic or paramagnetic<sup>1-3</sup> depending upon relative modal abundance of magmatic opaque phases, viz. magnetite ± ilmenite in I-type (metaluminous) and ilmenite + sulphide ± magnetite in S-type

(peraluminous) granites<sup>4,5</sup>. However, a leucocratic facies of granitoids, i.e. aplite may correspond to diamagnetic type. Relative abundances of magnetic and weakly magnetic minerals in granites can be measured in terms of magnetic susceptibility (MS in 10<sup>-3</sup> SI units), which is used to characterize the magnetite series (MS >  $3.0 \times 10^{-3}$ SI units) and ilmenite series (MS  $\leq 3.0 \times 10^{-3}$  SI units) granites corresponding to oxidized type and reduced type granites respectively 4,6. MS values primarily indicate redox conditions (oxygen fugacity) of felsic melts commonly inherited from source regions. There may be a parallelism between the magnetite-/ilmenite series and the metaluminous (I-type)/peraluminous (S-type) granitoids8, commonly reflected in species and amount of opaque and ferromagnesian minerals<sup>4</sup>, biotite composition<sup>9</sup>, whole-rock molar  $Al_2O_3/CaO + Na_2O + K_2O$  (A/CNK) ratio<sup>8</sup> and isotopes<sup>6</sup>.

The Ladakh batholith located to the north of the Indus Tsangpo Suture Zone (ITSZ), considered to be formed by subduction-related calc-alkaline magmatism, forms an integral part of the Trans-Himalayan magmatic arc. The Ladakh batholith provides ample opportunity to evaluate the nature of magmatic accretion processes in space and time. Detailed MS mapping of mafic-felsic magmatic lithounits constituting the Ladakh batholith has been carried out recently 10. In the present communication, mineralogical and MS parameters of granitoids from northwestern, central and southeastern parts of the Ladakh batholith have been examined in order to understand the redox conditions (magnetite series versus ilmenite series) of arc-related magmatic evolution with implications on understanding the nature of the subducting-slab (source) materials undergoing partial melting processes in space and time.

Felsic and associated mafic magmatic rocks constitute the Ladakh batholith, which is an elongated body (600 km long and 20-80 km wide), roughly trending NW-SE and covers an area of ca. 30,000 km<sup>2</sup> (refs 11-13). The Ladakh batholith lies to the north of ITSZ and shows gradual shortening in width from northwest (Kargil) to southeast (Chuma Thang) through Leh-Upshi extending laterally in a linear fashion<sup>14</sup> (Figure 1). The Ladakh batholith is characterized by multiple intrusions of variable composition ranging from gabbro-diorite-tonalite to granodiorite-granite, which are in turn cross-cut by mafic dykes<sup>14,15</sup>. Mafic to hybrid microgranular (magmatic) enclaves (ME) and disrupted fragments of synplutonic (or synmagmatic) mafic dykes are ubiquitous, which have equally contributed in the magmatic evolution of the Ladakh batholith<sup>16,17</sup>. Several studies show that the Ladakh batholith is formed by subduction of the Neo-Tethyan oceanic crust beneath the Eurasian plate representing one of the largest Andean-type calc-alkaline magmatic arc found in an orogenic belt<sup>18</sup>. The Ladakh batholith has been formed by at least seven spatially and genetically linked subduction-related, calc-alkaline felsic



**Figure 1.** *a*, Geological structure of Himalaya showing location of Ladakh batholith<sup>38</sup>. *b*, Geological map of Ladakh batholith and associated lithogroups<sup>39,40</sup>. Locations of various places within the Ladakh batholith are also shown, where magnetic susceptibility is measured on the granitoids. Relative abundance of magnetite series (oxidized type) and ilmenite series (reduced type) granites in northwestern, central and southeastern parts of Ladakh batholith is shown as wheel diagrams<sup>10</sup>. See text for detailed discussion.

magma pulses that occurred during ca. 102, 67, 64, 59, 53, 50 and 45 Ma, when the most dominant felsic magmatisms prevailed between ca. 67 and 45 Ma, probably marking the duration of arc-related magmatism propagating from northwest to southeast of the Ladakh batholith 15,18-25.

Granitoids of the Ladakh batholith can be broadly classified into coarse-grained facies with abundant mafic (hbl-bt), medium-grained facies with less abundant mafic, and fine-grained leucocratic facies with very low content of mafic minerals (Figure 2  $\alpha$ –c). They exhibit medium to coarse-grained hypidiomorphic textures with varying amounts of opaque and mafic minerals (Figure 2 d-f). Medium to coarse-grained granitoids contain fine-grained mafic to hybrid ME, which are rare or absent in the leucocratic variety<sup>17</sup>. ME and the felsic host bear common mineral assemblages (hbl-bt-pl-kfs-qtz-ap-zrn-mag ± ttn ± ilm; mineral symbols after Kretz<sup>26</sup>), but differ in mineral proportions and grain sizes<sup>17</sup>. The Ladakh batholith is comprised of calc-alkaline, strongly metaluminous I-type (molar  $Al_2O_3/CaO + Na_2O + K_2O = 0.74-1.09$ ) granitoids, which become relatively less metaluminous in the southeastern parts of the Ladakh batholith (Table 1).

