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FLUID INCLUSION STUDIES ON HYDROTHERMAL MINERALIZATION ASSOCIATED WITH THE DURUCHAUS FORMATION: GENETIC ASPECTS OF LARGE QUARTZ. DOLOMITE BODIES AND ASSOCIATED ALTERATION

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ABSTRACT

In the southern portion of the Damara Orogen of Namibia large bodies of megacrystalline quartz and dolomite, many of them with a core of silicified dolomitic breccia that generally exhibits intrusive patterns, are abundant. Systematic fluid inclusion studies on the quartz-dolomite bodies by means of decrepitometry and microthermometry revealed that three phases of fluid activity led to the formation and alteration of the quartz-dolomite bodies. Most of the quartz and dolomite formed from a highly saline fluid phase which was generated by dehydration and leaching of the evaporitic Duruchaus Formation. The general characteristics of these genetic fluids are high salinity (38 vol% NaCI) and high oxygen fugacity (minimum log fO_2 -30). Minimum temperatures of formation ranged from 130°C to possibly as high as 330°C. A second fluid phase with a distinctly higher CO_2 content, but still high salinity, developed and formed a quartz stockwork in the surrounding wall rock. Total homogenization temperatures for the inclusions are 180° to 230°C. In the quartz-dolomite bodies this fluid phase formed secondary and pseudosecondary inclusions. In the quartz stockwork this phase is characterized by very variable gas-liquid ratios and high CO_2 content of the fluid inclusions which is interpreted as the result of boiling. A third fluid phase that led to strong alteration of the quartz-dolomite bodies consisted of almost pure CO_2. A minimum pressure of formation of 3 kbar was determined from these inclusions. Mineral parageneses were used to estimate pH-fO_2 conditions. The various fluid phases can be related to different stages of the tectonic development of the Damara Orogen.

1. INTRODUCTION

Previous studies on fluid inclusions in the Damara Orogen have established a comprehensive framework of fluid systems, that were active during metamorphic and tectonic processes (Behr and Horn, 1982; Behr *et al.*, 1983b). One of these fluis systems was characterized by extreme salinity. In many places it caused extensive hydrothermal alteration and led to emplacement of large quartz-dolomite bodies. The fluids were, at least partly, generated by dewatering of the evaporitic metaplaya-sequence of the Duruchaus Formation.

The physico-chemical conditions of formation and secondary alteration of quartz-dolomite bodies were studied at a representative example on the farm Hilton (Fig. 1).

Fluid inclusion studies were carried out in two steps:

(i) All samples were first analyzed by decrepitometry. Subsequently the resulting data were used to select samples for microthermometric analysis.

(ii) Samples were then studied by microthermometrical methods and the results from microthermometric studies were correlated with decrepitometrical data.

An estimation of the pH-fo_2 conditions of formation was made on the basis of mineral parageneses.

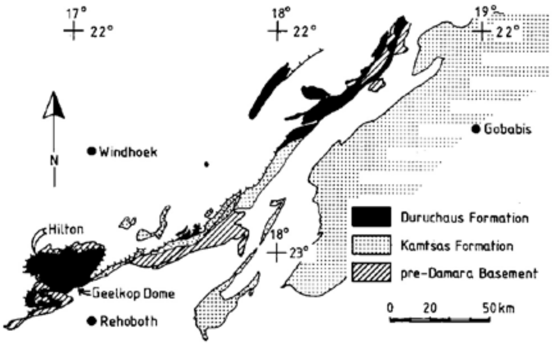


Fig. 1: Geological map showing the distribution of the Kamtsas and Duruchaus Formations and part of the basement (Billstein and Marienhof Formations). (Modified after: Geological Map of South West Africa/Namibia).

2. GEOLOGICAL SETTING

The quartz-dolomite bodies under investigation are located at the north-western margin of the Geelkop Dome (Figs. 1 and 2). The margin is defined by major thrust faults. The lowermost unit of the Damara Sequence in this area is the Duruchaus Formation, consisting of shale, dolomitic siltstone and, predominantly in the upper part of the section, intercalated layers of meta-evaporites. Most rocks are quite unaltered and undeformed, and even very delicate sedimentary structures are well preserved. In the evaporitic layers, pseudomorphs of various evaporite minerals were identified (for a detailed description see Behr *et al.*, 1983a).

In the west and north-west of the study area, outcrops of the Duruchaus Formation are terminated by a major thrust fault. Here, feldspathic quartzite of the Kudis Subgroup is thrust upon the parautochthonous Duruchaus Formation. The thrust zone shows effects of intense hydrothermal alteration. Up the sequence follow layers of graphitic schist interbedded with pebbly schist and some minor calcareous layers of the Kudis Subgroup. Towards the top, the pebble size in the pebbly schist increases and some quartzitic layers are intercalated.

