Reaction rate estimates based on muscovite dehydration and CSD of garnets in the Malé Karpaty Mts. metapelites

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Abstract

Independent petrological methods for reaction rate evaluation have been compared in metapelitic mineral assemblages (St + Ms + Bt + Grt + Sil + Pl + Qtz). Garnet growth rate has been evaluated on the basis of crystal size distribution (CSD) and the growth residence time has been calculated to be $\tau \approx$ 740–2180 years. Muscovite represents the dehydration reactants of the whole rock reactions that might have lasted ca. 940–2630 y. and proceeded at the rate $K_R \approx$ 6.1 x 10^{-11}–8.6 x 10^{-10} cm^{-2}s^{-1} with presumed $\Delta T \approx$ 0.15 °C and $\Delta S_R <$ 100 J/mol. Though the muscovite dehydration time and garnet population growth residence time have poor correlation ($r = 0.32$), they fall within the narrow geological time span of ca. 3000 years. Results confirm relatively rapid dehydration and transformation of metapelitic rocks in the Earth crust.

Key words: reaction rate, residence time, dehydration, crystal size distribution, metapelites

Introduction

Geophysical studies of Ague and Park (1998), Ague et al. (1998) emphasized the link of regional metamorphic dehydration reactions and induced seismic activities. They developed a hypothesis that the release of water during recrystallization of rocks within the fault zones can occur rapidly in a time frame measured in decades or centuries rather than eons and can lead to repeated volcanic and earthquakes activities.

Theoretical and field rock studies today are significantly shifted from a static thermodynamic mode to dynamic quantification of the recrystallization kinetics describing rock metamorphism associated with dehydration and volume changes. Many studies have been extended to the understanding of fluid production and fluid phase dynamics, especially in metapelitic rocks.

Growing recognition of natural effects that could be attributed to slower reaction rates justifies the need for a field-based quantification of reaction rates to asses the accuracy of lab-based predictions and bridge the gap between idealized conditions of the lab and the complexities of nature and provide useful reaction rate data (Baxter and DePaolo, 2002).

Generally, we presume that mineralogical equilibrium is attained between solid phases and metamorphic fluid. But if regional heating and reaction rate occur rapidly, reaction reactants and products may coexist in disequilibrium. Thus, in terms of dehydration reaction progress and its extent, the modal occurrence of reaction products can be an effective criterion of textural and mineral equilibrium, at least in a relatively small rock volume (e.g. some cm^3). Specific metamorphic dehydration reaction and pulse production of fluid is thus restricted to a narrow temperature interval. The reaction progress is generally controlled by the rate of heat supply and reaction extent depends then on the temperature increase in time.

Such estimates indicate that the variation of reaction rate with temperature can be highly non-linear in the crust and the evolution of metamorphic regime depends on different tectonic settings of a particular terrane determined by dynamic mode of rock recrystallization in time.

Sampling locations were chosen to reflect their structural position in periplutonic zones in close vicinity to the intrusive granitoidic rocks. Concept of crystalline basement evolution of Putiš (2006) has been generally accepted.

The metapelitic rock samples (Fig. 1, Tab. 1) represent post-tectonic metamorphic histories with the thermal culmination of ~580–650 °C and pressures of ~4.5–6.2 Kbar, leaving progressively the stability field of St + Qtz and crossing the Ms + Qtz stability curve. The post--culmination decompression retrograde features are absent with the exemption of diffusion processes and rare tiny chloritization of biotite grains (see e.g. Dyda, 2002). Mineral assemblages are considered to represent equilibrium mineral reactions and thus offer the useful data for estimation of their recrystallization kinetics.

