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Autor: Frank, Erik / Stettler, Anton

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K-Ar and ^{39}Ar-^{40}Ar systematics of white K-mica from an Alpine metamorphic profile in the Swiss Alps

by *Erik Frank** and *Anton Stettler***

Abstract

Mineralogical, K-Ar and ^{39}Ar-^{40}Ar isotope data are reported of metamorphic white K-mica (illite, muscovite) from Triassic phyllites, collected along a NW-SE profile of increasing Alpine metamorphism from the lower greenschist facies into the upper staurolite zone. Our study demonstrates that the formation of illite in the *low grade* rocks could not be dated by the K-Ar method. Mineralogical investigations show that the illites were formed by the reaction series mixed-layer illite/montmorillonite → $1M_d$ illite → $2M_l$ illite. Their disturbed ^{39}Ar-^{40}Ar spectra indicate that they were only rejuvenated but did not complete recrystallize during Alpine metamorphism. Diffusion was an important mechanism for the $^{40}Ar_{radiogenic}$ loss, and was controlled by temperature as well as deformation processes. Apparently elements were rearranged during Alpine metamorphism within preserved crystal lattices of presumably diagenetic origin. Complete convertion of $1M_d$ to $2M_l$ illite indicates the beginning of total outgassing of all pre-existent radiogenic argon.

$2M_l$ muscovites from the *higher grade* part of the metamorphic profile show rather uniform K-Ar ages of 8–13 my which indicate the time at which the loss of radiogenic argon essentially ceased. We conclude that complete recrystallization of muscovite, with breaking up all lattice bonds, occurs at higher temperatures than those critical for argon retention.

1. INTRODUCTION

White K-mica is an important petrogenetic indicator for metamorphic processes in sediments: the reaction sequence smectite → smectite/illite → illite → muscovite can be used as a sensitive tool to estimate the grade of diagenesis and metamorphism in lowgrade metasediments (DUNOYER 1970, HOWER et al., 1976). At higher metamorphic grade, muscovite is very common in pelitic schists of greenschist and amphibolite facies and can be used to obtain important informations on intensive parameters (temperature, pressure, activity of H_2O) as was deduced by GUIDOTTI and SASSI (1976).

* Mineralogisch-petrographisches Institut, University of Berne, Switzerland, present address: Mineralogisch-petrographisches Institut, Universität Basel, Bernoullistr. 30, CH-4056 Basel.

** Physikalisches Institut, University of Berne, Switzerland.

In the present study the regional increase of metamorphic grade within a Triassic phyllite formation in the Swiss Alps is investigated by mineralogical methods and K-Ar isotope analyses on white K-mica. The geological significance of K-Ar data and possible relations between mica mineralogy and the Alpine metamorphism are discussed in the light of the results obtained by the ^{39}Ar-^{40}Ar stepwise heating experiments.

2. GEOLOGICAL SETTING

Mesozoic sediments can be followed from the outer border of the Swiss Alps (Helvetic nappes) into the Pennine zone (Fig. 1). These sediments have been overprinted only by the Alpine metamorphism and are therefore well suited for a study of the Alpine thermal events on a regional scale (NIGGLI + NIGGLI 1965, WENK 1962, TROMMSDORFF 1966). The regional distribution of index minerals (e. g. stilpnomelane and staurolite) and of isogrades (calcite-diopside) indicate

