

Data Repository Item

Part A: Database

sample	d_g [μm]		d_p [μm]	f_p [fraction]	$Z=d_p/f_p$ [μm]	T [$^{\circ}\text{C}$]
Gs-126-a	10.6	\pm 3.2	0.81	0.065	13	356
Gs-126-b	17.9	\pm 7.0	1.31	0.003	461	356
Gs-126-c	11.8	\pm 4.8	1.00	0.039	26	356
Gs-126-d	12.6	\pm 4.7	0.75	0.036	21	356
Gs-126-e	11.2	\pm 3.7	1.06	0.061	17	356
Gs-126-f	13.1	\pm 5.1	0.64	0.021	30	356
Gs-126-h	18.1	\pm 7.5	0.71	0.001	550	356
Gs-126-i	17.1	\pm 5.7	3.45	0.013	265	356
Gs-126-k	16.3	\pm 6.2	1.83	0.033	55	356
Gs-126-m	18.2	\pm 6.6	2.90	0.025	116	356
Gs-126-n	16.7	\pm 6.0	1.40	0.012	117	356
Do-28-a	12.2	\pm 4.0	0.95	0.076	12	361
Do-28-b	14.0	\pm 4.9	1.10	0.076	15	361
Do-28-c	22.4	\pm 9.1	1.17	0.010	113	361
Do-28-d	20.7	\pm 10.2	1.06	0.004	281	361
Do-28-e	18.8	\pm 6.6	2.98	0.061	49	361
Do-28-g	22.8	\pm 8.1	3.79	0.006	632	361
Do-24-a	25.6	\pm 10.2	2.68	0.020	134	371
Do-24-b	26.8	\pm 8.8	2.36	0.011	215	371
Do-24-c	20.2	\pm 8.5	4.81	0.133	36	371
Do-24-d	26.3	\pm 10.8	1.64	0.029	56	371
Do-24-e	20.6	\pm 7.3	3.25	0.086	37	371
Do-24-f	22.5	\pm 9.3	3.17	0.074	43	371
Do-24-g	27.2	\pm 9.9	2.03	0.004	508	371
Do-24-h	28.4	\pm 11.5	3.01	0.010	301	371
Do-24-i	21.0	\pm 8.2	3.44	0.075	46	371
Do-24-m	22.2	\pm 9.5	4.3	0.126	34	371
Do-24-n	24.9	\pm 11.2	3.87	0.076	51	371
Do-24-o	21.6	\pm 9.4	3.94	0.097	41	371
Do-24-p	23.8	\pm 10.3	3.31	0.057	58	371
Jung-986-a	29.4	\pm 11.7	1.90	0.060	31	380
Jung-986-b	27.0	\pm 10.5	2.00	0.085	24	380
Jung-986-c	27.1	\pm 10.0	2.21	0.069	32	380
Jung-986-d	19.7	\pm 7.8	1.42	0.077	19	380
Jung-986-e	29.0	\pm 11.6	1.03	0.025	42	380
Jung-986-f	39.0	\pm 12.8	1.39	0.011	131	380
Jung-986-g	40.6	\pm 13.2	0.79	0.002	337	380
Jung-986-h	31.4	\pm 12.5	1.10	0.031	36	380
Jung-986-i	40.2	\pm 13.3	1.11	0.007	151	380
Jung-986-m	37.4	\pm 12.2	0.98	0.011	89	380
Jung-986-k	39.7	\pm 13.0	1.14	0.011	104	380
Jung-986-n	37.0	\pm 16.5	1.56	0.008	195	380
Jung-986-o	34.3	\pm 11.0	3.18	0.038	84	380

Table DR1: Database for Figure 2 presenting microstructural parameters of carbonate mylonites with variable second-phase contents. Note that sheet silicates are the predominating second-phase in these samples. d_g : calcite grain size, d_p : second phase grain size, f_p : second phase volume fraction, Z : Zener parameter = d_p/f_p , T : temperature.

Part B: Equation derivations

Temperature-dependent Zener relation

$$d_g = c \cdot Z^m, \text{ where } Z = \left(\frac{d_p}{f_p} \right) \quad \text{eq. 2}$$

Eq. 2 relates Z [μm], as ratio of the grain size d_p [μm] and the volume fraction of the second-phases f_p , in dependence of a dimensionless exponent m and a constant c [μm^{1-m}] to the size of the matrix grains d_g [μm]. This equation is valid only for isothermal conditions but a temperature-dependence can be incorporated in the following form:

$$d_g = c' \cdot \exp(-Q_g / RT) \cdot Z^m \quad \text{eq. 3}$$

where Q_g [J mol^{-1}] is the activation energy for growth of the matrix grain and c' [μm^{1-m}] is a constant, R is the gas constant and T is the temperature.