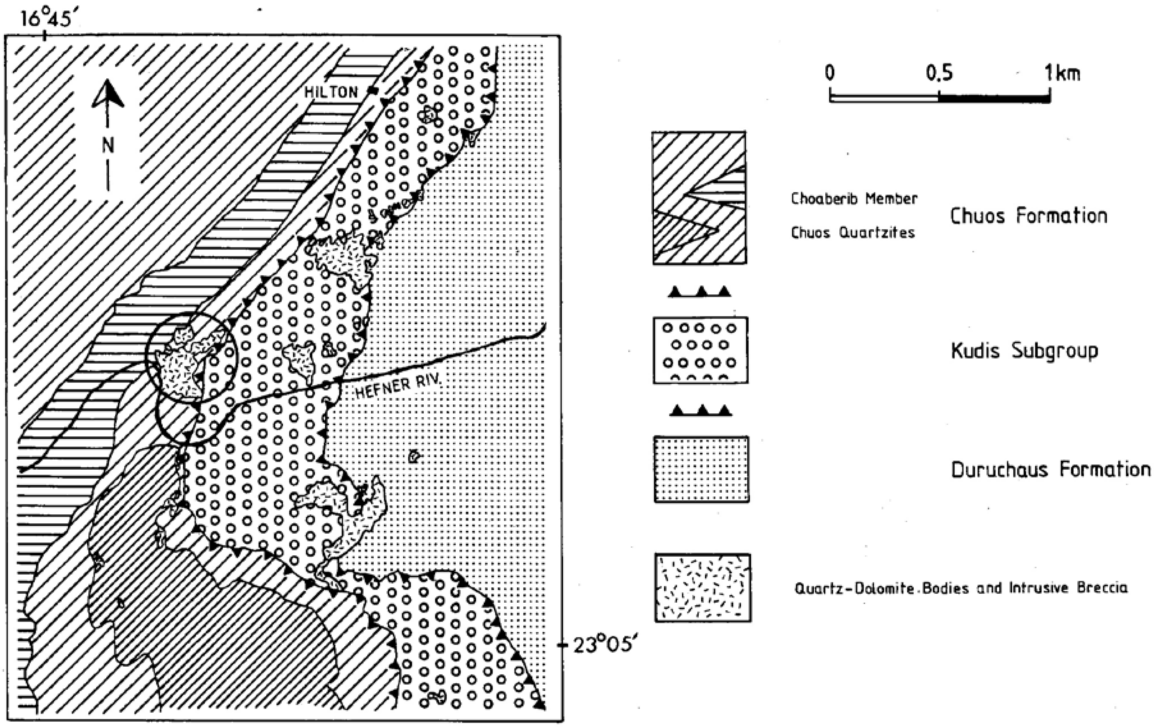


Fig. 2: Geological map of a portion of Farm Hilton (portion of Vaalgras 38), showing distribution of quartz-dolomite bodies. Circle indicates body referred to in this study.

The exposed top of the sequence is defined by a zone of highly sheared and deformed graphitic schist and one to three layers of carbonate showing flow lamination and internal isoclinal folding. This defines a further thrust zone where rocks of the Chuos Formation are thrust upon the underlying Kudis Subgroup.

The base of the Chuos Formation consists of pebbly schist or conglomerate. In a south-westerly direction the sequence grades into thinly layered quartz-feldspar schist, followed by several tens of metres of feldspathic quartzite. Further up in the Chuos Formation talc-tremolite schist and dark, fine-grained amphibolite of the Choaberib Member are intercalated. The sequence continues with pebbly schist and a thick monotonous series of feldspathic quartz-muscovite schist containing rare pebbles and segregational quartz augens.

Quartz-dolomite bodies occur at all positions within this sequence, up to the base of the amphibolite (Fig.2). From field relationships it appears that they were emplaced preferentially in tectonically caused low pressure zones, e.g. along opening fractures and in extensional areas, in some cases subparallel to the main foliation. Field evidence shows that most of the mineralizing fluids originated from hydrous evaporitic layers of the Duruchaus Formation by dehydration. The highly saline fluids ascended along fractures, carrying large amounts of dissolved matter. The basal parts of the quartz-dolomite bodies show that besides fluids, a carbonatic breccia was mobilized, consisting mainly of fragments or clusters of crystalline carbonate and intraclasts of very variable size. During tectogenesis of the

