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Distribution and occurrence of lithium-bearing minerals in the Helikon pegmatites in Karibib, Namibia.

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ABSTRACT

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Keywords:

Fractionation-Coefficients, Lithium, Mineralization, Pegmatite, Lepidolite, Syn-Collisional Granite, Helikon Lithium is a unique metal and a raw material for the batteries of electronic equipment and electric cars, making it one of the most valuable elements in the Industry 4.0 era. The Karibib Pegmatite Belt (KPB) has been prospected for lithium mineralization since the 19th century. There is however a need for detailed studies of lithium-bearing pegmatites in this area. This research therefore focuses on the distribution and occurrence of lithium-bearing minerals within the Helikon pegmatites (5 and 6), and develops a criterion that can be used for lithium exploration. The Helikon pegmatites are some of the pegmatites that occurs within the KPB. Both Helikon 5 and 6 pegmatites show well developed zonation, with discrete and discontinuous mineralization restricted to areas with well-developed internal zonation. The primary lithium mineralization is characterized by purple and grey lepidolite, with minor amounts of petalite. Furthermore, lithium mineralization occurs as both massive and disseminated at the Helikon 5 and 6 pegmatites, but it also occurs as banded at Helikon 6. ICP-MS analysis determined an overall lithium grade of 3.01%_Li₂O and XRD was used to identify all the lithium bearing minerals. The provenance of the Helikon 5 and 6 pegmatites is found to be from Damaran S-type granites. The fractionation coefficients showed that Helikon 5 and 6 pegmatites are both highly fractionated pegmatites, but Helikon 5 is more prospected for lithium mineralization than the Helikon 6 pegmatite.

1. Introduction

The Karibib Pegmatite Belt (KPB), which lies in the southern Central Zone of the Damara Orogen (Ashworth, 2014), contains valuable Lithium-Cesium-Tantalum (LCT) type of pegmatites. The pegmatites of this belt are subdivided into the Etusis, Kaliombo-Okongava and Dernburg-Abbabis-Naob portions. The Helikon Pegmatites, located 30 Km southeast of Karibib and explored by Lepidico Chemicals Namibia (Pty) Ltd (formerly known as Desert Lion Energy (Pty) Ltd), lies in the Kaliombo-Okongava portion of the KPB. These pegmatites are classified as LCT family following on the petrogenetic classification scheme (Cerny P, 1985), and are characterized by a high degree of alkali fractionation (Stewart, 1978). Most of the pegmatites with the LCT signature have compositional affinity with S-type granites (Cerny et al., 2012). Fractional crystallization of fertile leucogranitic magmas resulted in a regional differentiation and Nb-Ta-Sn enrichment of lithiumcesium-tantalum pegmatites (Hulsbosch et al., 2017). Furthermore, the Helikon Pegmatites Pegmatites intruded into the Swakop Group of the Damaran Supergroup. Table 1 below shows the stratigraphy for the regional geology where the pegmatite occurs. The lithium mineralization appears predominantly as petalite and sometimes lithium mica, spodumene and montebrasite (Knorring, 1985). The pegmatites may have intercalated zones (Diehl, 1990), in addition to the normal core, intermediate, boundary and wall zones.

Table 1. Stratigraphy for the Karibib area, in whichthe Helikon pegmatite occurs (from Ashworth,2014).



The occurrence and distribution of lithium minerals in different zones of the Helikon pegmatites are not well understood at present, due to the lack of detailed studies of lithiumbearing pegmatites in the area. This study aimed to establish the zonation sequence, geochemical characteristics, occurrence and variation of lithium minerals in different zones of two pegmatites, Helikon 5 and 6. Lithium is a unique metal and a raw material for the batteries of electronic equipment and electric cars, making it one of the most valuable minerals in the Industry 4.0 era. The results from this study will be used to establish the distribution of lithium mineralization within the different zones of the Helikon 5 and 6 pegmatites, in order to establish criteria to use the internal zonation as a basis to support LCT-type pegmatites exploration.

