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Formation of Clays and Chlorites in the Upper Icelandic Crust

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ABSTRACT

Zeolites, clays and chlorites form the basis of the alteration zonation in the upper part of the Icelandic geothermal environment. Detailed studies using XRD, binocular and petrographical microscope data from Nesjavellir high-temperature field in SW-Iceland show that the dominant clay formation occurs as smectite within the smectite-zeolite zone both as direct precipitation as well as the alteration of volcanic glass. Near the base of that zone a distinct coarse-grained clay crystallizes overlying the fine-grained clay lining in the voids. This clay crystallization occurs at same depth as zeolites become unstable and appear to grow from underneath the gradually dissolving zeolites. The final alteration of the zeolites is represented by their replacement mainly into quartz and less wairakite. The progressive hydrothermal alteration is dominantly caused by the suppression of the volcanic strata into the normal geothermal gradient but can also be affected by the propagation of the high-temperature system into the former. This causes the gradual transformation/recrystallization of the already formed smectite clays into mixed layer clays and later into chlorites. Clay analysis show that smectites and mixed layer clays persist to much higher temperatures. Detailed studies combining geological structures and permeability variation within the geothermal reservoir show that their persistence to higher temperature is closely linked to low permeability, while their transition is fast where permeability is high.

1. INTRODUCTION

Hydrothermal alteration is the reaction between a geothermal fluid and the rock it percolates through and is heavily dependent on temperature, rock/fluid composition and degree of porosity and permeability. The use of hydrothermal alteration is highly useful in geothermal exploration and utilization as it pictures the longtime condition of the respective geothermal system and often mirrors the fluid geochemistry, resistivity structures and many other geophysical parameters used during exploration (e.g. Kristmannsdottir 1979, Gunnlaugsson 2004, Gíslason et al. 2005, Ármannsson 2012, 2016, Árnason et al. 2000).

Hydrothermal alteration is to a large extent a function of the geothermal gradient within the volcanic rift zone and outside the central volcanic provinces is estimated to be around 100°C/km outside the high-temperature areas but more than 200°C/km within. (e.g. Flóvenz and Sæmundsson 1993).

The highly temperature dependent alteration is well documented in the eroded tertiary succession first described by Walker (1960) and Kristmannsdóttir and Tómasson (1976a, 1976b), Kristmannsdóttir (1979) and Tómasson and Franzson (1992). Similar studies have been done outside high-temperature areas in the volcanic zone (e.g. Tómasson and Franzson 1992). The main emphasis for this zonation was to trace the progressive change in the zeolite mineralogy from low-temperature ones such as chabazite, thomsonite to stilbite, scolecite, and to mordenite and lastly laumontite as the highest temperatures, which lastly is replaced by wairakite zeolite at temperature higher than 200°C. The essential feature is that when a volcanic layer moves downwards due to the overlying volcanic accumulation it passes through progressively higher temperature and consequently thermal alteration. This gradual progressive change is expected to be interrupted where a high-temperature system "intrudes into" the former. However, after the high-temperature system is established, this progressive change continues in line with the accumulation rate.

Effusive basaltic rocks show relatively elevated porosity due to the high vesicularity formed as a result of trapped exolved magmatic gases during consolidation (Sigurðsson et al. 2000, Franzson et al. 2001). This high vesicularity is the prime locations for the mineral deposition during the progressive increase in alteration. Basaltic rocks are also relatively reactive within the geothermal environment in comparison with many other such locations in the world where trachytic compositions dominate. Several studies have focused on the mineral assemblages in vesicles and veins (e.g Fridleifsson 1983, Franzson 1983, 1987, 1994, 1995, 2000, et al.2002, et.al. 2010a) and in particular the sequence in which they deposit, often with the assumption that the oldest minerals lie at the margin of the void and the youngest in the central part. In this paper the mineral sequence is reconsidered with respect to the relation between the disappearance of zeolites and the deposition of coarse grained clays. Further we will also focus on the formation of clays within the geothermal system in relation to other minerals. This involves both clays as alteration product of the primary minerals in the basaltic rocks as well as its deposition within voids.