Methodical approach

At first methodical approximation, the metapelitic rock samples (ca. 250–750 g) have been chosen for garnet separation in heavy liquids. Then the garnet
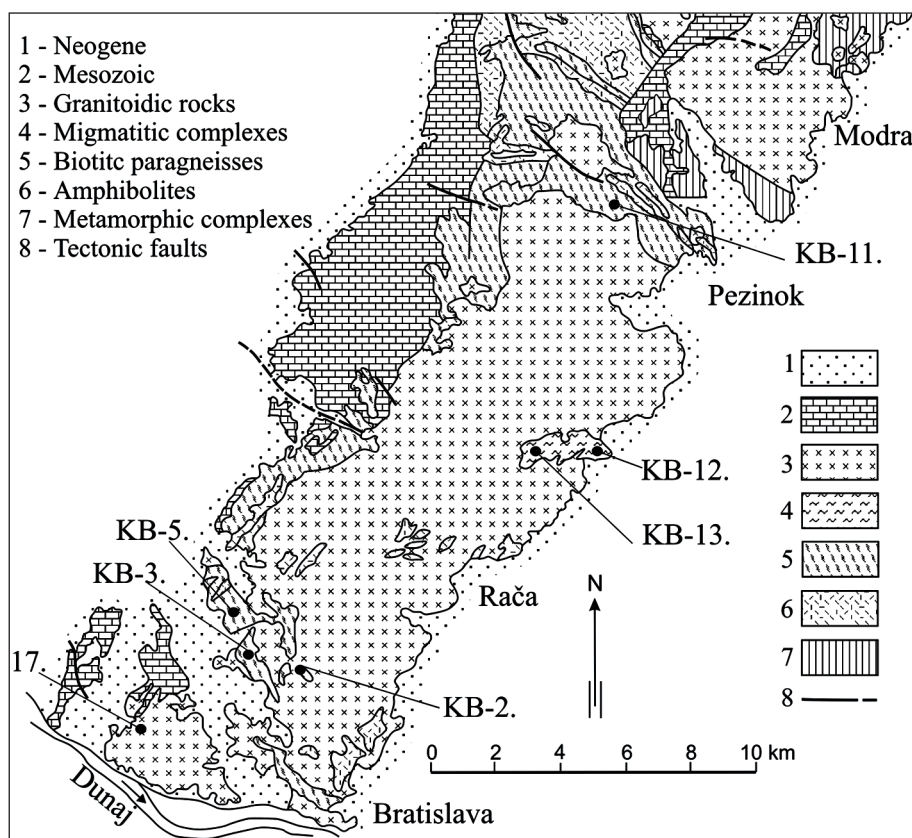
1 - Neogene
2 - Mesozoic
3 - Granitoidic rocks
4 - Migmatitic complexes
5 - Biotitc paragneisses
6 - Amphibolites
7 - Metamorphic complexes
8 - Tectonic faults

Fig. 1. Schematic map of the Malé Karpaty Mts. crystalline complexes and localization of studied metapelitic samples. For more details see Dyda (2002).

heavy fraction has been weighted and compared with the garnet modal content obtained under microscope. Garnet mineral separation from rocks in heavy liquid is frequently connected with fracturing of larger grains and loss of tiny ones. This fact leads obviously to obtaining a shifted garnet crystal size distribution (CSD) histograms. To avoid such uncertainties steaming from this mechanical garnet separation, the microscopic evaluation of garnet CSD has been preferred. Altogether 200–250 garnet grains have been measured and the crystal size frequency in standard rock volume (1 cm^3) has been determined (Fig. 2).

Plotting the population density ln (n) versus garnet crystal size (L) is usually a line with the slope of $k/G\tau$ and intercept n_o, where G is the average growth rate of a particular mineral, τ is the growth residence time and k is the constant. As the average growth rate is known, the growth residence time may be calculated directly. Numerical definition of CSD determines the kinetic and residence growth characteristics of given mineral population in rock.

These methods of mineral nucleation and growth evaluation have been defined earlier in chemical industrial processes by Randolph and Larson (1971, 1988). Later, for silicate systems by Baronnet (1982, 1984) and for geological materials used by Kretz (1973), Lasaga (1982), Brandeis et al. (1984), Carlson (1989), Cashman (1986,1998), Cashman and Ferry (1998) and Marsh (1988, 1998).

Lasaga's (1998) model of heterogeneous kinetics is dealing with the value of specific surface (A) of a particular mineral. Because ΔG and ΔT are nearly linear in a narrow range of the regional metamorphic regime at ~600 °C, the rate law has been approximated as $R = 251 \times 10^{-17} \Delta T^{2.69}$ (moles cm^{-2}s^{-1}) and the minimum time (τ) to reach completion for dehydration reaction given by $\tau = (2.5/RA)$.

Wood and Walther's (1983) model is based on Arrhenius plot of the reciprocal absolute temperature (1/T) versus logarithm of the rate constant (K_R) in gm atom oxygen cm^{-2}s^{-1}, determined from phase equilibrium studies. The rate constant is given approximately by log $K_R = (-2900/T) - 685$. If the rate is predicted to be linear function of ΔG of the overall reaction, then the rate of mass transfer is determined by dm/d$t = -K_R (\Delta G/R$T) in moles cm^{-2}s^{-1}.