2. Materials and Methods

The field work comprised of detailed geological mapping and core logging to establish the stratigraphy, depth and extent of the ore body. Samples were prepared from diamond drill cores for petrographic analysis, in order to study the mineralogy of both the country rock and pegmatites. Petrographic analysis involved the study of microstructures and the mineralogy of both the country rock and pegmatites via thin sections under a transmitted light microscope and polished sections under a reflected light microscope.

A total of 17 samples from diamond drill cores were selected for petrographic analysis, of which 15 (7 from Helikon 5 and 8 from Helikon 6) were prepared for thin sections and 2 (from Helikon 5) were prepared for polished sections respectively. Diamond drill holes HDH046 (drilled from 0– 34.90 m) and HDH061 (drilled from 0-79.50 m) were selected as representative drill holes for the Helikon 5 and H6DH001 (drilled from 0- 122.20 m) was selected as the representative drill hole for the Helikon 6 pegmatites. The drill holes have a spacing of 60 metres apart.

Samples were analysed (courtesy of Lepidico Chemicals Namibia (Pty) Ltd), by means of X-ray Diffraction (XRD), to establish the various mineral compositions and by Inductively-coupled Plasma Mass spectrometry (ICP-MS), in order to determine the concentrations of minor, trace and Rare Earth Elements (REE) and to establish the variation of lithium grades in various zones of the pegmatites.

Geological maps were produced using ArcGIS, and core logging information was used to draw the cross-sections for both Helikon 5 and 6 pegmatites. Fractionation coefficients (FC) adopted from Cerny et al., (1985) were used to infer on the degree of fractionation for the Helikon 5 and 6 pegmatites. Tectonic discrimination diagrams were plotted in order to infer on the possible type of parental granite source for the Helikon 5 and 6 pegmatites. The geochemical analysis was aided by Microsoft excel, in order to produce various geochemical variation diagrams, graphs and plots.

3. Results

3.1 Field observations

3.1.1 Helikon 5

The Helikon 5 pegmatite is a lens-shaped, wellzoned pegmatite body that is continuous along a strike of 480 m. It intruded the marble of the Karibib Formation, and has a strike and dip direction of $048^{0}/62^{0}$ southeast, which almost conforms to that of the country rock ($105^{0}/48^{0}$ southwest).

The zones present in the Helikon 5 and 6 pegmatites are described below based on field observation, the work of Roering and Gevers (1964) and Cameron et al.'s (1949) models for mapping zoned pegmatites.

No visible lithium mineralization was observed within the wall zone. The Helikon 5 has a width of 6 m and it is composed of quartz, albite and muscovite (qtz-alb-musc). Flakes of muscovite are thick (3 cm) and quartz occurs as large, blocky crystals. The intermediate zone is characterized by alternating sequences of perthite-quartz-muscovite with quartz-albite-muscovite zones. The quartz-albitemuscovite zone is characterized by radial muscovite and fine grained garnets, and the perthite-quartzmuscovite zone is characterized by quartz-albite matrix. In addition, the intermediate zone also comprises of lithium ore-zones, containing either massive or disseminated lepidolite

92

Three distinctive lepidolite zones were mapped, of which two are characterized by massive grey lepidolite mineralization, and are intergrown with quartz and albite. The Core Zone, which has a width of 3 m, consists of blocky massive quartz, enclosed by the lithium-mineralized zone.

3.1.2 Helikon 6

Two distinct lens-shaped pegmatite bodies were observed at Helikon 6. They are referred to as Helikon 6A and 6B. Both contain massive and disseminated lepidolite, with banded lepidolite also occurring at Helikon 6B. The Helikon 6 pegmatites intruded calcsilicate and dolomitic marbles of the Karibib Formation. Whereas the Helikon 6A pegmatite is well exposed, Helikon 6B is partially covered by some calcrete and mostly dolomitic marbles. The pegmatites dip northwards (Helikon 6A- 250°/48° northwest, Helikon 6B- 300°/50° northeast), whereas the dip of the country rock is southwards (055°/50°southeast), similar to that of Helikon 5. The Helikon 6A pegmatite is adjacent to a marble intruded by an aplitic granite.