Mafic minerals (bt-hbl) were analysed by the electronprobe micro-analysis (EPMA) technique at Indian Institute of Technology, Roorkee, and whole-rock major oxides were determined by X-ray fluorescence (XRF) at the Wadia Institute of Himalayan Geology, Dehradun. MS measurements of granitoids exposed along various transects (northwestern, central and southeastern parts) of the Ladakh batholith have been carried out on smooth and fresh rock surfaces using SM-20 model Kappameter. MS values (in 10<sup>-3</sup> SI units) were corrected further using recommended factors for degree of rock-surface unevenness: 1 mm - 1.07, 2 mm - 1.15, 3 mm - 1.23, 4 mm - 1.32and 5 mm - 1.41. Mineralogical parameters and average MS values measured at various outcrops of granitoids are summarized in Table 1. Relative abundance of magnetite series  $(MS > 3 \times 10^{-3} \text{ SI units})$  and ilmenite series  $(MS \le 3 \times 10^{-3} \text{ SI units})$  granites<sup>4</sup> corresponding to oxidized type and reduced type granites respectively<sup>6</sup>, has been shown as wheel diagrams placed on the geological map of the Ladakh batholith (Figure 1).

In the northwestern part of the Ladakh batholith average MS values measured at 14 selected outcrops vary from  $3.21 \times 10^{-3}$  to  $27.69 \times 10^{-3}$  SI units (Table 1). About 97% of the observed MS represents the magnetite series and 3% the ilmenite series granites. Relatively lower average MS values  $(3.21-18.3\times 10^{-3} \text{ SI units})$  have been recorded for the granitoids exposed in and around Dras region, which most likely represents local reducing environment of granitoid magma as a result of interaction with volcanics. Granitoids enclosing ME usually record

Table 1. Mineralogical and magnetic susceptibility parameters of granitoids from northwest, central and southeast parts of Ladakh batholith

Parameter	Northwest	Central	Southeast
Texture	Coarse-grained, occasional megacrystic	Medium to coarse-grained	Medium to coarse-grained, occasional megacrystic
Mineral® assemblage	Plg–Kf–Hbl–Bt–Qtz– Mag–Ap–Ttn–Zrn (Hbl > Bt)	Plg-Kf-Hbl-Bt-Qtz-Mag-Ap- Ttn-Zm (Bt > Hbl)	$Plg-Kf-Bt-Qtz-Zrn + Mag \pm Ap \pm Ttn (Bt >> Hbl)$
Mafic/felsic minerals	High	Moderate	Low
Colour index $(M')^+$	Mesocratic	Mesocratic	Mesocratic to leucocratic
Modal composition	Bt–Hbl quartz monzodio- rite, granodiorite	Hbl-Bt quartz monzodiorite, granodiorite	Bt ± Hbl granodiorite, tonalite
ME occurrence	Less frequent	Frequent	Scarce
Average pressure (kbar) (Al-in-hornblende barometer*) Biotite	3.35, 4.27 (± 0.5 kbar)	2.99 (± 0.5 kbar)	2.17 (± 0.5 kbar)
Fe/(Fe + Mg) pfu	0.45-0.80 (N = 21)	$0.42-0.52 \ (N=35)$	$0.42-0.60 \ (N=23)$
FeO <sup>t</sup> /MgO	$1.44-2.19 \ (N=21)$	$1.31-1.90 \ (N=35)$	$1.71-2.63 \ (N=23)$
Buffer stability	FMQ > NNO	FMQ≡NNO	FMQ < NNO
Whole-rock molar A/CNK	0.74-1.04 (N = 9)	$0.88-1.03 \ (N=13)$	$0.90-1.09 \ (N=12)$
Average magnetic susceptibility <sup>#</sup> (× 10 <sup>-3</sup> SI units)	19.80 (19), 3.21 (20), 9.45 (62), 18.30 (16), 23.17 (29), 12.03 (20), 10.85 (19), 14.88 (5), 14.61 (20), 8.81 (20), 19.05 (20), 27.69 (38), 5.98 (6), 13.15 (10)	0.28 (10), 3.86 (14), 11.27 (18), 7.06 (12), 9.75 (10), 9.04 (6), 9.46 (20), 25.82 (20), 44.54 (18), 42.65 (9), 21.98 (15), 0.19 (29), 16.46 (6), 17.63, (15), 15.55 (14), 20.86 (5), 0.81 (14), 13.91 (29), 17.21 (15), 25.95 (6), 28.14 (6),	2.36 (44), 6.30 (8), 0.64 (10), 0.69 (7), 13.18 (20), 10.79 (8), 13.65 (21), 4.45 (10), 9.74 (15), 18.12 (29), 13.96 (29), 12.22 (12), 18.82 (6), 27.15 (10), 17.52 (20), 15.78 (10), 12.40 (29), 15.49 (20), 0.02 (10), 3.99 (13), 0.10 (40), 0.07 (15), 0.02 (10), 0.10 (29),
		25.30 (28), 9.42 (20), 18.13 (21), 11.13 (29), 18.60 (16)	0.14 (32)