For more detailed methodical steps, the reader is recommended to the above cited works.

Tab. 1
Modal composition of the studied metapelites

Sample	Qtz	Pl	Bt	Ms	Grt	St	Sil	Opq
171	38.7	26.8	30.4	0.6	1.6	1.3	0.4	0.2
KB-2	34.3	32.7	27.5	0.2	3.5	0.7	0.9	0.2
KB-3	31.1	38.1	27.6	0.3	1.2	1.2	0.2	0.3
KB-5	24.3	35.1	33.8	2.1	1.8	0.4	2.3	0.2
KB-11	34.8	30.8	28.8	2.2	1.6	0.9	0.8	0.1
KB-12	33.5	30.5	29.7	1.7	2.1	1.0	1.2	0.3
KB-13	29.3	31.6	30.8	1.9	3.8	1.2	1.1	0.3

Garnet producing dehydration reactions

Garnets are the progressive peak metamorphic reaction products in studied rocks. Their appearance in mineral assemblages Grt + St + Ms + Bt + Sil + Pl + Qtz is consistent with mass-balance reactions in simplified metapelitic system K_2O-MgO-FeO-Al_2O_3-SiO_2-H_2O based on analytical data of minerals used for geothermobarometry (see Dyda, 2002).

The lowest variance mineral assemblages of the studied paragneisses include St, Bt, Grt, Ms, Pl, Ilm, Sil, Qtz. The metamorphic whole rock reaction among these minerals is formally represented by a system of linear equations for components chosen. This algebraic approach may include all components and simultaneously exclude degenerative composition relations among minerals in the assemblage. Each composition balance for given component in the assemblage is defined as

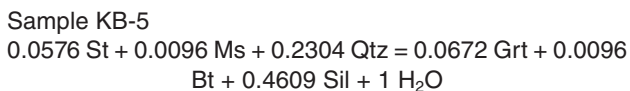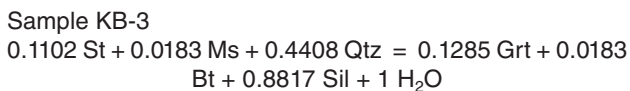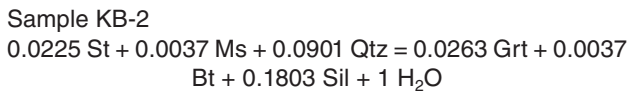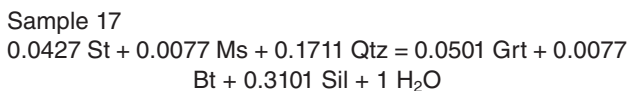$$A_i = \nu_1 X_{1i} + \nu_2 X_{2i} + \nu_3 X_{3i} + \dots \nu_n X_{ni} \qquad (1)$$

where ν – is the stoichiometric coefficient, X_i – element concentration in the mineral and A_i – the concentration of the same element in the reference mineral. The linear equation e.g. for Si and Al in the simplified metapelitic KMFASH system in the assemblage Grt + St + Ms + Bt + Qtz is then expressed:

$$0 = \nu_{Grt} Si_{Grt} + \nu_{St} Si_{St} + \nu_{Ms} Si_{Ms} + \nu_{Bt} Si_{Bt} + \nu_{Qtz} Si_{Qtz} \qquad (2)$$

$$0 = \nu_{Grt} Al_{Grt} + \nu_{St} Al_{St} + \nu_{Ms} Al_{Ms} + \nu_{Bt} Al_{Bt} + \nu_{Qtz} Al_{Qtz} \qquad (3)$$

The formulated set of chemical composition of minerals participating in particular metamorphic reaction thus divides and quantifies the minerals of the assemblage on the reaction reactants and reaction products. Critical importance still has the appearance of equilibrium rock structure (see Figs. 3 and 4).

The mineral abundance in paragneisses (Tab. 1) and mineral composition (see Dyda, 2002) can be thus arranged in the numerical form to express the whole rock dehydration reactions normalized to 1 mol of H_2O:

Sample 17
0.0427 St + 0.0077 Ms + 0.1711 Qtz = 0.0501 Grt + 0.0077 Bt + 0.3101 Sil + 1 H_2O

Sample KB-2
0.0225 St + 0.0037 Ms + 0.0901 Qtz = 0.0263 Grt + 0.0037 Bt + 0.1803 Sil + 1 H_2O

Sample KB-3
0.1102 St + 0.0183 Ms + 0.4408 Qtz = 0.1285 Grt + 0.0183 Bt + 0.8817 Sil + 1 H_2O

Sample KB-5
0.0576 St + 0.0096 Ms + 0.2304 Qtz = 0.0672 Grt + 0.0096 Bt + 0.4609 Sil + 1 H_2O

The calculated reaction stoichiometry compared with microscopic modal study of the samples thus assures the resemblance to the calculated whole rock reaction that actually runs in the rock. Both petrological approaches are thus consistent in the studied metapelitic assemblages.