Similar to Helikon 5, the zonation of the Helikon 6 pegmatites are classified as follows:

The wall zone is composed of quartz, with radial muscovites surrounded by albite, and contains no mineralization. The intermediate zones are comprised of the lepidolite-albite-quartz (lep-alb-qtz) and quartz-K-feldspar-muscovite zones. The lepidolite-albite-quartz zone is characterized by banded lepidolite mineralization, of width varying from 3 mm up to 2.5 cm, and intergrown with quartz and albite crystals. Radial muscovites surrounded by K-feldspar are present within the quartz-K-feldspar-muscovite zone, which also contains fine-grained garnets, measuring less than 2 mm in diameter.

The core zone is represented by a blocky mass of quartz and a grey lepidolite zone occurs adjacent to the core zone.

3.1.3 Core-logging

Core logging determined the depth at which the mineralization occurs. Diamond drill hole HDH046 (from Helikon 5) and H6DH001 (from Helikon 6) showed good intersection of mineralization. In addition, it is established that the lepidolite zone is mostly associated with quartz and albite. The quartz-albite-muscovite zone usually forms the hanging and foot wall zones to the mineralization.

3.2 Petrography

The lepidolite zone shows massive occurrence of lepidolite mineralization. Figure 1 shows a lepidolite zone under Plane Polarized Light (PPL) and Crossed Polarized Light (XPL).



Figure 1. Lepidolite zone under A) PPL and B) XPL.

3.3 Geochemistry

- 3.3.1 Lithium Grade distribution
- **3.3.1.1** Lithium grade distribution in various pegmatite zones

Helikon 5 has a generally higher average Li grade (0.9%) compared to Helikon 6 (0.4%). The grade variation with the internal zonation is depicted in figure 2 below. Within each pegmatite, the grade varies with the internal zonation. In Helikon 5, the Li grade is highest in the Lep-alb-qtz zone (1%) and lowest in the Qtz-alb-musc zone (0.1%). The highest grade in Helikon 6 was recorded in the Lepidolite zone (1.8%) whereas the lowest grade was recorded in the Qtz-alb-musc zone (0.4%). Helikon 6 has a wider grade distribution (1.4%), while Helikon 5 has a slightly narrower distribution (0.9%).



Figure 2. Li Grade distribution with zones in Helikon 5 (left) and Helikon 6 (right).

3.3.1.2 Lithium concentrations in petalite and lepidolite

The lithium grade appears to be mostly concentrated in lepidolite compared to petalite. The lepidolite, petalite and the lithium oxide were averaged in every zone, and the lepidolite and petalite concentrations were plotted against the lithium grade (Figure 3), in order to determine the variation of the lithium concentration in every zone. The results were obtained from HDH046, drilled from Helikon 5.



Figure 3. Average concentrations of lepidolite and petalite in relation to the lithium grade.

3.3.2 Geochemical characteristics of Rare Earth Elements (REE)

Helikon 5 and 6 show a similar REE pattern, depicted in figure 4 below. This shows an overall enrichment in Light Rare Earth Elements (LREE) and Lanthanum/Ytterbium ratio (LaN/YbN) ranging from 4.59-10.75, and a depletion in Heavy Rare Earth Gadolinium/Ytterbium Elements (HREE) with (GdN/YbN) ranging from 1.62-2.70. From the pattern, Helikon 5 shows relative enrichment in Strontium with respect to Cerium (SrN/CeN = 5.45) whereas Helikon 6 shows relative depletion in Sr with respect to Ce (SrN/CeN = 0.59). Helikon 5 also shows relative enrichment in Samarium with respect to Neodymium (SmN/NdN = 1.64), whereas Helikon 6 shows the contrary (SmN/NdN= 0.60). The Helikon 5 and 6 REE patterns are compared against that of the Upper continental crust (Figure 4).



Figure 4. Chondrite (Sun & McDonough, 1989) normalized REE pattern for Helikon 5 and 6 pegmatites. The Upper continental crust values are taken from Taylor and McLennan (1981).

3.3.3 Fractionation coefficient diagrams

The Rb/Sr ratio tends to increase with an increase in the lithium concentration. The following fractionation coefficients (FC), Rubidium/Lithium (Rb/Li), Rubidium/Strontium (Rb/Sr), and Niobium/Tantalum (Nb/Ta) are adopted from Cerny et al. (1985), with exception of the Rubidium/Titanium (Rb/Ti) ratio which was plotted as an additional FC. The FCs were plotted in order to infer the degree of fractionation for the Helikon 5 and 6 pegmatites. Figure 5 shows the variations of these FC versus the Li concentration in the different pegmatite zones.