The cuttings retrieved from the wells studied are analyzed by mainly three methods where each sample is analyzed through binocular microscope followed by petrographic analysis of selected samples and lastly samples are selected for clay analysis in order to establish alteration zonal boundaries. The mixture of these three methods establishes a comprehensive picture of the rock types and alteration. There is, however, a basic difference between XRD-clay analysis versus binocular and petrographic analysis. The former involves grinding the sample, sorting out the clay and analyzing indiscriminately the whole clay content. This is opposed to the petrographic and partly the binocular analysis where one can assert the origin of the clay, such as whether a deposition or an origin from alteration product of the primary constituents. Furthermore, the latter can estimate the clay type and crystallinity. The clay hydrothermal zonation used in the Icelandic environment has been determined largely by the XRD-method, where the uppermost occurrence of the specific clay determines the boundary. In latter years, other minerals have also been considered in zonal boundary determination.

Studies of permeability within high-temperature fields are very important in exploration and a special attention has been given on finding the relation between aquifers and geological features. This has made it possible to attach changes in alteration with variations in permeabilities as a separate variable in addition to temperature. This is very important with respect to variations in clay mineralogy (e.g. Franzson 1983, 1987, 1994, 1995, 2000, et al.2002, et al.2010a).

The process of subsidence of volcanic rock within the rift zone was first put forward by Pálmason (1971). The accumulation and subsidence rate within the rift zone is variable depending on whether in- or outside the central volcanic complexes. This accumulation rate has been assessed in tertiary regions (Harðarson et al. 2008) and range from 400-2000 m/Ma. Estimates on successions belonging to the western rift zone seem to lie around 1000 m/Ma. Estimates on accumulation rates within the Hengill central volcano, based on number of glacial hyaloclastite horizons, seem to show somewhat higher values (Franzson 2000) where the bottom of the stratigraphy belonging to the central volcano is found at about 600 m depth which is the base of the third/fourth last glacial (300-400x10^3 y).

In this paper we will first give a short introduction of the geological and alteration character of the Nesjavellir high-temperature area followed by a more detailed focus on the relationship between clay formation, temperature and permeability and the depositional trends in the voids. The data presented is based on the research done on the first eighteen wells drilled in the area, but here we focus mainly on the data from wells 6 and 7 which were studied in the greatest detail (Franzson 1994).

NESJAVELLIR HIGH-TEMPERATURE AREA

Nesjavellir is located in the northern part of the Hengill central volcanic complex in the western volcanic zone in SW-Iceland (figure 1). The geological cross section through the southern part of the geothermal system is exemplified in figure 2. The succession is dominated by lava series and hyaloclastites. The latter which dominates down to about 350 m b.s.l. is interpreted to represent highland condition in line with the build up of a central volcano, while the underlying lavas imply low altitude landscape predating the volcano. This puts an age restriction to the volcano at 300-400 x 10^3 y and thus the maximum age of a high-temperature activity (Franzson 1994, 2000). This demonstrates how volcanic accumulation gradually suppresses the rock units into the underlying increasing geothermal gradient.

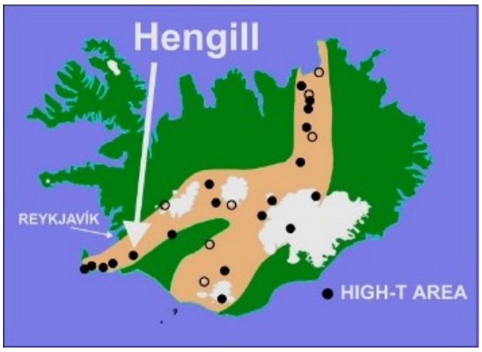

Figure 1. Simplified geological map showing the volcanic rift-zones, location of Hengill volcano and other high-temperature areas.

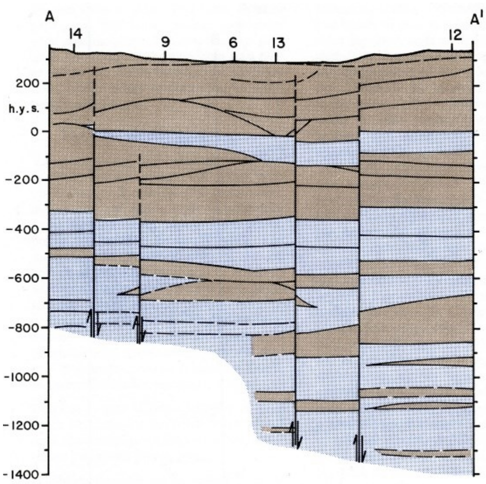

Figure 2. An E-W geological cross section through Nesjavellir high temperature area, showing the main stratigraphical units where brown colour indicates glacial hyaloclastites and blue sub-areal lava series.