Processes that effect modal changes and element distribution among minerals in rocks are strictly path dependent and reflect P-T conditions along the reaction path. The dehydration of rocks thus coincides with temperature culmination of rock's P-T trajectory and the equilibrium forming of peak metamorphic assemblage.

However, differences in protolith compositions of the studied rocks (Dyda and Miklóš, 1993) and their tectonic positions may attribute the differences in the reaction extent and modal changes of particular minerals in the assemblages with respect to their growth or consumption.

Garnet crystal size distribution (CSD) is represented by histograms in Fig. 2. The obtained CSD data are typical for garnets of regionally metamorphosed rocks. Consequently, the CSD data enables to generalize the kinetic evolution of garnet population in the given rock. Thus, the basic kinetic characteristics of garnet nucleation rate (N/cm^3s), growth and residence time (τ) of garnet population in samples have been calculated (see Tab. 2).

Crystal size distribution with maximum number of crystals at intermediate size is typically bell-shaped and similar to other garnet size distributions in regionally
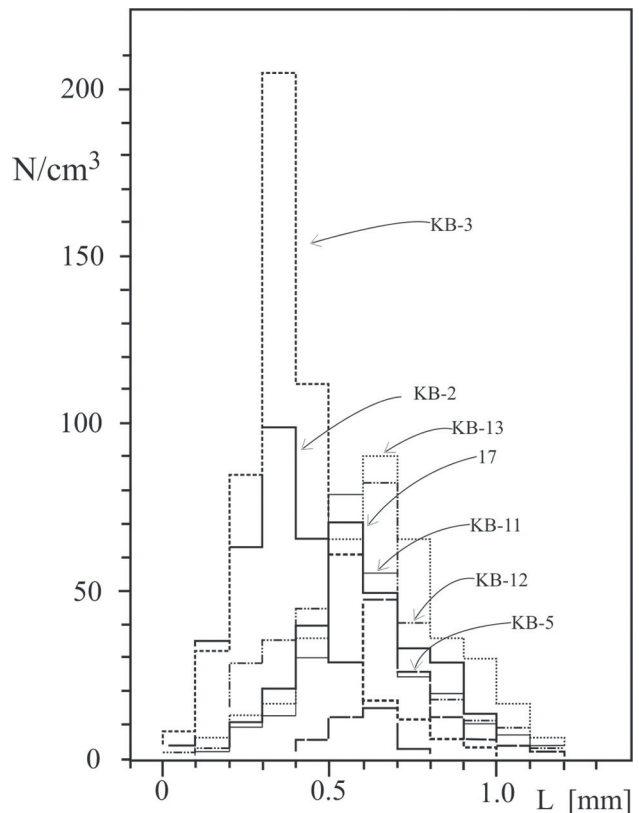

Fig. 2. Garnet crystal size distribution (CSD) in studied metapelitic rocks used for growth characteristics calculation.

Tab. 2
Garnet crystal size, nucleation and growth characteristics*

Sample No	Average grain size [mm]	Standard deviation S	Number of crystals [N_V/cm^3]	Slope	Intersection	Nucleation rate [N/cm^{3}s]	Residence time [τ, years]
17	0.604	0.174	267	37.71	10.964	2.93×10^{-8}	1 660
KB-2	0.366	0.148	313	83.98	12.543	1.40×10^{-7}	740
KB-3	0.384	0.154	544	67.78	12.765	1.08×10^{-7}	920
KB-5	0.718	0.180	116	66.19	12.765	1.77×10^{-7}	940
KB-11	0.630	0.193	253	28.92	10.147	1.29×10^{-8}	2 180
KB-12	0.594	0.218	326	31.82	10.356	1.59×10^{-8}	1 970
KB-13	0.679	0.195	363	36.66	11.082	3.29×10^{-8}	1 710