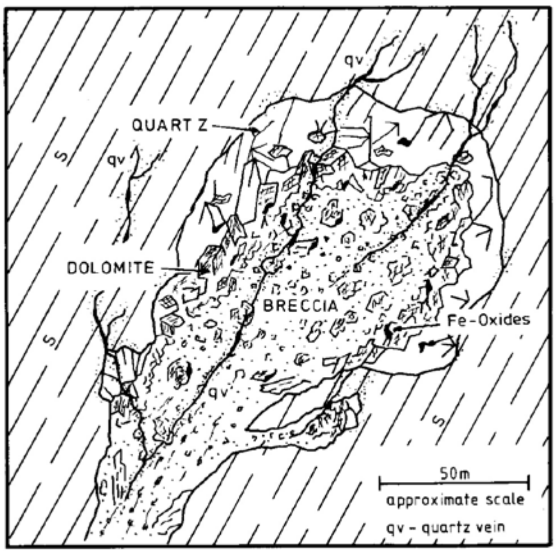


Fig. 3: Schematic section across a quartz-dolomite body. For explanation see text. qv-quartz veins, s-main foliation.

Damara Orogen this mobile mush was also injected into thrust planes, where it acted as a lubricant for nappe movement. The fluid content of this mush was then released and additionally contributed to the formation of quartz-dolomite bodies.

3. THE QUARTZ- DOLOMITE BODIES

Regional geological studies have shown that most of the quartz-dolomite bodies are associated with, or situated in the Duruchaus Formation. In the area under investigation the setting of the quartz-dolomite bodies is tectonically controlled. The geological map (Fig. 2) shows only major occurrences. Two features of these bodies are a well-developed zoning and a quartz stockwork impregnation of the wall rock.

3.1 Zoning of the Quartz-Dolomite Bodies

The zoning is reconstructed from exposed parts of different bodies present in the study area. The top part of a quartz-dolomite body is -formed by a thick quartz cap and an inner dolomite shell. In many cases the quartz cap is poorly developed or eroded, but crystalline dolomite is always present. In the outer parts of deeply eroded, concentric occurrences (Fig. 3), the quartz zone is only sparsely developed, forming isolated blocks at the margin. Breccia is the most common rock type representing the inner and lowermost part. At even more deeply eroded bodies, the breccia can be seen to intrude into faults and fractures in the wall rock. At these deeper levels fragments of wall rock of all sizes, many of them highly silicified or penetrated by a network of quartz veins, are enclosed in the breccia. Flow structures are obvious, indicating mass movement and an intrusive character of the breccia. At one locality the carbonatic breccia mush can be seen to have intruded into a thrust plane.

The zoning is best seen in the large body that is crossed by the Hefner River (Fig.2). Here, the uppermost part of a quartz-dolomite body was studied in detail. Fig. 3 shows a schematic cross-section through a quartz-dolomite body. Three zones can be determined.

3.1.1. Zone of Massive Quartz

A cap of massive quartz with well developed crystal faces forms an outer zone several metres in thick. Large single crystals (megacrystals) of several metres in size or large crystal faces of closely intergrown or twinned quartz are present. The quartz is milky white and generally shows trails of secondary generations of fluid inclusions. Towards the centre of the body, cavities of up to Im size, filled with crystalline dolomite, occur.

3.1.2. Zone of Massive Dolomite

Tightly intergrown with the quartz megacrystals is a 3 to 4 m thick shell of megacrystalline (several tens of centimetres) dolomite, characterized by closely intergrown and twinned dolomite crystals of brown or dark green colour. Many vesicles are filled with anhedral nodules of iron oxide. Iron deposits can also be seen along fine fissures and on crystal faces.

3.1.3. Breccia Zone

The massive dolomite gradually changes into a clast-supported breccia, the transition being defined by increasing fracturing of the holocrystalline dolomite; but the change may also be very abrupt. The breccia consists of large dolomite and quartz fragments (up to 5m in size) and clasts of wall rock or breccia intraclasts of variable size. The fragments are set in a silicified matrix of relatively small (up to several centimetres) fragments of quartz and dolomite. Clast size gradually decreases towards the centre of the quartz-dolomite bodies where the breccia in general is matrix supported.

3.2. Quartz Stockwork and Secondary Alteration

All quartz-dolomite bodies are cross-cut by quartz veins with distinct alteration haloes, mostly only a few centimetres across. Iron and carbonate precipitation is often associated with these quartz veins. Similar veins can be found in the surrounding country rock forming a.quartz stockwork. A very late generation of fine crystalline quartz is commonly precipitated on the walls of small cavities. The surrounding country rock shows only slight hydrothermal alteration in the form of a bleached zone of a few centimetres thickness.