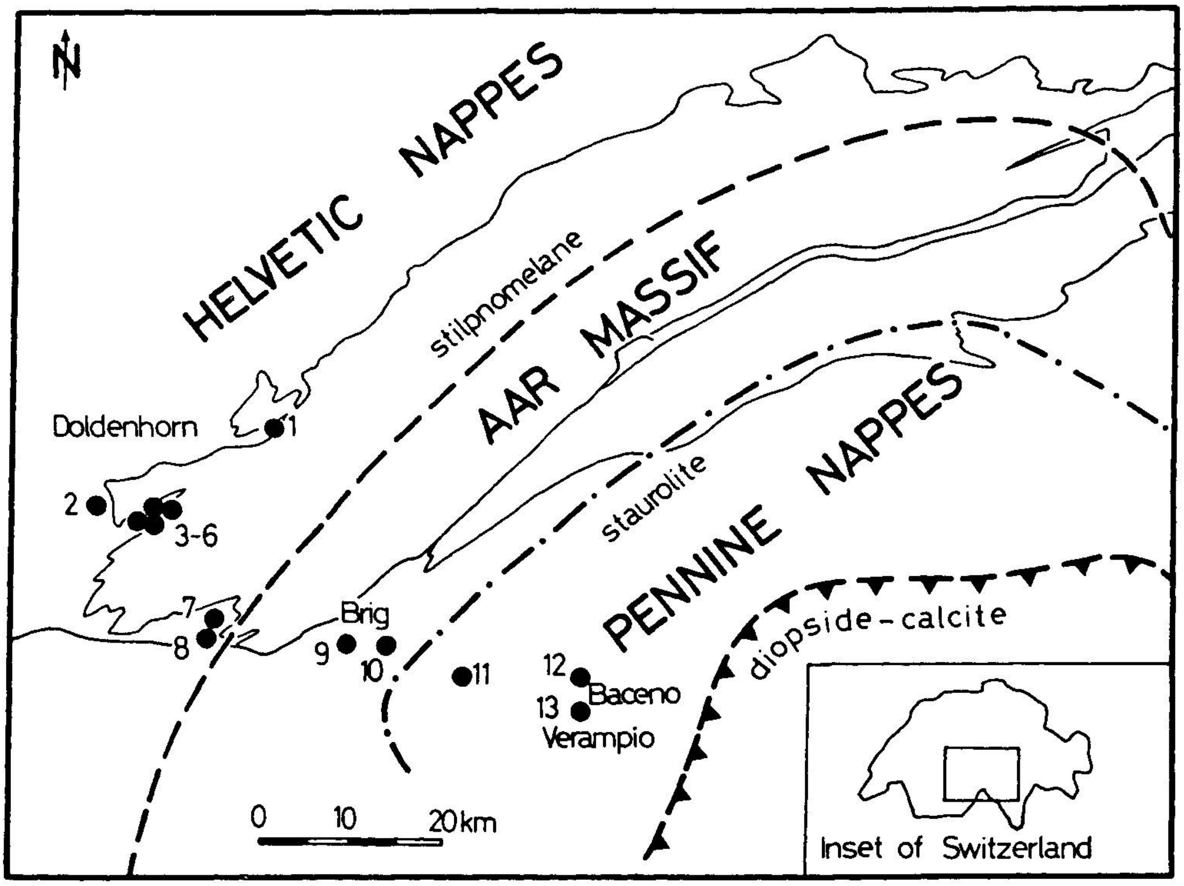

Fig. 1: Generalized geological map of the Central Alps illustrating our sampling localities (Ns. 1-13). Metamorphic mineral zones (Stilpnomelane, staurolite) after NIGGLI and NIGGLI 1965, diopside-calcite isograde after TROMMSDORFF (1966).

an increase of metamorphic grade from the lowest greenschist facies in the NW (Helvetic nappes) to the higher amphibolite facies in the SE (Fig. 1).
Upper Triassic rocks may be traced more or less continously over the whole profile. Mineralogical changes as well as the behaviour of K-Ar isotopes in white K-mica may so be studied in one single type of rock. The geological history and mineralogy of this formation is well known from the detailed work of FREY (1969) on a corresponding cross section in the eastern part of the Swiss Alps (Glarus Alps). This unit was therefore chosen for a detailed K-Ar and ^{39}Ar-^{40}Ar study. The sampling region extends from the zone of the Helvetic nappes (Doldenhorn) to the Pennine zone (Baceno-Verampio, cf. Fig. 1).

Metamorphic P-T conditions can be estimated from fluid inclusion studies on fissure quartz reported by MULLIS (1979) which indicate minimum temperatures of about 260 °C and pressures of 1-2 kb in the low-grade Doldenhorn area, whereas mineralogical geothermometry and barometry suggest P-T conditions of about 400 °C/2-4 kb near Brig, raising up to a maximum of 580-600 °C/5-7 kb at Verampio (FRANK 1979).

3. EXPERIMENTAL PROCEDURE

3.1. Sample preparation

Mica size fractions were separated out of about 500 g of powdered rock sample by sedimentation in Atterberg cylinders. Any carbonate minerals present were removed by dissolution in 5% acetic acid and subsequently the mica fractions were rinsed with distilled water. All fractions were filtered and dried at a temperature of about 40 °C. Three samples (40-60 mesh) were separated by classical techniques. Sample purity was determined by detailed x-ray diffraction analyses using the method devised by HENDERSON (1971), PETERS (1965) and FREY (1978).