* calculated on the basis of the methodical approach of Randolph and Larson (1971, 1988), Baronnet (1982, 1984), Brandeis et al. (1984), Cashman (1986), and Cashman and Ferry (1987)
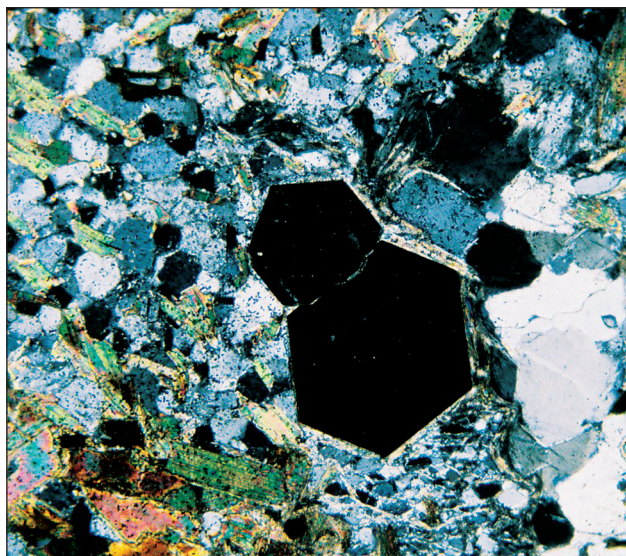

Fig. 3. Idioblastic garnets in rock's texture document their appearance as the products of the whole rock progressive reaction. Base ~0.9 mm. Crossed polars. Photo by author.

metamorphosed rocks. Therefore, certain generalization about the development of the crystal population can be made from the linear correlation between crystal sizes and their population density.

CSD of a garnet population determines garnet nucleation and growth rates, garnet producing reaction overstepping and average residence time of a garnet population. These data provide a numerical comparison of metamorphic recrystallization conditions based on detailed textural and reaction extent studies.

Nucleation rate depends significantly on the type of metamorphic regime. High nucleation rates occur during contact metamorphic conditions, whereas high-grade regional metamorphism is represented by slow rate of nucleation. Thus, the calculated garnet nucleation rates 2.9×10^{-8}/cm^3/s to 1.0×10^{-7}/cm^3/s (Tab. 2) confirmed the periplutonic regional metamorphic recrystallization regime for analysed metapelites. The calculated estimates of garnet growth residence time based on garnet CSD may be bracketed within the time span of ca. 1 000–2 000 years

(Tab. 2). These calculated residence time characteristics for particular samples differ no more than in one order of numerical magnitude.

For comparison purposes of the calculated garnet residence time data, the method of Walter and Wood (1984) has been used and the results are given in Tab. 3.

Garnets crystallized as the peak metamorphic reaction products during metamorphic culmination. The peak temperatures (see Dyda, 2002) were taken as the equilibrium temperatures of garnet growth. The maximum limiting value of temperature overstepping, ΔT, for garnet producing reaction was taken, at first approximation, to be 5 °C (Tab. 3). Two others, lower temperature overstep limits chosen $\Delta T = 0.15$ °C and $\Delta T = 0.04$ °C reflect, however better, the range of temperature overstep for regional metamorphic terrains.
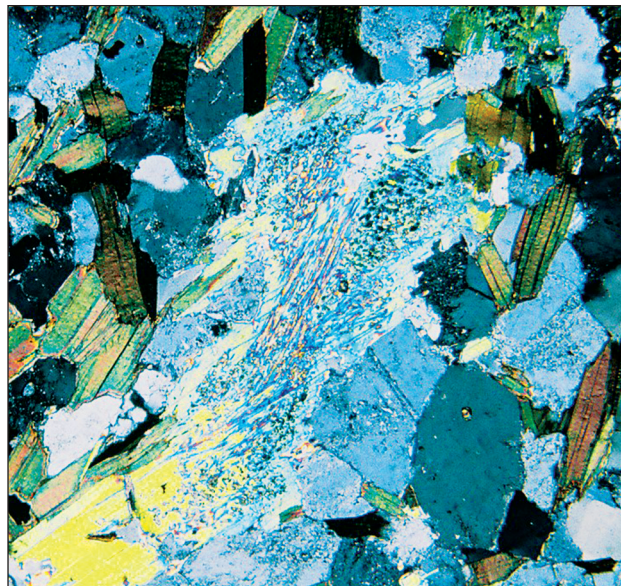

Fig. 4. Muscovite (in central part) represents the reaction rests of the whole rock metamorphic reaction in which muscovite vanishes as the dehydration progresses. The extent of overall reaction is given by molar ratio limits, determining the dehydration consumption of mineral reactants – staurolite and muscovite. Base ~0.75 mm. Crossed polars. Photo by author.