4. GENESIS OF QUARTZ-DOLOMITE BODIES

Data from fluid inclusion studies and the mineralogical composition of the hydrothermally generated quartz-dolomite bodies allow an estimation of physicochemical conditions of formation. Fluid inclusion studies were used to determine pressure-temperature conditions and, in combination with geological field studies, helped to estimate the relative time of emplacement. The mineral assemblage allows a good estimation of fO_2-pH conditions at the time of emplacement.

4.1. Fluid Inclusion Studies

Fluid inclusion studies were carried out by decrepitometry and microthermometry to determine the processes of formation and alteration of the quartz-dolomite bodies. The nomenclature used in the fluid inclusion study of this paper follows the guidelines given by Roedder (1984).

4.1.1. Decrepitometry

Decrepitometry was used to determine the overall statistical distribution of the various generations of fluid inclusions in different parts and settings of the quartz-dolomite bodies and the wall rock.

The decrepitometer was developed at the Institut für Geologie und Dynamik der Lithosphare (IGDL), Universität Göttingen. It was designed to be used as both a field and laboratory instrument. In the field it helped as a “guide in unknown territory” to distinguish the fluid systems that left their “fingerprints” on the hydrothermally generated or altered rocks. Thus it was obvious from the first field meas-urements, that formation and alteration of the quartz-dolomite bodies underwent several phases of fluid activity, each one leaving its traces as primary, pseudosecondary or secondary inclusions. The significance of each generation of fluids for the development of the quartz-dolomite bodies and alteration of surrounding rocks can be read from the decrepitograms. Variations of physicochemical conditions are also manifested in the decrepitograms (Hladky and Wilkins, 1987).

4.1.2. Microthermometry

Microthermometric studies revealed three phases of fluid activity that affected the quartz-dolomite bodies. Each of these phases can again be subdivided into several generations of fluid activity. Fig. A shows the microthermometric results of all studied fluid inclusion generations.

Inclusions of the earliest phase are characterized by high salinity. They always contain an NaCI daughter mineral, but in some cases up to five different solid phases are present. CO_2 is sometimes present in very low concentration. Primary inclusions containing CO_2 are attributed to a later stage of activity of the first fluid phase. Inclusions are often large (50 µm), sometimes exhibiting negative crystal forms.

Most of the quartz and dolomite megacrystals were precipitated from these highly saline fluids. Microthermometric measurements revealed a low temperature of homogenization of the vapour phase (Th) (130° - 200°C) but, depending on the setting, it can be as high as 330°C. Homogenization temperature of NaCI daughter minerals is rarely more than 10°C above or below Th, corresponding to a maximum salinity of 38 wt per cent (Keevil, 1942). Necking down is observed on many of these primary inclusions and contributes to the variation of Th. A minimum pressure of 0.5-1 kbar is estimated from the above temperature characteristics of the CO_2-bearing inclusions. Freezing measurements on these inclusions show that dissolved components other than NaCI must be present. Eutectic temperatures (Te) of about -42° to -60°C but even as low as -75°C, melting temperature (Tm_{ice}) of about -20° to -30° and hydrate or clathrate melting temperatures of up to +28° indicate the presence of other dissolved species. It is assumed that they are Ca^{2+} and Mg^{2+}. The presence of dissolved carbonic and sulphuric compounds must also be considered here.

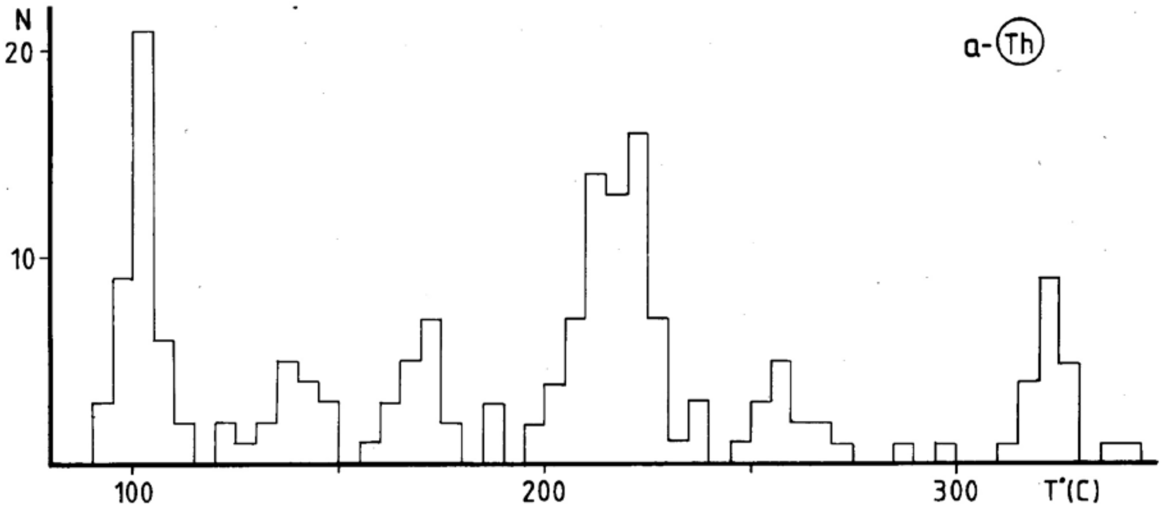
The second fluid phase formed the quartz stockwork in the surrounding country rock and also led to Further quartz precipitation in the quartz-dolomite bodies in the form of cross-cutting veins. In the quartz of the large quartz-dolomite bodies this fluid phase is present as trails of pseudosecondary or secondary inclusions aligned along distinct planes and healed fissures. Here, they often have negative crystal shape and a constant gas-liquid ratio.