<sup>&</sup>lt;sup>®</sup>Mineral's symbols are after Kretz<sup>26</sup>;  $\mathcal{M}' = 100$  – light coloured minerals;  $\mathcal{M}' = 35$ –65 Mesocratic;  $\mathcal{M}' = 10$ –35 Leucocratic (after Le Maitre<sup>36</sup>); ME, Microgranular enclave; \*Schimdt<sup>37</sup>; pfu, Per formula unit; FMQ, Faylite–magnetite–quartz; NNO, Nickel–Nickel–Oxide; Molar A/CNK, Molar Al<sub>2</sub>O<sub>3</sub>/CaO + Na<sub>2</sub>O + K<sub>2</sub>O; \*Kumar<sup>10</sup>.

higher MS values compared to ME-free outcrops, which strongly suggests an increase of oxygen fugacity ( $fO_2$ ) due to mafic–felsic magma mixing and mingling processes that prevailed in the open magma chambers.

In the central region of the Ladakh batholith, the average MS values measured at 26 selected outcrops vary widely between 0.19 and  $44.54 \times 10^{-3}$  SI units, corresponding to both ilmenite and magnetite series granites respectively (Table 1). The observed large MS variations are probably related to modal variations of opaque and ferromagnesian minerals (Table 1). Medium to coarse-grained granitoids intruding the diorite body become strongly reduced to ilmenite series measuring a very low average MS value  $(0.81 \times 10^{-3} \text{ SI units})^{10}$ . Mafic-felsic magma-mingled outcrops characterized by the presence of randomly oriented small mafic to hybrid ME<sup>17</sup> usually reflect high average MS value. Highly oxidized (magnetite series) nature of the granitoids was mostly observed at Khardung La  $(21.98-44.54\times10^{-3} \text{ SI units})$ . Granitoids are reduced to moderately magnetite and ilmenite series towards the northern (Khardung) and southern (Karu) margins, respectively and also observed at some places in and around Pullu and Leh regions. About 87% MS values of granitoids represents magnetite series, whereas 13% belongs to the ilmenite series. The ratio of the two granite series (magnetite/ilmenite series) was thus found to decrease from the northwestern to the central parts of the Ladakh batholith.

In the southeastern parts of the Ladakh batholith average MS values measured on 25 selected outcrops vary from  $0.02 \times 10^{-3}$  to  $27.15 \times 10^{-3}$  SI units (Table 1). Moderate to high oxidizing nature of granitoids has been observed at Sakti  $(4.45-13.65 \times 10^{-3} \text{ SI units})$ , Zingral (9.74- $18.12 \times 10^{-3}$  SI units) and ChangLa  $(15.82-27.15 \times 10^{-3})$  SI units). Granitoids at Karu  $(0.64-2.36\times10^{-3} \text{ SI units})$ , Upshi  $(0.02-3.99 \times 10^{-3} \text{ SI units})$ , Litse  $(0.10 \times 10^{-3} \text{ SI})$ units) and Himiya  $(0.02-1.03 \times 10^{-3} \text{ SI units})$  correspond to highly reduced type ilmenite series. Progressive increase in the reducing (ilmenite series) nature of granitoids has been observed from north (ChangLa) to south (Himiya)<sup>10</sup>. In the southeastern part of the Ladakh batholith, about 56% and 44% of MS values represent magnetite series and ilmenite series granites respectively. The ratio of the two granite series (magnetite/ilmenite series) decreased substantially in the southeastern part compared to the northwestern and central parts of the Ladakh batholith.