Tab. 3
Calculated garnet growth residence time (τ – years)

Sample No	Reaction rate $K_R[cm^{-2}s^{-1}]$	Temperature overstep ΔT = 5 °C		Temperature overstep ΔT = 0.15 °C		Temperature overstep ΔT = 0.04 °C	
		ΔS = 5 J/mol	ΔS = 100 J/mol	ΔS = 1 J/mol	ΔS = 100 J/mol	ΔS = 5 J/mol	ΔS = 100 J/mol
17	6.16×10^{-11}	940	37	31 620	1 260	118 600	4 740
KB-2	1.10×10^{-10}	340	13	11 490	450	43 100	1 720
KB-3	8.67×10^{-10}	290	11	9 840	390	36 900	1 470
KB-5	6.73×10^{-11}	1 250	50	41 970	1 670	157 300	6 290
KB-11	6.72×10^{-11}	1 070	43	35 880	1 430	134 500	5 380
KB-12	8.41×10^{-11}	830	33	27 940	1 100	104 700	4 190
KB-13	7.11×10^{-11}	1 100	44	36 950	1 470	138 500	5 540

* using model of Walther and Wood (1984)

The exact value of $\Delta S_{reaction}$ for garnet producing dehydration reaction in samples is unknown. However, the chosen upper limit for $\Delta S_{reaction}$ = 100 J/mol/deg may be taken as an appropriate maximum value for muscovite dehydration reaction in studied metapelitic rocks.

The comparison of garnet growth residence time estimates of Tabs. 2 and 3 shows schematically that the residence time estimates based on CSD of garnets match well with metamorphic reaction conditions of ΔT = 0.15 °C and $\Delta S_{reaction}$ = 100 J/mol/deg (Fig. 6). The reaction temperature overstep of ca. 0.15 °C is thus consistent with the regional metamorphic thermal regime.

Duration of thermal conditions in the regional or contact metamorphic environments forms specific textural features of a particular rock. CSD of contact metamorphic rocks gives usually information of high nucleation rates. Probability of CSD modification after peak recrystallization is generally lesser. The regional metamorphic rocks are usually subjected to prolonged cooling after thermal culmination and CSD histograms are mostly modified. This process is represented by the change of shape of the crystal size histograms, as small unstable garnet crystals dissolve and the material is precipitated on larger crystals in the process of Ostwald ripening. The annealing mass transfer is temperature and time dependent process, where large crystals retain the original crystal size frequency distribution. Thus the extent of this process may reflect the duration of high temperature conditions to which the rock sample was subjected.

Discussion and conclusions

The calculated average residence time estimates, based on CSD of garnets (Tab. 2), and the thermodynamic limitations of metamorphic dehydration reactions (Tab. 4) offer the coincidental garnet residence time data with the bracketed reaction conditions limited approximately by temperature overstep of ca. 0.15 °C and the reaction entropy of ca. 100 J/mol/deg. These limiting parameters are in a good agreement with heating rate and reaction characteristics of regional metamorphism; see e.g. England and Thompson (1984), Thompson and England (1984), Schumacher et al. (1990), Bucher and Frey (1994).

For comparison of calculated residence and dehydration time data (τ), the statistical tests of difference in means was used that gives comparative values of *t*. As *t* approaches to zero, the data sets are more similar. Identical numerical sets have *t* = 0.

Each calculation method applied is independent and uses different numerical entries. Elaboration of garnet CSD and muscovite dehydration data (Fig. 5) does not include any tabled thermodynamic data, whereas the Wood and Walther (1984) method works with modeled thermodynamic entries such as ΔT and ΔS_R.