Figure 3 shows the alteration zonation through the same cross section. The geological structures controlling the permeability in the upper part of the geothermal system is either controlled by sub-horizontal stratigraphic boundaries or by vertical fault structures, and this is clearly observed in figure 3. Strong evidence is found of higher permeability near the upper boundaries of the alteration zones indicating the connection between alteration and permeability (Franzson 2000).

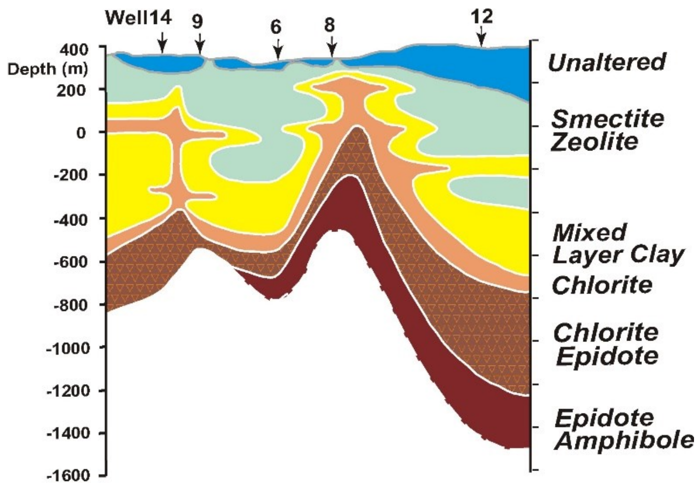

Figure 3. The same E-W cross section as in figure 2 showing the hydrothermal zonation deduced from detailed alteration studies.

THE FORMATION OF CLAYS.

Clays are assessed using XRD, binocular and petrographic methods as discussed above. In the early eighties clay crystallization became a part of the routine observations in the cutting analysis in boreholes (Franzson 1983), mainly done through the microscopes. A comprehensive study was done in well NG-6 at Nesjavellir, and the results are shown in figure 4 below, plotted against depths of observations. The left side of the picture shows the three main successive clay depositions in voids. Amorphous clay lining is the first to form and that occurs as soon as the rocks enter the smectite-zeolite zone. At slightly deeper levels indications are seen that the clays are becoming slightly radial in shape. In well NG-6 the third clay episode occurs with coarse radial crystallization lying on top of the finer clay. This third episode of clay is subdivided into three parts on grounds of petrographical (and from binocular microscope) characteristics. In the top part, or from about 530 down to about 720 m, the coarse grained brownish coloured clay dominates with some pleochroism and is termed as smectite. Then at about 700 m this clay obtains higher order colour and develops a very high pleochroism. This clay type, which is the mixed layer clay, predominates down to the bottom of the well. The third type is the coarse-grained clay where the clay becomes grayish in colour in cross polar and the pleochroism disappears with a faint green colour in plane polarized light. This clay is termed chlorite. What is of particular importance is that clays can only be seen depositing in these three episodes, but the petrographical changes we see in the third episode is a transformation of this fine- and coarse-grained clay from one type to the other. Indeed, as this transformation proceeds in the coarser clay, the same petrographical changes are seen in the finer underlying clay types. In the central part of figure 4 the results of the XRD analysis is shown for comparison. There, smectite is the only clay down to 550 m though it extends further down to about 650 m. Mixed layer clay and swelling chlorite starts to appear at about 550 m and becomes dominant below 650 m down to about 900 m where first signs of chlorite appears along with MLC to the bottom of the well. There is a close correlation between the petrographic character of the clays and the XRD analysis given the basic difference between the methods used where XRD gives the dominant clays present while the former gives the more specific view to individual crystals.

Figures 5a and 5b show a petrographic (plain- and cross-polarized) view of the crystallization of the very pleochroic coarse-grained clay on top of the finer grained one (mixed layer clay), and then figure 6 shows the non-pleochroic clay (chlorite).