3.2. X-ray analyses

Illite and muscovite clay fractions were analyzed by X-ray diffractometry and Guinier camera technique. D-spacings were determined using Si or quartz as internal standards, with an accuracy better than ±0.002 Å. The illite crystallinity index after KÜBLER (1967) was measured as the width at half peak height of the first illite basal reflection at 10 Å, using standardized conditions. The illite polytypes were determined following the method of MAXWELL et al., (1967) and REYNOLDS (1963). The present study indicates that the (025), (115) and (116) reflections of $2M_1$ muscovite can be used for identification of this polytype, while the d (2.57 to 2.59) region contains peaks of $1M_d$ and $2M_1$ mica. For quantitative estimation of $2M_1$ and $1M_d$ contents we used the method described by REYNOLDS (1963).

A Cambridge scanning electron microprobe was used to investigate the separated mica fractions.

3.3. Conventional K-Ar analyses

Potassium was determined in duplicate by flame photometry analyses. The isotopic composition and absolute amounts of argon were measured on a mass spectrometer (Varian Mat GD 150) using a ^{38}Ar spike (from Prof. Clusius, Zürich), previously calibrated against the B4 muscovite standard of Berne. Further analytical details are described by PURDY (1972, 1976). The following constants have been adopted in the present work (STEIGER + JÄGER 1977):

abundance of ^{40}K = 1.17×10^{-4} mole/mole

$\lambda (^{40}K_\beta)$ = $4.962 \times 10^{-10}/y$

$\lambda (^{40}K_\varepsilon) + \lambda'(^{40}K_\varepsilon)$ = $0.581 \times 10^{-10}/y$

atomic ratio $^{40}Ar/^{36}Ar_{atmospheric} = 295.5$

3.4. ^{39}Ar-^{40}Ar analyses

Five samples were prepared for irradiation using a technique described earlier (STETTLER et al., 1973, 1974). Together with four B4 muscovite monitors (JÄGER et al., 1963) they were exposed to fast neutrons in the core of the FR-2 reactor at Karlsruhe. All samples were enclosed in a Cd envelope within a single Harwell-type capsule. Spatial variations in neutron flux were monitored by Ni wires attached to each individual sample vial. The relative uncertainty of the fast neutron-flux values could thus be reduced to $\leqq 1\%$ by measuring the Co^{58} activities. The average fluence for neutrons with energy above 0.1 MeV was 3.46×10^{17} cm^{-2}. In view of the low Ca/K ratios of the samples (Ca/K $\leqq 0.2$) Ar interferences from neutron reactions on Ca were negligible. Contributions from K were corrected using $^{38}Ar/^{39}Ar/^{40}Ar = 0.0138/1.0/0.0055$ as inferred from K_2SO_4 salts irradiated previously in the same reactor position (MAURER 1973). ^{38}Ar produced by ^{37}Cl (n, β, γ) was used to estimate the Cl content of the samples comparing with previously irradiated NaCl (STETTLER et al., 1979). For the conversion factor $c_{39}(K)$ determined from the B4 muscovite monitors we obtained

$$c_{39}(K) = 1.108 \times 10^{-5}\ cm^3\ STP\ ^{39}Ar/g\ K.$$

No Ca standards was included in this irradiation run. However, the results of a series of 40 irradiations performed during the last six years reveal an extremely constant ratio

$$(Ca/K) / (^{37}Ar/^{39}Ar) = 1.95 \pm 0.03$$

so that it seems reasonable to assume a conversion factor

$$C_{37}(Ca) = 5.68 \times 10^{-6}\,cm^3\,STP\,^{37}Ar/g\,Ca.$$

After neutron irradiation the illite samples were degassed stepwise and the argon composition measured on-line with the double magnetic mass spectrometer assemblage at the Physikalisches Institut in Bern (SCHWARZMÜLLER, 1970).The detailed argon results given in Table 3 are corrected for spectrometer blanks and non-linearities, and the decay of ^{37}Ar and ^{39}Ar using $\lambda_{37} = 0.01975$ day^{-1} and $\lambda_{39} = 7.2 \times 10^{-6}$ day^{-1}.