Thus, in comparison, the garnet CSD residence time data correspond to the calculated muscovite dehydration time data with *t* = 0.60, which indicates, that both data sets reassemble well each other in the given time span. Garnet CSD residence time values τ (Grt_{CSD} τ) versus garnet growth τ modeled by Wood and Walther (1983), at ΔT = 0.15 °C and ΔS_R = 100 J/mol gives *t* = 1.20 in comparison
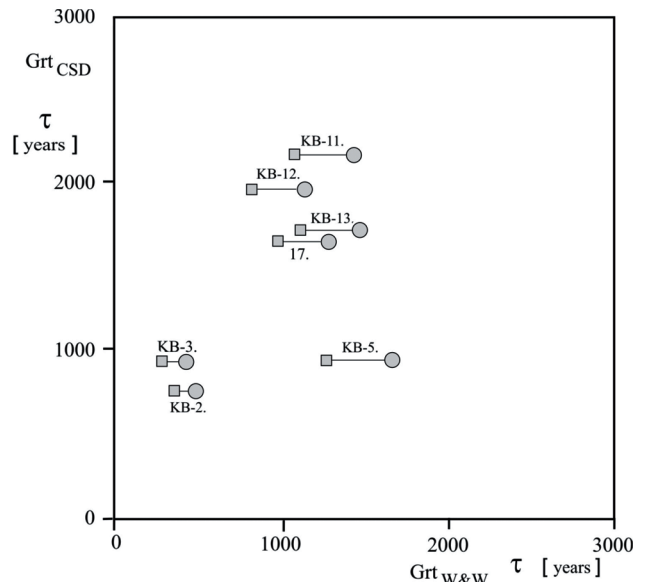

Fig. 5. Muscovite dehydration and garnet growth data in metapelitic rocks with the correlation r = 0.32 are bracketed in relative narrow time span of ca. 3000 years and document a good coherence of the independent calculation methods used.

Tab. 4
Muscovite dehydration rate*

Sample No	Modal %	Average grain size [cm]	Specific surface [cm²/kg] A	Temperature [°C]	Dehydration rate [mol·s⁻¹]	Dehydration time [τ – years]
17	0.6	0.055	125.8	592 ± 17	2.39×10^{-13}	2 630
KB-2	0.2	0.011	209.7	657 ± 25	3.99×10^{-13}	940
KB-3	0.3	0.018	192.3	623 ± 21	3.66×10^{-13}	1 120
KB-5	2.1	0.161	151.4	601 ± 19	2.88×10^{-13}	1 810
KB-11	2.2	0.123	206.3	613 ± 18	3.93×10^{-13}	970
KB-12	1.7	0.144	136.2	627 ± 16	2.59×10^{-13}	2 240
KB-13	1.9	0.141	155.4	607 ± 21	2.96×10^{-13}	1 720

*model of Lasaga (1982, 1998)

to t = 3.70 for Grt_{CSD} τ versus ΔT = 0.04 °C and ΔS_R = 100 J/mol (Tab. 3, Fig. 6).

Consequently, in studied metapelitic rocks, the regional metamorphic equilibrium reaction temperature overstep of about ~0.15 °C is more probable than e.g. the overstep of ΔT = 5 °C or ΔT = 0.04 °C and for metapelitic dehydration reaction the ΔS_R < 100 J/mol may be presumed.

Although kinetics of muscovite dehydration and garnet growth differs in used numerical approach, the release of water entrapped in crystalline structures is likely very rapid. Probably in the order of magnitude as calculated in the used models. The data show further that relatively small increase in temperature beyond equilibrium conditions which can rapidly accelerate the process of fluid release.

During dehydration reactions the metastability is presupposed to be minimal in comparison with the

Fig. 6. Calculation of garnet population growth residence time based on reaction rate, reaction temperature overstep and modeled reaction entropy change ($Grt_{W\&W}$) present small differences at ΔT = 5 °C, ΔS_R = 5 J/mol (■) and at ΔT = 0.15 °C, ΔS_R = 100 J/mol (•), showing thus the consistency with the muscovite dehydration data.

solid phase reactions. Schramke et al. (1987) studied the dehydration reaction Ms + Qtz = And + Kfs + H_2O and concluded that the reaction rates are fast, even at conditions close to equilibrium and that this reaction would rapidly reach completion during metamorphism even when the equilibrium is overstepped only by 2 °C, if no significant nucleation barriers exist. At specific experimental conditions the dehydration of muscovite + quartz, required 4 days to reach 10–50 % completion. In experiments without added water the reactions are generally very sluggish.

However, using mineralized natural quartz and pulverized MgO, ~1–2 cm forsterite crystals may grow at ~1650 °C at atmospheric pressure during 1 day (unpublished results).

At temperature which significantly oversteps the equilibrium, the rate of dehydration can be very rapid. Rubie and Brearley (1990) showed that an initial temperature overstep of ~100 °C caused melting of ~20–50 % of a rock within 1 year.

During such rapid dehydration reactions, equilibrium of major and trace elements partitioning between solid phases and fluid may not occur, because intracrystalline structure diffusion distances will be negligible on the considered time scale (see e.g. Prinzhofer and Allegre, 1985).