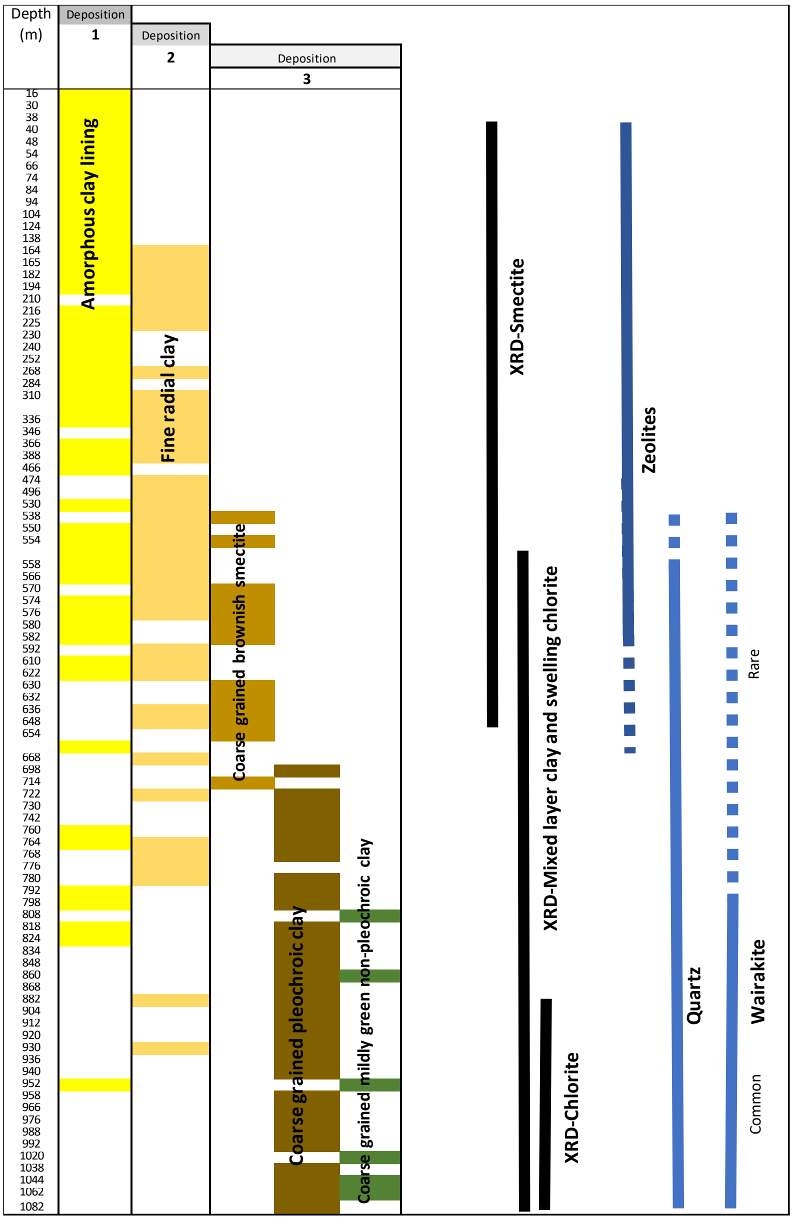

Figure 4. Nesjavellir, well NG-6. Binocular- and petrographic observations on clay deposition in voids with depth to the left in comparison with XRD-clay analysis and then the relative abundance of zeolites, quartz and wairakite.

Fig. 5A

Fig. 5B

Figure 5. A part of a vesicle filling showing early opal/chalcedony (later altering to quartz) lining the void followed by finely crystallized clay and coarse-grained high-coloured radial clay. A. plane polarized light, B. Cross-polarized light.

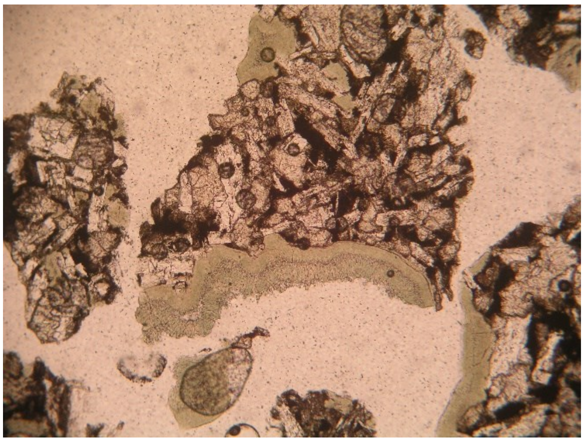

Figure 6. Part of a vesicle filling showing a sequence of finely crystallized clay followed by a coarse clay. Both types show faint green colour indicating its transformation into chlorite.