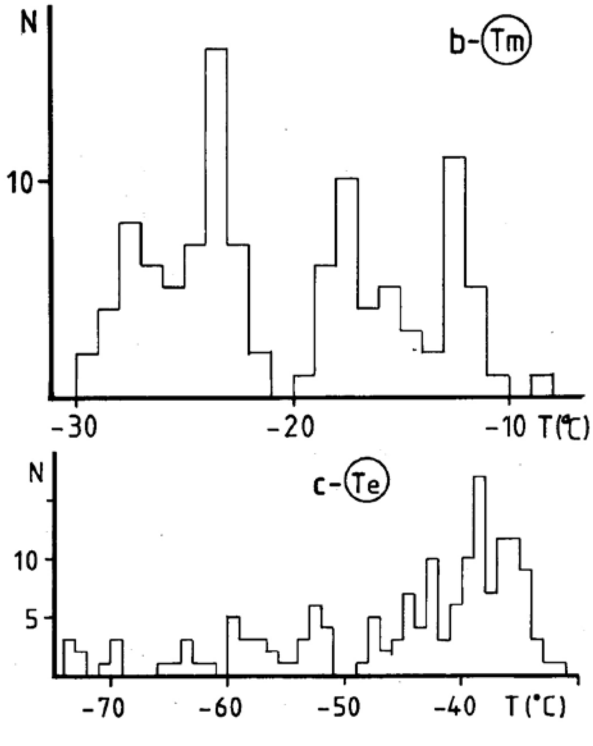
In the vein quartz these fluids form primary or secondary inclusions, characterized by a distinctly higher CO_2 content than equivalent inclusions in the quartz of the quartz-dolomite bodies. The gas-liquid ratios in these inclusions are very variable (up to 90 vol. per cent gas), especially in secondary inclusions. This indicates boiling of the fluids. All second phase inclusions contain NaCI daughter minerals. Other solid phases are only rarely present and are suggested to be captured solids.

In many cases there is a gradual transition from the first, highly saline fluid phase in the quartz-dolomite bodies to secondary, CO_2-rich inclusions. Thus, some of the late primary inclusions contain minor CO_2. The differences in the microthermometric data are quite distinct in respect to the end members. The total homogenization temperature of 180° to 230°C is higher than for the inclusions of the first phase, still they overlap. The homogenization of the NaCI daughter mineral indicates a less saline (about 32 wt per cent) but still saturated solution. Melting temperatures (Tm_{ice}) can be as low as -28°C but are higher in most cases (-12° to -18°C). Eutectic temperatures spread from -38° to -43°C. Together with the melting temperatures of the hydrates or clathrates, with a maximum at +6.8°C, this is interpreted to be due to a lower content of dissolved salts other than NaCI.

The third phase of fluid activity is distinctly different from the first two. All inclusions of this group are purely secondary. Almost all samples studied were affected by this phase of fluid activity. The quartz and dolomite are strongly altered. Thin sections of the altered rocks reveal a strong internal fissuring, thus allowing the fluids to invade the rock. Clouds of minute inclusions surround these fissures, giving the quartz the milky texture. Where alteration is strong, primary inclusions are partly or completely emptied (natural decrepitation), many leaving behind their daughter minerals as solid inclusions or multicomponent inclusions with 80 vol per cent or more of solids and only minor amounts of liquid and gaseous phases. These altered inclusions were not used for measurements.