Extraction blanks (in 10^{-8} cm^3 STP Ar^{40}) were typically 0.5 for $T \leqq 1100\,°C$ increasing to 2.5 at 1600 °C. To separate the radiogenic argon component all ^{36}Ar was assumed to be atmospheric. Apparent gas retention ages were calculated using the constants listed in section 3.3. and an adopted age of 18.3×10^6 y for the B4 muscovite monitors (JÄGER, 1963). All errors given correspond to the 95% confidence level. For further analytical details see SCHWARZMÜLLER (1970) and STETTLER et al., (1973, 1974).

4. RESULTS AND DISCUSSION

4.1. Mineralogy

Chemical composition, crystallinity index and polytype of illite as well as its paragenesis can be used to estimate the physico-chemical conditions of its formation during diagenetic and low-grade metamorphic processes (DUNNOYER 1970, FREY 1969, KÜBLER 1967). According to KÜBLER (1967) a crystallinity index of <7.5 marks the beginning of the anchizone, whereas values of less than 4.0 already indicate greenschist facies conditions. As inferred from field studies (FREY 1969) and experimental work (YODER & EUGSTER 1955), $1M_d$ illite tends to recrystallize to $2M_1$ K-mica in the anchizone or at the beginning of greenschist facies.

For the present study different clay size fractions of each sample were analyzed in detail by X-ray diffraction methods and by electron-microscopy in order to see whether different mica generations are present. Unmetamorphosed equivalents of our Upper Triassic formation can be found in the Swiss Jura

Locality No.	Sample No.	Whole rock mineralogy*	Type of white K-mica	d(002) (Å)	b_o (Å)	Polytype	Crystallinity index (KUEBLER)	Estimated P-T conditions of alpine metamorphism
	KAW							
1	1844	Ill,trChl,Qz, trDol,Häm	phengitic illite	9.961	9.048	$1M_d, 2M_1$	3.8	260-300°/ 1-2 kb
2	1764	Ill,Qz,Ab, tr Dol		9.970	9.048	$1M_d, 2M_1$	3.4	
3	1840	Ill,Chl,Qz, trAb		9.968	9.018	$1M_d, 2M_1$	3.2	
4	1846	Ill,trChl,Qz, trDol		9.964	9.042	$1M_d, 2M_1$	3.6	
5	1762	Ill,Chl,Qz, trAb,Dol		9.963	9.042	$1M_d, 2M_1$	3.8	
6	1763	Ill,trChl,Qz, trAb		9.968	9.018	$1M_d, 2M_1$	3.9	
7	1760	Ill,trBi,Qz, Ab		9.957	9.054	$2M_1$	3.5	380°C/2-3kb
8	1761	Ill,trChl,Qz, trAb,Cc		9.956	9.030	$2M_1$	3.6	
9	1757	Mu,Qz,Cc	muscovite	9.954	8.982	$2M_1$	3.0	420°C/3-4kb
10	1714	Mu,trChl,Qz		9.942	8.988	$2M_1$	2.6	
11	1703	Mu,Bi,Qz,Plag, Cc		9.975	8.994	$2M_1$	2.4	530°C/4-6kb
12	1709	Mu,Bi,Qz,Plag, Gr		9.940	9.000	$2M_1$	2.4	580-600°C / 5-7 kb
13	1755	Mu,Bi,Qz,Plag, Gr,Stau		9.938	8.994	$2M_1$	2.5	

*Abbreviations used:

Ill = illite, Chl = chlorite, Bi = biotite, Qz = quartz, Ab = albite, Plag = plagioclase, Gr = garnet, Cc = calcite, Dol = dolomite, Stau = staurolite, Häm = hematite, Mu = muscovite, tr = traces

Table 1: Mineralogy and white K-mica data of Upper Triassic rocks in the metamorphic profile Doldenhorn-Verampio.

(claystones). The clay mineralogy of these rocks is well known by the work of PETERS 1964 and FREY 1969: the clay-fractions consist mainly of 1M illite and an irregular mixed-layer illite/montmorillonite. No detrital high temperature 2M white mica is present.

Following this formation along our metamorphic profile from NW (Doldenhorn) to SE (Brig to Verampio-Baceno), the transition of phengitic illite to muscovite can be studied with increasing grade (cf. Table 1): in the low-grade samples, illite shows strong basal reflections (001), (002) and (003) and apparently no expandable mixed-layer phase as tested by treating the samples with ethylene-glycol. The measured illite crystallinity values vary from 3.9 to 3.2 in the low-grade samples of the Helvetic nappe zone and decrease to values of less than 3.0 in the higher grade rocks of the Brig-Verampio region (cf. Table 1).