LOWER BOUNDARY OF ZEOLITE AND ITS RELATION TO CLAY DEPOSITION

Literature on zeolite distribution and zonal boundaries in basaltic crust is well documented as described above and is an integral part of the alteration analysis in geothermal wells in Iceland. Their ease of recrystallization into higher temperature varieties as the volcanic accumulation is suppressed to deeper levels is well documented and clear and bear evidence of their sensitivity to reaction. The bottom of the zeolites is less studied though, in particular where they disappear or change to wairakite which is reported to occur at around 200°C (e.g. Kristmannsdóttir and Tómasson 1976b). The bottom of the zeolite zone has been studied petrographically in detail at Nesjavellir and the relation between zeolites and the accompanying alteration void minerals. Figure 4 above shows the zeolite distribution where their abundance starts to decline below about 550 m depth in well NG-6 and disappear altogether below about 650 m. Of importance is to observe that quartz appears at about 540 m and becomes common below 560. Wairakite is first seen at a similar depth to quartz but only becomes a common occurrence below about 800 m depth. Figures 7 and 8 show quartz being formed from laumontite and from a finely radial zeolite. These observations confirm the close relationship between the disappearance of the latter and the formation of quartz. Table 1 summarizes the occurrence of zeolite alteration into other minerals derived from petrographic observations in four wells at Nesjavellir.

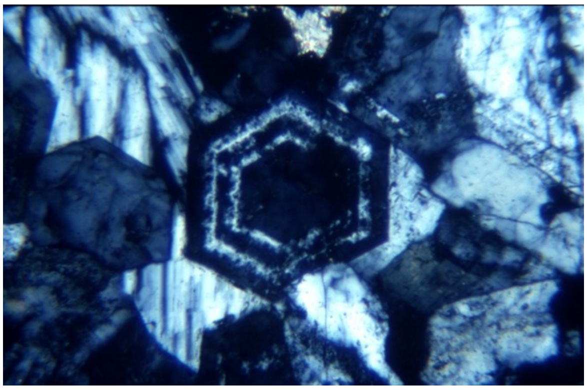

Figure 7. Alteration of laumontite into hexagonal quartz. Note the laumontite remains within quartz growth zones.

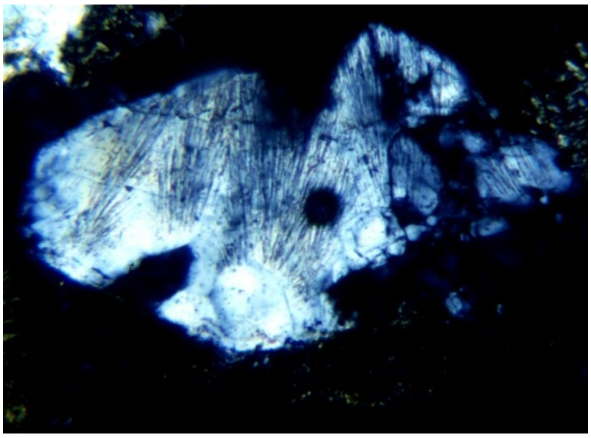

Figure 8. Alteration of fine radial zeolite into quartz.

The most conspicuous change is the alteration of zeolites into quartz which is found in more than half of the observations, most commonly as an alteration of fine radial zeolites (mesolite/mordenite?) and laumontite. Wairakite alters zeolites, in particular stilbite and laumontite, but is not as common as further seen in figure 4 above. A few examples are found of zeolite alteration into prehnite and calcite, but it should be noted that those two minerals relate closely to the last depositional phases within the geothermal system (Franzson 2000). Lastly, but importantly, there is a clear indication of laumontite being replaced by clay. We have also noted clear evidence of coarse-grained clay penetrating the base of the overlying zeolite indicating the growth of clay underneath the zeolite.

Table 1. Alteration of zeolites in voids at Nesjavellir high-temperature field (Franzson 1994).

	Laumontite	Quartz	Wairakite	Prehnite	Clay	Calcite
Silbite	2	6	4			
Fine radial zeolite		11	1			
Mordenite	1	5				1
Mesolite		3				1
Scolecite		2	1			
Laumontite		9	7	1	9	
Undefined zeolite		1	1			2
Analcime						1
"Total"	3	37	14	1	9	5

It is interesting to observe the progressive clay crystallization in figure 4 and in particular that the coarse-grained clay starts to form in the zone where the zeolites are disappearing. But in no instances do we observe the zeolite/quartz alteration underlying the coarse clay deposition or indeed the presence of quartz or wairakite in that position, which conclusively suggests that the coarse-grained clay is depositing prior to the above transformation and must occur near the initiation of the zeolites disappearance.