The determination of physico-chemical characteristics of this secondary generation of fluid inclusions is difficult due to the small inclusion size. Largest inclusions formed are about 5 µm in size. Most incrusions are monophase and did not undergo a phase change during cooling or heating. Most measured inclusions contained pure CO_2 only few cogenetic low-saline, aqueous inclusions were present. No H_2O-CO_2 mixed inclusions were found. Eutectic temperatures were about -35°C. Tmice of the aqueous inclusions was -10° to -14° C corresponding to a salinity of about 14 wt per cent NaCI equiv. Total homogenization occurs at 130° to 150°C. The cogenetic CO_2 inclusions have homogenization temperatures as low as -50°C but most inclusions homogenize in the vicinity of -45°C. Pressure estimations from these data indicate at least 3 kbar as the minimum





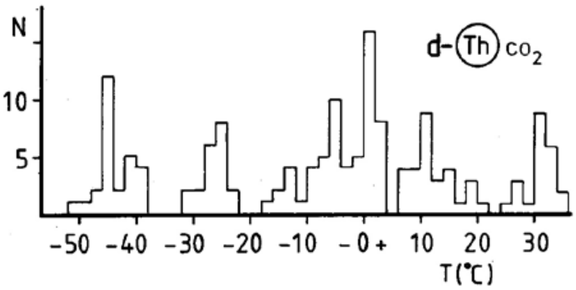


Fig. 4: Results of microthermometric measurements on fluid inclusions in quartz-dolomite bodies and associated quartz stockwork. 'a': Th - Total homogenization temperatures of aqueous inclusions, mostly homogenization of vapour into liquid phase. 'b': Tm-Melting temperatures of ice. Please note: If a solid phase is present, Tm may only serve as a measure of the degree of saturation of the liquid and gives information about the presence of dissolved component other than NaCl. 'c': Te-Eutectic temperatures of aqueous inclusions. Values as low as -75°C indicate the presence of polyvalent cations (Ca^{2+}, Mg^{2+}). 'd': Th-CO_2. Homogenization temperatures of CO_2 allow an estimation of minimum pressure of formation of the inclusion. The very low values of -39° to -51° correspond to a density of 0.8 to 1.2 g/cm³

pressure of formation.

4.2 Correlation of Decrepitometry and Microthermometry

Due to the numerous parameters (e.g. inclusion size, shape and distance from grain boundary, density and composition of liquid and vapour phase, heating rate and sample grain size) that influence the decrepitation temperatures, the decrepitograms have to be seen in connection with microthermometric measurements, if any valid interpretation is to be made. Fig. 5 shows decrepitograms which best represent the three phases of fluid activity. Decrepitograms show the statistical distribution of fluid inclusions in a sample.

The sample of Fig. 5a was taken from the outer part of the quartz cap of a quartz-dolomite body. In this section only primary and pseudosecondary inclusions of phase one and two are present. The decrepitogram shows effects of phase one and two fluids. A distinction of the two fluid phases is not possible by decrepitometry alone, but with information from microscopy and microthermometry, the decrepitogram can readily be interpreted.

The large peak from 200° to 500°C is due to the overlapping of the primary and CO_2-rich secondary or pseudosecondary inclusions. The wide spread of the decrepitation temperatures is due to strong variation of inclusion size and CO_2-H_2O ratios. Also, necking down of primary inclusions, which is observed in thin section, is reflected in the decrepitogram. According to Roedder (1984, p. 248) CO_2-rich inclusions can decrepitate before homogenization due to the pressure dependent immiscibility of CO_2 and H_2O, especially if dissolved