Figure 5. The relationship between Li concentration and Rb concentration (A1-A2), Rb/Sr ratio (B1-B2), Rb/Ti ratio (C1-C2) and Nb/Ta ratio (D1-D2). The ratios are adopted from Cerny et al. (1985).

3.3.4 Tectonic discrimination diagrams

Tectonic discrimination diagrams are used in order to determine the source of the pegmatites. Figure 6 below shows the Rb/Nb + Y and Nb/Y discrimination diagrams. The Helikon 5 and 6 pegmatites both plot in the Syn-Collisional granites (Syn-COLG) field for the Rb/Nb + Y diagram, and in the Volcanic arc granites and Syn-Collisional granites (VAG + Syn-COLG) field for the Nb/Y diagram.



Figure 6. Rb versus (Nb + Y) on the left and Nb versus Y (on the right) tectonic discrimination diagrams (After Joshi et al., 2014). Syn- COLG: Syn-Collisional granites, WPG: Within plate granites, ORG: Ocean ridge granites, VAG: Volcanic arc granites.

4. Discussion

4.1 Field observations

The Helikon 5 pegmatite has an east-west trend, similar to the strike of the Karibib marble in which it occurs. The pegmatite could have been emplaced during the deformation of the Karibib marble. It intruded in a hinge zone, oriented 480 towards southwest. The Helikon 5 pegmatite does not continue beyond the hinge zone, indicating that it may be structurally controlled. On the other hand, the Helikon 6 pegmatite does not appear to be structurally controlled since it cuts across the interbedded zones of the Karibib marble (0550/500 SE). The difference in attitude could be attributed to the fact that the Helikon 5 pegmatite intruded on the southern limb, whereas the Helikon 6 pegmatite has intruded on the northern limb of another fold.

Core-logging data suggests that the lithiummineralization is mainly associated with albite and quartz, which conforms to field mapping. The quartzalbite-muscovite assemblage mostly occurs in the hanging and foot wall zones, as a result these coincide with wall zones. The cross-section of the Helikon 5 pegmatite shows that it is a sigmoidal body; whereas that of Helikon 6 shows that, the pegmatites are typically occurring as dikes.

4.2 Petrographic studies

Petrographic studies have indicated that lepidolite (lithium mineralization) usually occurs with quartz and albite, and this is in agreement with core logging results. Quartz forms the matrix, which is partly replaced by albite with late formed lepidolite crystals. Thin sections of the lepidolite-albite-quartz zones have disseminated lepidolite, whereas the lepidolite zone generally contains massive lepidolite with minor quartz and albite. Muscovite usually occurs with quartz and albite within the border zones and is generally absent in lithium rich zones. The guartz core formed from silica rich hydrothermal fluids that intruded at high temperatures. The sulphides contained in the marble probably indicates hydrothermal overprint of the Helikon 5 and 6 pegmatites (Geo Experts Consulting Services CC, 2018).

4.3 Lithium grade distribution

The highest lithium concentration is associated with quartz-albite-muscovite zones at Helikon 5 and the lepidolite zone at Helikon 6. These zones proved to be the dominating lithium mineralized zones (Figure 2). Figure 3 shows that lepidolite is positively correlated with lithium concentration. The figure also shows that the lepidolite-albite-quartz zone in the Helikon 5 pegmatite has the highest lithium concentrations (2.20 %_Li2O); whereas the lowest values (0.31 % Li2O) occur in the quartz-albitemuscovite zone. Significant lithium concentrations are mainly attributed to the presence of lepidolite (which has an average concentration of 35%) in the lepidolite-albite-quartz zone, whereas petalite occurs in very low concentrations (which has an average concentration of 1.07%), such that it has little effect on the overall lithium grade.