The chemical composition of white K-mica was estimated from d(002) and b_0 spacing values. The relation between these two parameters is illustrated in Fig. 2 using the grid proposed by GUIDOTTI & SASSI (1976). This diagram allows an estimate of femic contents (Fe, Mg) in terms of RM-values and the evaluation of the Na/Na+K ratio of white K-mica. The low-grade illites (filled circles in Fig. 2) show strong compositional differences in respect to femic contents, pointing to a solid-solution muscovite-phengite for our samples. The chemical

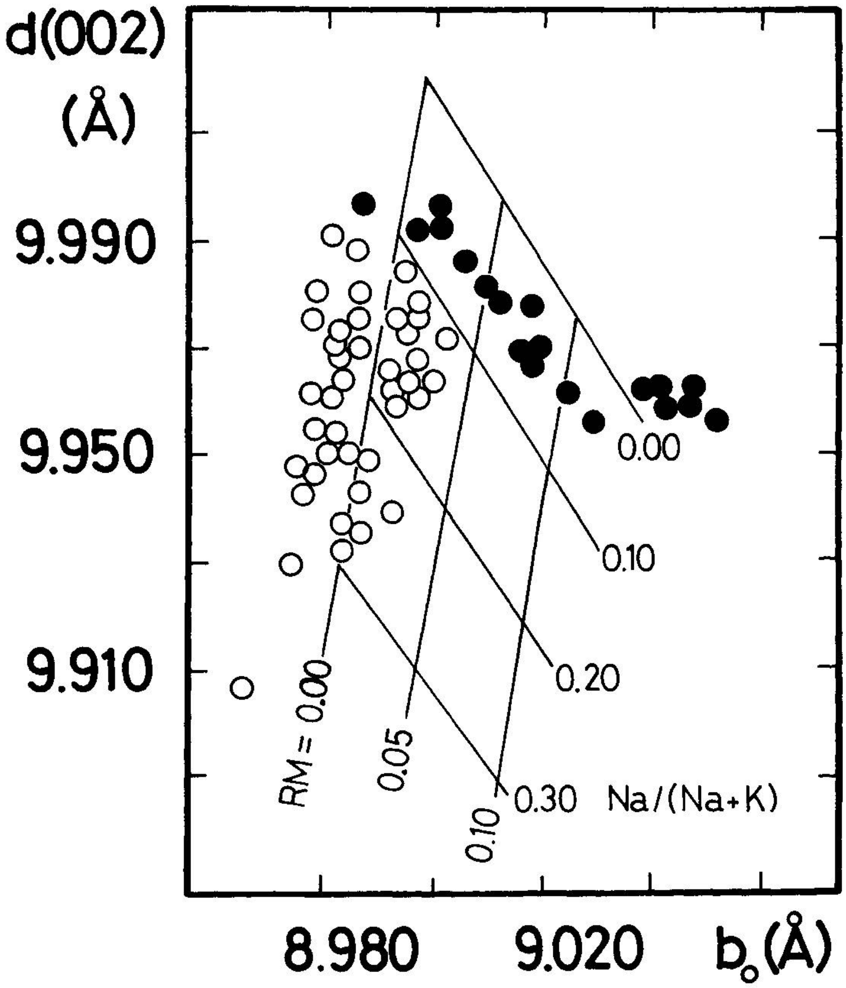

Fig. 2: Observed variation of white K-mica composition from Mesozoic rocks of the metamorphic profile Doldenhorn-Verampio (FRANK 1979), using the b_0-d(002) plot of GUIDOTTI and SASSI 1976. Filled circles: low-grade illites; open circles : higher grade muscovites (greenschist- to higher amphibolite facies). RM = mol. prop. 1/2 Fe_2O_3 + mol. prop. FeO + mol. prop. MgO.

Table 2: K-Ar and mineralogical data of illite and muscovite samples.