Calculation of the activation energy Q_g and the constant c'

The basic equation

In order to reduce first the dimensions in eq. 3, both sides of the equation are divided by [μm]. Then the logarithm is taken.

$$\log(d_g) = \log(c') + m \cdot \log(Z) + (-Q_g / RT) \cdot \log(e) \quad \text{eq. 3.1}$$

For a specific Z value, the terms $\log(c')$ and $m \cdot \log(Z)$ are constant and can therefore be replaced by $c'' = \log(c') + m \cdot \log(Z)$, yielding in:

$$\log(d_g) = c'' + (-Q_g / R) \cdot \log(e) \cdot 1/T \quad \text{eq. 3.2}$$

Taking a series of d_g values for different T but constant Z and plotting them in a $\log(d_g)$ versus $1/T$ diagram, the slope s_l of the resulting linear trend corresponds to $(-Q_g/R) \cdot \log(e)$. Hence, Q_g can be calculated by eq. 3.3

$$Q_g = (s_l \cdot R) / \log(e) \quad \text{eq. 3.3}$$

Finally, c' can be calculated by solving eq. 3:

$$c' = d_g / (Z^m \cdot \exp(-Q_g / RT)) \quad \text{eq. 3.4}$$

Second-phase grain growth

The basic equation

The data obtained from field samples subjected to different deformation temperatures suggest a power law relationship between d_p and f_p

$$d_p = k \cdot f_p^n \quad \text{eq. 4}$$

with a dimensionless and constant exponent n and a constant k [μm]. Since second-phase coarsening is temperature dependent, an activation energy term can be added in the same manner as already done for eq. 3.

$$d_p = k' \cdot \exp(-Q_p / RT) \cdot f_p^n \quad \text{eq. 5}$$

where k' [μm] is a constant and Q_p [J mol^{-1}] the activation energy for second-phase growth.

Calculation of the activation energy Q_p and the constant k'

To calculate Q_p , both sides of eq. 5 are divided in a first step by [μm] to eliminate the units and the logarithm is taken.

$$\log(d_p) = \log(k') + n \cdot \log(f_p) + (-Q_p / R) \cdot \log(e) \cdot (1/T) \quad \text{eq. 5.1}$$

For a series of data with constant f_p but different T and d_p , the term $\log(k') + n \cdot \log(f_p)$ is constant and can be replaced by k'' :

$$\log(d_p) = k'' + (-Q_p / R) \cdot \log(e) \cdot (1/T) \quad \text{eq. 5.2}$$

Hence, in a graph $\log(d_p)$ versus $1/T$, the slope s_2 corresponds to $(-Q_p/R) \cdot \log(e)$ allowing the calculation of Q_p . Using the new Q_p , eq. 5 can be solved for k' .

The final equation

The variable d_p , which is incorporated in the Z term of eq. 3, can now be replaced by eq. 5 resulting in:

$$d_g = c' \cdot \exp(-Q_g/RT) \cdot \left(\frac{d_p}{f_p} \right)^m = c' \cdot \exp(-Q_g/RT) \cdot \left(\frac{(k' \cdot \exp(-Q_p/RT) \cdot f_p^n)}{f_p} \right)^m$$

which finally yields in:

$$d_g = c' \cdot k'^m \cdot \exp(-Q_g/RT) \cdot \left(\exp(-Q_p/RT) \right)^m \cdot f_p^{m(n-1)} \quad \text{eq. 6}$$

Eq. 6 shows that d_g basically depends on four parameters: (1) the constants c' and k' , (2) & 3) activation terms for growth of the matrix grain and growth of the second-phase and (4) the volume fraction of the second-phases.

The role of the pre-exponential constants

Normal grain growth for monomineralic aggregates is classically defined by:

$$d_g^n - d_{g0}^n = k \cdot \exp(-Q/RT) \cdot (t - t_0) \quad \text{eq. 1}$$

where d_g and d_{g0} are the final and starting grain size, respectively, n is the growth exponent, k a pre-exponential constant, Q an activation energy, R the gas constant, T the

temperature and $(t-t_0)$ the time interval during which grain growth occurred. Here, the pre-exponential parameter k can be subdivided into:

$$k = C' \frac{\Omega D_{gb}}{\delta k_{BZ} T} \gamma \quad \text{eq. 1.1}$$

indicating that normal grain growth also depends on parameters like a constant C' , the unit cell volume Ω , the grain boundary diffusion coefficient D_{gb} , the grain boundary width δ , the Boltzman factor $k_{BZ}T$ and the interface specific Gibbs free energy γ (see also Evans et al., 2001).

Similar to the pre-exponential constant of eqs. 1 and 1.1, eq. 6 could additionally be used to obtain information about mass transfer in case of two-phase systems (e.g. calcite – muscovite). Here, the constants k' and c' also comprise information with respect to mass transfer coefficients. In comparison with eq. 1.1, the parameters C' , Ω , δ , k_{BZ} and γ will be nearly constant for both k' and c' , i.e. for calcite and the second phases, respectively. The only parameter variable for different geologic environments will be the mass transfer coefficient. In this way, mass transfer coefficients for second-phase minerals can be investigated for polymineralic aggregates undergoing coarsening via dissolution - mass transfer - precipitation cycles. Since, mass transfer is drastically enhanced under wet conditions, the mass transfer coefficient may deliver additional information about the fluid activity in such systems. This information is crucial for comparison of identical polymineralic rock types from different geological environments to learn more in a quantitative way about the processes associated with the microstructural evolution.