Figure 9 is the distribution of selected alteration assemblage in well NG-7 at Nesjavellir and shows a very similar alteration relation as in well NG-6 where zeolites diminish rapidly at the top of the mixed layer clay zone accompanied by the appearance of quartz and wairakite. Of importance is to note the continual appearance of smectite to deeper levels and mixed layer clays down to about 1000 m depth.

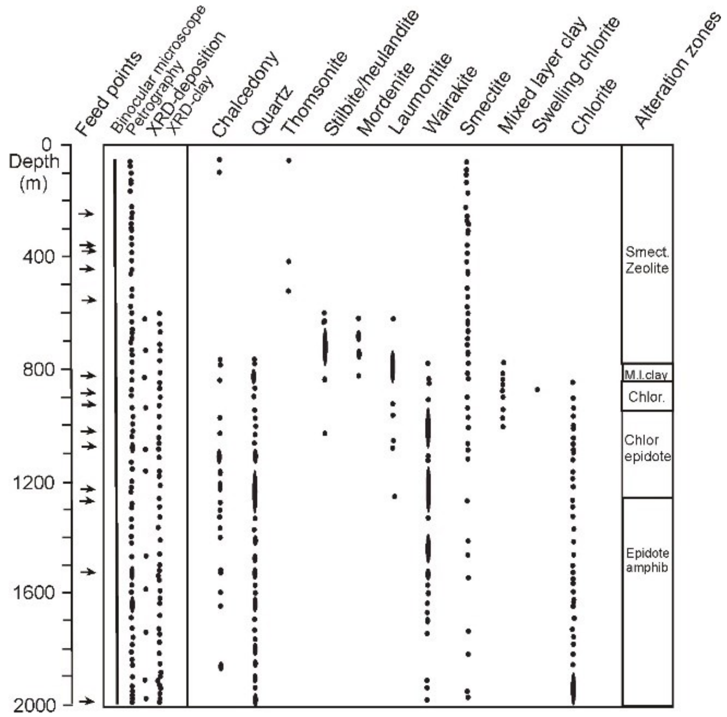

Figure 9. Selected alteration mineral distribution in well NG-7 at Nesjavellir showing feed-points, analytical methods and frequency, selected alteration minerals distribution (zeolites and clay minerals) and alteration zonation.

The overall deposition/transformation trend that we can now establish with respect to clays zeolites quartz and wairakite is as follows:

1. Clay smectite lining at the boundary of voids possibly followed by finely crystallized smectite. Silica is sometimes underlying the smectite (opalline silica later transforming to quartz as seen in figure 5).
2. Zeolites zonation with depth with increasing temperatures up to around 160-180°C.
3. Coarse-grained clays depositing at the base of the zeolites.
4. Transformation of the remaining zeolites mainly into quartz, wairakite and even to clays.
5. Transformation of the precipitated clays into mixed layer clays and lower down into chlorites.

The cross section of the hydrothermal zones along the same line as the lithology is shown in figure 3. The relatively complex zonal boundaries shown there is in some respects the result of the observational detail of the research. The detailed mineralogy was compared closely with feed zones in the succession which made it possible to project the zonal boundaries more accurately by taking geological structures into the model. A clear relationship is found of zonal boundaries and permeability, which demonstrates how important the latter is with respect to the clay transition.

CLAY ALTERATION OF GLASS AND PRIMARY MINERALS

Primary minerals and volcanic glass show a very variable sensitivity to hydrothermal alteration. Basaltic glass is probably the most sensitive constituent. It, however, shows two stages. The first one is the palagonitization of the glass which involves hydrolysis and chemical exchange which most likely occurs at the same time as the volcanic eruption. This palagonite has composition approaching that of smectite composition (Franzson et al. 2010b). This results in a rapid alteration into smectite as soon as it reaches into the smectite-zeolite zone. The second stage is the alteration of the un-palagonitized glass which shows a much more resistance to alteration. Although most of the glass has altered roughly halfway down into the smectite zeolite zone, a small portion of it remains fresh all the way down to the upper boundary of the mixed layer clay zone. The dominant alteration of glass is smectite, but other minerals are also common such as silica, zeolites and calcite, which often relate to location of permeable zones in the geothermal system. Olivine is similarly sensitive to alteration and has completely altered above the mixed layer clay zone into clay and iddingsite. Alteration into other minerals is also observed but less common. Plagioclase is more resistive to alteration and first signs of its instability is micro-fracturing, mainly seen in phenocrysts, filled with chlorite as defined petrographically (figure 10). This alteration starts in the lower part of the smectite zeolite zone and the mixed layer clay zone. This alteration is though only a minor part of the alteration, as further down the albitization of the plagioclase dominates along with other minerals such as calcite and wairakite.