Locality No.	Sample No.	Grain Size Fraction	IK*	sample purity**	Estimated $(2M_1/1M_d+2M_1)$ ratio in %	K (%)	$^{40}Ar_{rad}$ $(10^{-6}cc/g)$	$^{40}Ar_{rad}/^{40}Ar$	age (my)	
	KAW									
1 Obersteinberg	1844	<1μ	3.6	100% ill	55	7.29	9.65	0.805	33.5	± 1.2
		1-2μ	3.6	98% ill,2% hem,tr qz	45	7.07	9.64	0.780	34.5	1.3
		2-6μ	3.8	97% ill,3% hem,tr qz	45	6.11	9.40	0.660	38.8	1.8
2 Ferdenpass	1764	<1μ	3.4	100% ill	80	8.26	5.99	0.694	18.4	0.8
		1-2μ	3.5	100% ill	80	8.37	6.27	0.773	19.0	0.7
3 Kummenalp	1840	<1μ	3.2	100% ill	74	7.42	6.32	0.713	21.6	0.9
		1-2μ	3.2	92% ill, 6%chl,2% qz	67	6.86	6.48	0.743	24.0	0.9
		2-6μ	3.6	86% ill,12%chl,2% qz	60	6.10	6.47	0.711	26.9	1.1
		6-20μ	3.9	84% ill,13%chl,3% qz	62	5.46	6.90	0.773	32.0	1.2
4 Kummenalp	1846	<1μ	3.4	100% ill	75	7.53	6.64	0.651	22.4	1.1
		1-2μ	3.6	98% ill,2% qz	70	7.37	6.61	0.677	22.8	1.0
		2-6μ	3.6	96% ill,4% qz	72	6.18	5.99	0.589	24.5	1.4
5 Kummenalp	1762	<1μ	3.8	100% ill	80	7.67	5.95	0.674	19.7	0.9
		1-2μ	3.8	100% ill,tr qz	75	7.43	5.79	0.707	19.8	0.8
6 Kummenalp	1763	<1μ	3.8	100% ill	82	7.42	6.32	0.713	21.6	0.9
		1-2μ	3.9	96% ill, 4%chl,tr qz	70	6.86	6.48	0.743	24.0	0.9
7 Ausserberg	1760	2-3μ	3.5	98% ill, 2%qz	100	7.49	2.40	0.232	8.2	1.1
8 Ausserberg	1761	2-3μ	3.6	93% ill, 7%chl	100	6.62	2.14	0.287	8.2	0.9
9 Brig	1757	40-60 mesh	3.0	90% musc,8%chl,2%qz	100	5.04	1.88	0.558	9.5	0.5
10 Eisten	1714	6-20μ	2.6	92% musc,6%chl,2%qz	100	5.39	2.38	0.415	11.2	0.8
11 Alpe Veglia	1703	6-20μ	2.4	100% musc	100	7.75	3.68	0.454	12.1	0.8
12 Baceno	1709	40-60 mesh	2.4	100% musc	100	7.41	4.01	0.451	13.7	0.9
13 Baceno	1755	40-60 mesh	2.5	100% musc	100	7.64	3.91	0.414	13.0	1.0

*Illite crystallinity index after KUEBLER

** abbreviations for mineral names: ill = illite, musc= muscovite, chl = chlorite, hem = hematite, qz = quartz, tr = traces (<1%)

composition of these illites is mainly controlled by the limiting assemblage illite-albite and/or the host rock composition. With higher metamorphic grade, the transition of phengitic illite to muscovite occurs and the variation of the Na/Na+K ratio becomes more important (Fig. 2). As shown by FRANK (1979), the Na-content of muscovite, coexisting with paragonite, increases gradually with increasing metamorphic grade from Brig to Verampio from x_{Na}^{Mu} = 0.12 to 0.30: the estimated temperatures from the muscovite-paragonite solvus calibrations, using the method of ROSENFELD (1969), are consistent with other temperatures deduced from calibrated mineral equilibria. This indicates, that interlayer sites of white K-mica exchanged up to the thermal peak of Alpine metamorphism.