4.4 REE distribution patterns

The REE distribution pattern for both Helikon 5 and 6 pegmatites shows a similar enrichment in LREE and depletion in HREE (Figure 4). The Helikon 6 pegmatite generally has a higher enrichment in REE concentrations relative to the Helikon 5 pegmatite. Their LREE pattern is the same, but Helikon 6 shows a much higher enrichment in HREE compared to the Helikon 5 pegmatite. These patterns show a similar trend to that of the Upper continental crust, which indicates that rocks of the upper continental crust are the source of the lithium in the Helikon 5 and 6 pegmatites. This is in agreement with London (2008), that the sources of LCT pegmatites are undepleted upper- to middle crustal S-type rocks.

4.5 Fractionation coefficient diagrams

The Rb versus Li concentration (Figure 5A1-A2) yield a well-defined correlation for both Helikon 5 and 6, with Li concentration increasing with an increase in the Rb concentration. Rb typically behaves like Potassium (K), and it remains in the melt (Barrat, et al., 2012), crystallizing only at a later stage. The lithium incorporation may be eased by an increase in the substitution of K by Li (Clark & Cerny, 1987), therefore lithium becomes incorporated in early crystallizing phases (including plagioclase) before its accumulation reaches the level sufficient to trigger the crystallization of lithium minerals. The bulk of lithium then becomes incorporated into enriched micas (Cerny et al., 1985), such as lepidolite. This is in agreement with both Figure 5 A1 and A2, which show the highest lithium concentration in the lepidolite-containing zones.

There is a positive correlation between Rb/Sr ratio and the Li concentration for Helikon 6 (Figure 5 B2), with the highest Rb/Sr ratio recorded in the lepidolite zone. During crystallization of the pegmatite, Sr is removed from the melt and is built into plagioclase and partly K-feldspar (Geo Experts Consulting Services CC, 2018). This triggers the Rb concentration to be higher than that of Sr in late melts, because Sr is more compatible than Rb and it therefore decreases with an increase in the degree of fractionation. As a result, the higher the Rb/Sr ratio, the more differentiated the zone (Geo Experts Consulting Services CC, 2018). The Li concentration therefore increases with the degree of differentiation, for it is positively correlated to the Rb/Sr ratio which is in turn positively correlated to the degree of fractionation. An explanation for this is that although Li has a suitable ionic radius, it has a low charge such that it is excluded from earlier crystallizing minerals (Cerny et al., 1985). It is therefore enriched in the melt and its concentration in the melt increases with the degree of differentiation.

Conversely, the zones with high Li concentration in Helikon 5 generally recorded a low Rb/Sr ratio (Figure 5 B1). In the highly differentiated zones (with high Li concentration) the Sr concentration is high (reversed behavior), such that the Rb/Sr ratio is low. In less differentiated zones (with low Li) the Sr is low (reversed behavior), such that Rb/Sr ratio is high. Sr behaves abnormally in highly fractionated because its pegmatites, concentration is exaggerated by the radiogenic ⁸⁷Sr in Rb-rich fields (Cerny et al., 1985). Clark and Cerny (1987) also agree to this, adding that highly differentiated Rb pegmatites may consist predominantly of radiogenic ⁸⁷Sr, which obscures the original relationship of Rb to common Sr at the time of crystallization.

This indicates that the Helikon 5 pegmatite is more highly differentiated than the Helikon 6 pegmatite, such that its Sr behavior is reversed. This is also in agreement with the average Li grades of the two pegmatites, with Helikon 5 having a higher average grade (0.9% Li) than Helikon 6 (0.4% Li). Highly fractionated pegmatites can therefore be said to be generally more prospective for Li than less highly fractionated pegmatites.

The Li concentration is positively correlated to the Rb/Ti ratio (Figure 5 C1-C2). High Rb/Ti ratios indicate high degrees of differentiation, since Rb is enriched in late melts while the enrichment of Ti in late phases is very restricted (Barrat et al., 2012). Consequently, the incorporation of Rb in lithophile micas will be higher than that of Ti, hence high Rb/Ti ratios will be associated with zones such as the quartz-albite-muscovite zone in Helikon 5 and the Lepidolite zone in Helikon 6 pegmatites.