Granitoids from the northwestern, central and south-eastern parts of the Ladakh batholith have recorded changing ratios of mafic to felsic minerals (high-moderate-low), corresponding to modal compositions classified as mesocratic bt-hbl quartz monzodio-rite/granodiorite, hbl-bt quartz monzodiorite/granodiorite and mesocratic to leucocratic bt-hbl granodiorite/tonalite respectively (Table 1). Occurrence of ME is less frequent, frequent and scarce respectively, in the northwest

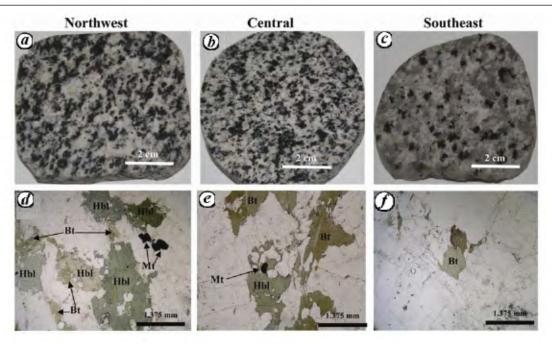


Figure 2. Megascopic features showing textural and mineralogical variations (a-c), and hypidiomorphic textures showing variations in ferromagnesian and opaque oxide (magnetite) contents (d-f) in granitoids from northwest, central and southeast parts of Ladakh batholith.

(P = 3.35 - 4.27 kbar), central (P = 2.99 kbar) and southeast (P = 2.17 kbar), exposing differentially unroofed (deeper to shallower) parts of the Ladakh batholith, as evident from total Al-in-hornblende geobarometer (Table 1). Magnetite and ilmenite series granitoids are primarily governed by the prevailing  $fO_2$  indicated by biotite (Fe/Fe + Mg) crystallized with K-feldspar, quartz and magnetite<sup>9</sup>. As a result of changing  $fO_2$  of felsic melts, biotite in the magnetite series bears Z colour: greenishbrown, is rich in Mg because iron is consumed to form early magnetite rich in  $Fe^{3+}$ , whereas Z colour in ilmenite series biotite is reddish-brown because of Fe<sup>2+</sup> enrichment<sup>27</sup>. Biotite in magnetite series therefore should stabilize above the Ni-NiO buffer and the ilmenite series biotite below it<sup>4,28</sup>. Fe/Fe + Mg ratios in biotites of granitoids from the northwestern (0.42–0.80), central (0.42– 0.52) and southeastern (0.42-0.60) parts of the Ladakh batholith are variable in nature, which is primarily controlled by the relative amount of their stability at various buffers corresponding to FMQ > NNO, FMQ ≡ NNO and FMQ < NNO respectively (Table 1). Biotites in calc alkaline I-type suites are moderately magnesian-rich, with an average FeO<sup>t</sup>/MgO of 1.76, whereas biotites in S-type (peraluminous) suites are siderophyllitic<sup>29</sup> with an average FeO<sup>t</sup>/MgO of 3.48. FeO<sup>t</sup>/MgO ratios of biotites in the northwestern (1.44-2.19), central (1.31-1.90) and southeastern (1.71–2.63) parts of the granitoids suggest dominance of calc-alkaline, magnetite series (I-type) felsic host magmas in the Ladakh batholith, with subordinate amount of ilmenite series (S-type) felsic melts in the southestern part of the Ladakh batholith (Table 1), consistent with the observed MS values.

Felsic magmatism of bimodal nature (magnetite and ilmenite series granites) observed in the Ladakh batholith spans from 102 Ma in the northwestern part to 45 Ma in the southeastern part of the Ladakh batholith, which broadly correlates with a shortening of the batholith width and decreasing ratios of magnetite/ilmenite series granites. Redox state of granitoids gradually changing laterally along the strike-direction of the Ladakh batholith and longitudinally (N-S) in the central and southeastern parts largely appears inherited from contamination of subducted materials with the mantle wedge source, which primarily controlled the redox state of granitic magmas<sup>30</sup>. It has been inferred elsewhere that the most likely source of sea-water sulphate required for magnetite series granites is subducted altered oceanic crust, whereas sedimentary sulphide source materials are needed for the generation of ilmenite series granites, prevalent in the southeastern parts of the Ladakh batholith. Although redox states of granites are intrinsic properties of felsic magma source regions<sup>31</sup>, some granites may acquire oxidation or reduction state due to prevailing physico-chemical conditions of magma chambers<sup>32</sup>, or may occur locally by degassing or assimilation of wall rocks in situ<sup>30</sup>, or deeper-derived lithology with the ascent of felsic magma<sup>33</sup>, or later tectonic and deformational processes<sup>34</sup>, or mafic-felsic magma mixing and mingling processes<sup>35</sup>, or a combination of two or more of the above processes.

In the present study, the observed magnetite to ilmenite series granitoids of the Ladakh batholith were most likely inherited from subducting materials undergoing partial melting process and contamination with mantle wedge source, and to some extent granitoid melts have reduced partially and locally, to varying degrees close to tectonic or intrusive contacts. However, magma mixing and mingling processes that operated in an open system may have also played an important role in elevating the oxidizing conditions (f O<sub>2</sub>) of magma chambers of the Ladakh batholith.

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