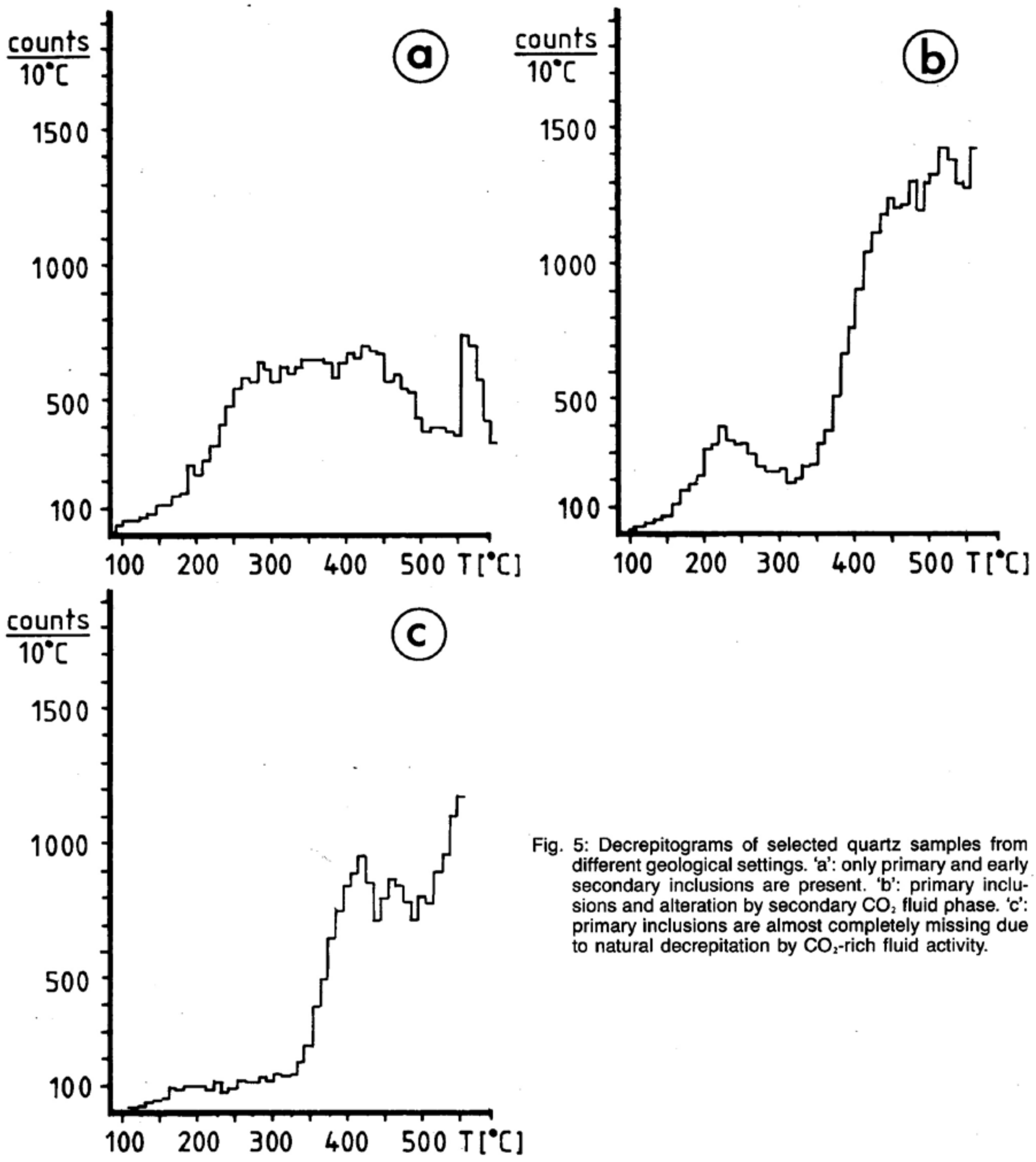


Fig. 5: Decrepitograms of selected quartz samples from different geological settings. 'a': only primary and early secondary inclusions are present. 'b': primary inclusions and alteration by secondary CO_2 fluid phase. 'c': primary inclusions are almost completely missing due to natural decrepitation by CO_2-rich fluid activity.

salts are present.

The sample of Fig. 5b was taken from a quartz vein associated with a quartz-dolomite body. The peak from 220°to 320°C represents the primary and secondary fluid generations. Primary inclusions show variable CO_2-H_2O ratios, and one or two daughter minerals. Total homogenization temperatures spread from 155° to 260°C, overlapping with the decrepitation temperature. At least two generations with different CO_2 densities can be correlated with different temperatures of decrepitation. The rapid increase of counts at 360°C is due to the third type of fluids, which formed high density, pure CO_2 inclusions. The reason for their relatively high temperature of decrepitation is their small size (Leory, 1979).

Fig. 4c shows the result of the extreme alteration by this secondary generation of CO_2-fluids. Primary or phase two inclusions are all emptied by natural decrepitation. The minor peaks between 190° and 350°C now represent the few remaining inclusions of these early phases.

This type of alteration has affected almost all quartz-dolomite bodies and associated quartz mineralizations, but the degree of alteration varies widely.

5. MINERAL PARAGENESIS

The mineral composition of the quartz-dolomite bodies is rather simple as they consist almost entirely of quartz and dolomite; a minor component is Fe^{3+}-oxide. The existing parageneses allow an estimation of pH-

fo^2 conditions of formation. Oxygen fugacity has been calculated using the presence of Fe^{3+}-oxides after the equilibrium reaction:

$$3Fe_2O_3 \leftrightarrow 2\ Fe_3O_4 + \frac{1}{2}\ O_2$$

For a temperature of 250° to 300° C, values of log fO_2 of -32 to -29 are required for the stability of Fe^{3+}-oxides (Helgeson, 1969).