Nb becomes available for crystallization of solid phases in early stages relative to the bulk of Ta, leading to extensive ranges of the Nb/Ta ratio (Figure 5 D1- D2), even in a single body of pegmatite (Cerny et al., 1985). The higher the degree of fractionation, the more Ta (vs. Nb) remains in the melt, and Ta accompanies Li mineralization (Geo Experts Consulting Services CC, 2018). Consequently, the Nb/Ta ratio decreases with increasing degree of fractionation. The Li concentration in both pegmatites (Helikon 5 and 6) behaves counter to the Nb/Ta ratio. This is attributed to the fact that the Li concentration increases with an increase in the degree of fractionation, whereas the Nb/Ta behaves opposite to this.

97

Therefore, the Nb/Ta is low in zones that contain high Li concentration (such as the lepidolitealbite-quartz zone for Helikon 5 and the lepidolite zone for Helikon 6 pegmatite) and the opposite for zones with low Li concentration (such as the quartz-albite-muscovite zones for both Helikon 5 and 6 pegmatites).

4.6 Tectonic discrimination diagrams

The Helikon 5 and 6 pegmatites both plot in the field of Syn-COLG on the Rb/ Nb + Y and in the VAG + Syn COLG field on the Nb/Y tectonic discrimination diagrams respectively (Figure 6). This suggests that both pegmatites have a similar source, which could be either S- or I-type granites. S-type granites are derived from melting of upper continental crustal rocks (meta-sediments), linked to over thickening of continental crust during collision (Bradley & McCauley, 2013). The inferred peraluminous, S-type granitic parent coupled with calc-alkaline nature and Syncollisional + VAG of these pegmatite signatures indicate that the parent granitic melt and consequent pegmatitic bodies could be related to subduction processes (Joshi et al., 2014). Most LCT pegmatites are late syn-tectonic to early posttectonic with respect to enclosing rocks (Bradley & McCauley, 2013; Xu et al., 2020). Although Itype granites can also form during collision or subduction, they are derived from melting deep crustal igneous rocks. With the pegmatites having an REE plot indicating an upper continental crustoriginating source, the two pegmatites are therefore sourced from S-type granites rather than I-type granites.

During magmatic fractionation, Li does not behave as a typical alkali element, and during consolidation of granitic intrusive bodies, exclusion of Li from crystallizing minerals leads to its accumulation in residual magmas (Cerny et al., 1985). The degree of fractionation plays an important role in constituting significant concentrations of Li in the lepidolite-albite-quartz and lepidolite zones respectively. The fractionation coefficients showed that both the Helikon 5 and 6 pegmatites are highly fractionated pegmatites, with the Rb/Sr ratio pointing to the Helikon 5 pegmatite as the more highly fractionated pegmatite. These fractionation coefficients can therefore be used as basis for Li exploration. Extreme fractionation is necessary for pegmatite melts to become saturated in lithium concentrations. The Helikon 5 pegmatite is more prospective for Li mineralization than the Helikon 6 pegmatite.

5. Conclusion

The fractionation coefficients showed that both the Helikon 5 and 6 pegmatites are highly fractionated pegmatites, with the Rb/Sr ratio pointing to the Helikon 5 pegmatite as the more highly fractionated pegmatite. These fractionation coefficients can therefore be used as basis for Li exploration. Extreme fractionation is necessary for pegmatite melts become saturated lithium to in concentrations. It can therefore be concluded that the Helikon 5 pegmatite is more prospective for Li mineralization than the Helikon 6 pegmatite.

The REE patterns of the Helikon 5 and 6 pegmatites show a similar trend to that of the upper continental crust. This shows that they are sourced from upper continental crustal rocks. This is in agreement with the tectonic discrimination diagrams, which indicate the Syn-collisional granites as the major source for the two pegmatites. The S-type Damaran granites are the parental rocks of the Helikon 5 and 6 pegmatites.

98

The results obtained through this research are significant for lithium exploration, as this can be used as a basis to determine the occurrence and distribution of lithium minerals within different pegmatite zones. It also helps to make an inference on the source of these pegmatites, which can help in explaining their signature or characteristics.

The geochemical analysis of this research was limited to REEs, minor and trace elements. The author therefore recommends further study on tracing the parental rocks of these pegmatites, by age-dating and assaying major elements of Damaran S-type granites, and relating them to the signature from these pegmatites.

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