When metamorphic reactions occur under marked disequilibrium conditions due to delayed nucleation, then reaction rates may be very fast. This is at least partly because the driving force for transformation, ΔG_R, is large under such conditions. When nucleation is catalysed by fluid influx, under disequilibrium conditions, the presence of fluid will also contribute to fast reaction rates. Some rocks react rapidly as soon as the influx of hydrous fluid occurred, perhaps on a time scale of days to months and the limited extent of reaction of some mineral grains is likely due to limited quantities of available fluid which is consumed by the hydration reaction (see Wayte et al., 1989; Rubie, 1990).

Not all dehydration reactions involve sluggish nucleation kinetics. Rutter and Brodie (1988) observed that nucleation during the reaction serpentine → olivine + talc + H_2O occurs within a few degrees of equilibrium boundary and Worden et al. (1991) conclude, that little overstepping is required for this reaction in a contact metamorphic
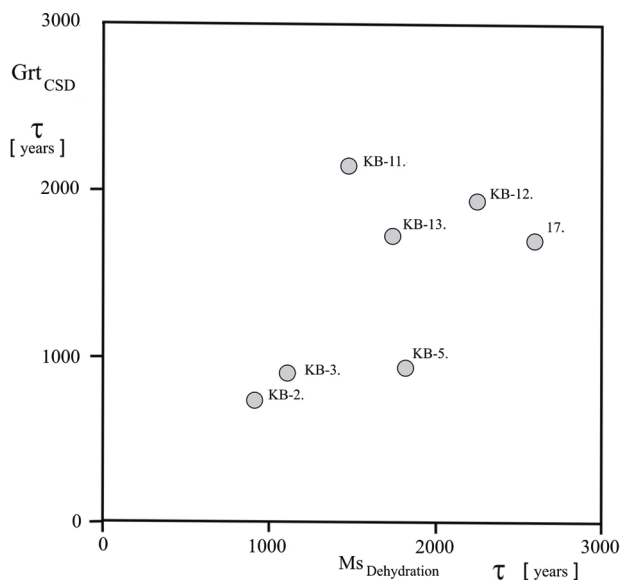
aureole. The small overstep required for nucleation during this reaction, compared with the large oversteps required for muscovite breakdown, may be related to differences in the interfacial energies, the rock strain and the structural mismatch between mineral reactants and products.

Rapid water release and simultaneous volume decrease can produce high internal fluid pressure that causes decrease of rock strength and involves rheological rock failure. Thus the rock may undergo transition from brittle to ductile properties and the massive escape of over-pressured fluid phase is impeded. As the result of internal stress due to hydrofracturing, the rocks may break apart and associated volume contraction could trigger e.g. an earthquake tremor in active dehydration fault zones.

Porosity is generated rapidly by dehydration or decarbonation for quite small degrees of overstepping, causing enhanced permeability behind the reaction front. This zone of enhanced permeability may provide a channelway for fluid advection parallel to the reaction front, even though the front itself advances by diffusion. The enhanced permeability will lower fluid pressure, creating an effective difference between fluid pressure and lithostatic pressure. Reaction rates here are sufficiently fast, that is, over times of tens of years at any point (Balashov and Yardley, 1998).

High fluid pressure could favour hydraulic fracturing and cataclastic deformation what may lead to the rapid fluid phase segregation especially along propagating fractures. The fluid infiltration and canalization could be a rapid process accompanied by cataclastic deformation involving rock dilatancy.

It is generally assumed that deformation combined with fluid infiltration may lead to equilibrium in rocks, at least on a thin section scale and that thermobarometric methods may be applied to such rocks in order to deduce the P-T conditions of metamorphism. In some rocks this is not the case and relics of the early assemblages survived the subsequent infiltration and deformation. Consequently, application of a geothermometer may lead to false temperature estimation.

It must be strongly emphasized that direct textural evidence for disequilibrium mineral composition in many rocks is not apparent (Koons et al., 1987; Frueh-Green, 1994). If such phases are not recognized as being metastable, the application of the thermobarometric techniques will clearly result in incorrect estimates of the P-T paths followed during metamorphism.

The kinetic data and the calculated recrystallization residence time $\tau \approx$ 740–2180 yrs obtained in this study (Tabs. 2, 3 and 4) support the presumption that the water release and associated volume decrease realized through metamorphic reactions can occur rapidly in the periplutonic zones.

Consequently, the recrystallization reactions in the crust may be swift and in the time frame spanning in decades or centuries rather than in long geologic time scale. Study may also emphasize a link between regional metamorphism and earthquake generation.

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