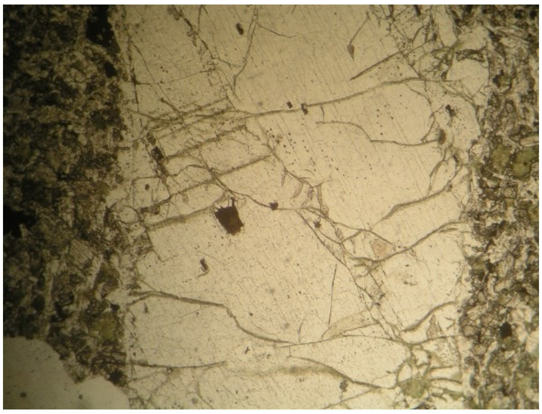

Figure 10. Plagioclase phenocryst in plane polarized light showing thin fracture fillings of chlorite like clays.

Pyroxene is resistive to alteration similar to plagioclase. Petrographically it is more challenging to observe its alteration but appears to start within the mixed layer clay zone or even within and below the chlorite zone boundary where it alters primarily directly to chlorite. Total alteration of pyroxene is rarely reached. It is interesting to observe that the pyroxene chlorite alteration changes to amphibole alteration when entering the epidote amphibole zone and deeper into that zone chlorite starts to alter to amphibole, both of which indicates diminishing chlorite stability and the preference of amphibole (actinolite) alteration. Figure 11 summarizes the relation between the clay crystallization and clay alteration of the primary minerals compared with the established alteration zones.

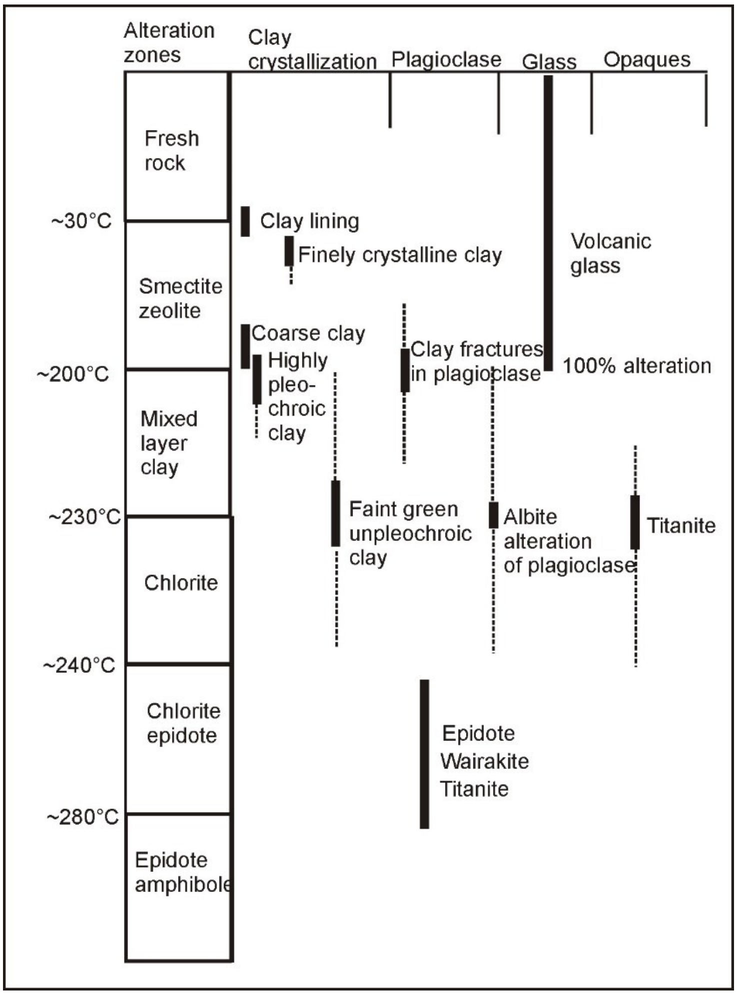

Figure 11. Alteration zones with approximate temperature at zonal boundaries to the left and upper boundaries of clay crystallization in voids and clay alteration of primary minerals (Franzson 1994). Note the non-linear temperature scale.