The pH was calculated after the pH controlled reaction for the formation of dolomite.

$$Ca^{2+} + Mg^{2+} + 2\ HCO_3^- \leftrightarrow CaMg\ (CO_3)_2 + 2\ H^+$$

For the given pressure and temperature, CO_2 saturation and a 0.8 molar (tot. Me^{2+}) solution, a pH of approximately 10.1 was calculated. For concentrations of Im total Me^{2+} the pH drops to 9.8 (log K and activity coefficients after Bowers *et al.*, 1984).

Another factor that controls the dolomite precipitation is the Ca^{2+}/Mg^{2+} ratio, (Rosenberg *et al.*, 1967). A distinct predominance of Ca^{2+} over Mg^{2+} is necessary for the dolomite precipitation at the given temperature. Thus it can be concluded, that Ca^{2+} was a major component of the solution. The presence of Ca^{2+} has already been postulated from fluid inclusion data.

6. GEOLOGICAL INTERPRETATION AND CONCLUSIONS

The evolution of the quartz-dolomite bodies and associated alterations is subdivided into five phases:

(i) Invasion of highly saline fluids into the thrust-zone and thereby heating of the fluids. This would have enhanced nappe movement. When the pore pressure equals the lithostatic pressure a state of flotation is reached. This results in the opening of fractures in the nappes due to decrease of tectonic stress.

(ii) Intrusion of discordant breccia into the thrust planes and into opening fractures. Migration of fluids into the fractures results in precipitation of quartz, mainly controlled by temperature and pressure decrease (Kennedy, 1950). Megacrystal growth suggests that the physico-chemical conditions were constant over a long period of time. Microthermometric measurements indicate minimum temperatures of formation of initially 300°C slowly decreasing to approximately 230°C. In some cases temperature decreased even below this value. Pressure estimations suggest a minimum pressure of 0.8 kbar.

(iii) Decreasing pressure and temperature and increasing CO_2 content of the fluids subsequently led to precipitation of dolomite. In a transgressive stage quartz and dolomite were cogenetic and are closely intergrown. Iron, which was also dissolved in the brines, could not be incorporated in the carbonate for reasons of fO_2, temperature and Ca^{2+}/Mg^{2+} ratios (Rosenberg *et al.*, 1967). It was therefore enriched in the residual fluids and precipitated as iron oxide. Further decrease of tectonic stress resulted in boiling of the fluids and opening of fractures into which a more CO_2-rich fluid phase migrated. The extensive quartz stockwork in the surrounding country rock was formed. The fluid inclusions are commonly very CO_2-rich and show boiling patterns. Inclusions of saline brines are present too.

(iv) A later phase of fluid activity resulted in fine fissuring and percolation of CO_2-rich fluids through the quartz-dolomite bodies and the wall rock. The most striking feature of the fluid phase is the extremely high CO_2 content. Most inclusions contain pure CO_2 and only few are found to contain aqueous solution. Pressure estimations indicate minimum pressures of formation of 3 kbar. Various mechanisms are suggested to explain the high pressure (Barker, 1972; Bradley, 1975; Gretner, 1979), viz. aquathermal pressuring, osmotic pressuring, dehydration, and tectonic loading. Future work on the fluid systems of the Damara Orogen has to explain the origin of this high pressure fluid phase.

(v) A remaining H_2O-rich phase, which was still highly saline, basic, and silica-rich, silicified the host rock. Very late segregational quartz was precipitated from the last residual fluids in vesicles, or cavities. The lack of fluid inclusions in this quartz generation implies undisturbed growth.

The variation of fluid composition within the quartz-dolomite bodies follows a distinct pattern. Differences between various bodies are minor and are mostly within the variational range of a single body. The strongest variation is observed in the intensity of secondary alteration by CO_2 fluids.

Fluid inclusion studies on quartz-dolomite bodies and wall rocks are related to the evolution of the Damara Orogen. Phases one and two of fluid activity reflect an early orogenic activity. Initial thrusting and nappe movement increased pressure and temperature in the hydrous Duruchaus Formation. Several phases of dehydration have to be considered. Initially, only pore water was released, subsequently also dehydration of evaporites and carbonates contributed to the high fluid activity. The increasing CO_2 content of the fluid inclusions is related to this progressive dehydration.

The main phase of tectogenesis in the Damara Orogen, with extensive southward thrusting and penetrative deformation, is reflected in the third fluid phase, charaterized by high CO_2 activity. The approaching CO_2-saturated, low saline aqueous front of tectonic fluids of the accretionary nappe pile of the high temperature metamorphic Kudis to Khomas units met with the previously established, Duruchaus-related fluid system. Due to immiscibility of the two fluid systems, large amounts of CO_2 were released which formed high pressure CO_2

inclusions. The remaining H_2O-rich phase will be the subject of Further studies.

7. ACKNOWLEDGEMENTS

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