DISCUSSION

Clays and chlorites play an important part in the hydrothermal alteration processes of high-temperature systems in Iceland and have been studied widely. However, less attention has been paid to where and when they precipitate or transform into higher temperature varieties. This paper attempts to focus on these issues within the upper part of the Icelandic crust. The main findings show clearly that the clay crystallization occurs dominantly within the smectite-zeolite zone, but is only insignificantly seen depositing below that depth, and there mostly as an alteration product of pyroxene and plagioclase. What is also a very significant finding is the relation of the crystallization of coarse-grained clay and the disappearance of zeolites. This crystallization occurs as a prelude to the disappearance of the zeolites as they grow from the inner part of voids towards the disappearing zeolites. Following the clay deposition, zeolites continue to disappear by a transformation to higher temperature minerals, mainly quartz and wairakite. The absence of quartz and wairakite underlying the coarse-grained clay deposition strongly implies that the latter is preceding the quartz/wairakite alteration of the zeolites. The upper boundary of the coarse-grained clays has become a standard observation during the cutting analysis in high-temperature drilling in Iceland. Very similar occurrences, further to Nesjavellir, are found in other high-temperature areas such as Hellisheiði, Svartsengi, Eldvörp and Reykjanes (e.g. Franzson, 1983, 1987, 1995, et al. 2002, et al. 2010a) and is proven to represent a specific alteration event. It must be mentioned, however, that alteration is to some extent related to the composition of bedrock, where the more basic varieties of basalts (olivine tholeiites) are more sensitive, while the more evolved ones (quartz tholeiites), are more resistive to alteration. This, possibly, may change the alteration trend and where coarse-grained clay starts to form. The rock series in the high-temperature areas in SW-Iceland are dominantly olivine tholeiite composition, while those in the northern volcanic zone may be within more evolved rock formations. Whether there is a fundamental difference in clay deposition has not been specifically studied.

Smectite and mixed layer clays are usually found far down into the chlorite and chlorite-epidote zone which to some extent contradicts the strict minimum temperature boundary of the clay zonation and this is mainly recognized through XRD- and petrographical analyses. The close study of the relation between clay and other alteration minerals with permeability variations shows a very clear relation where higher temperature clay varieties occur preferentially within high-permeability zones while the lower temperature ones prevail at low permeability. This demonstrates conclusively how permeability controls the rate of clay transformation, being very low at low permeability but fast at high permeability.

CONCLUSIONS

Clays and chlorite are abundant in the upper part of the Icelandic crust and play an important role in hydrothermal alteration studies, in particular in high-temperature systems. This study which is based on detailed alteration, geological and permeability analysis of wells at Nesjavellir, shows a clear trend in the evolution of clay in the geothermal system.

Smectite clays deposit in voids, first as microcrystalline clay linings at the top of the smectite zeolite zone followed by a finely crystallized one. At temperatures of ca. 160°C, near the lower limit of that alteration zone, coarse-grained smectite clays deposit on top of the fine grained ones.

Low-temperature zeolites dominate at shallow levels but are replaced by higher temperature ones with increasing depth. At the lower boundary of the smectite zeolite zone they are seen altering to quartz and wairakite. This zeolite alteration is preceded by the formation of coarse-grained clays which grow underneath the disappearing zeolites.

Smectite clay is the dominant alteration of volcanic glass and the more rare olivine. The more resistive plagioclase and pyroxene start to alter within the mixed layer clay and chlorite zone but this clay alteration is considered relatively negligible.

The volcanic accumulation within the rift zone leads to the suppression of the volcanic strata down into the geothermal gradient leading to a progressive alteration of individual volcanic formation with time. The dominant part of the clays within a high temperature system is considered to have originally been smectite. When the volcanic formations enter below the chlorite zonal boundary the existing clays are transformed into higher temperature varieties such as mixed layer clays and chlorites. These are not viewed as direct mineral deposition but more as recrystallization of earlier clays.

Clay zonation has to date been mainly related to temperature. Although true, another factor is believed to influence the clay and that is permeability, leading to that the clay transformation is very fast at high permeability but slow at low permeability. This explains why smectite and mixed layer clay persist to much higher temperatures.

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