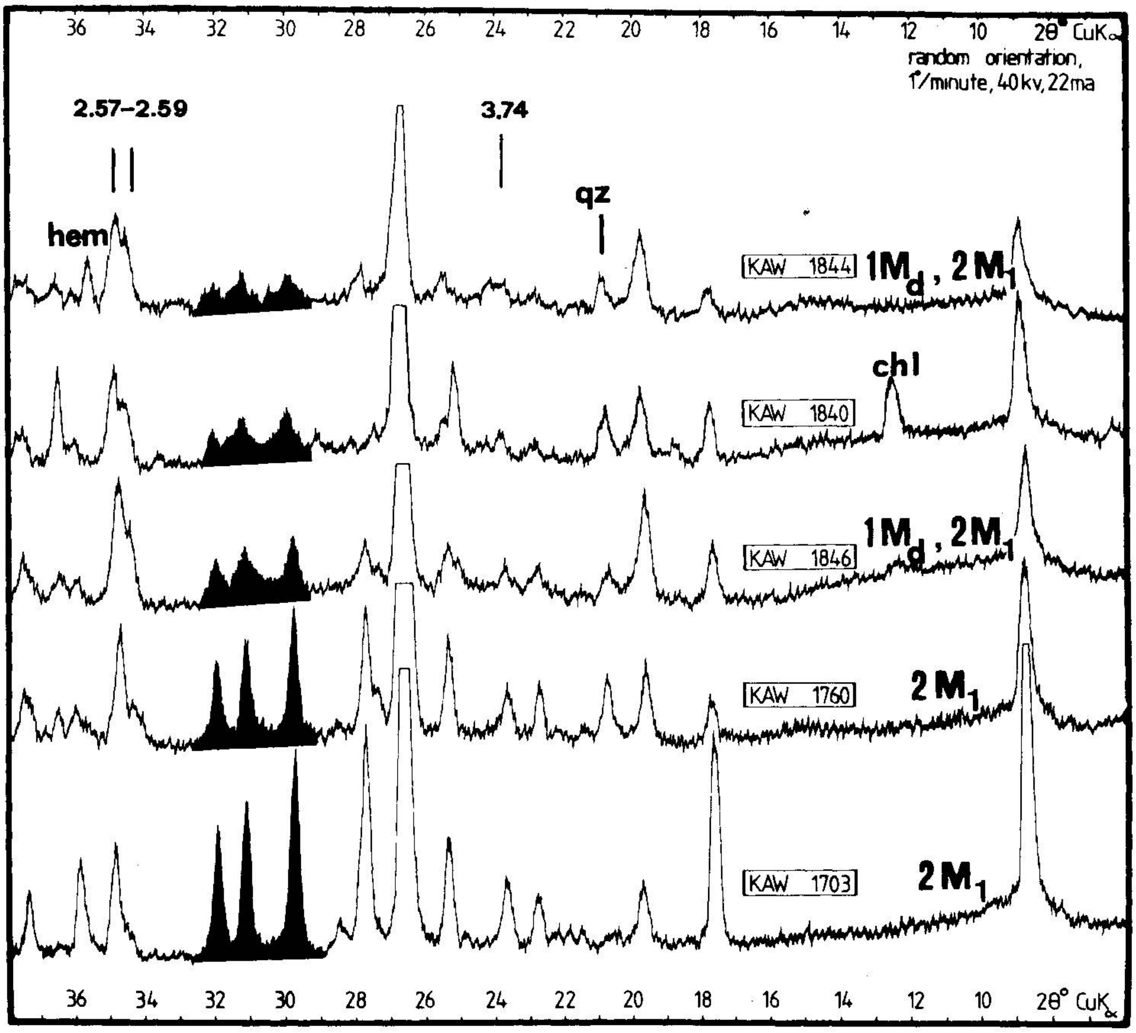

Fig. 3: X-ray patterns of illite ($1M_d$-$2M_l$) and muscovite ($2M_l$) (< 2μ fraction, randomly oriented specimens), shown as a function of increasing metamorphic grade. Sample 1844 contains 2% hematite, in sample 1840 chlorite is present too. The black area (peaks of 30-32° 2θ° CuK), diagnostic for $2M_l$-polytype, record a continous increase of $2M_l$ with increasing metamorphic grade. (For quantitative polytype determination, low goniometer speed of 1/4° per minute was used).

Polytype determinations indicate that the $1M_d$ structure is still present in the low-grade illites with approximately half its intensity. With increasing metamorphic grade the amount of $1M_d$ decreases continously whereas the $2M_1$-polytype becomes completely dominant in sample KAW 1760 (100% $2M_1$, cf. Table 2, Fig. 3). This systematic increase in the $2M_1/1M_d + 2M_1$ ratio is considered to be the result of increases in the metamorphic grade and excludes a detrital origin of the 2M material. Furthermore, analyzing different sizefractions of the same sample reveals slightly higher amounts of $2M_1$ polytype in the finer fractions. This may indicate that the $1M_d \rightarrow 2M_1$transformation also depends on grain size: finer grain size fractions recrystallize much better than coarser fractions. According to YODER & EUGSTER (1955) $1M_d$ muscovite is stable below temperatures of 200-350 °C, while a gradual transfer to the high temperature $2M_1$ polytype occurs above this temperature range. From their oxygen isotope results obtained on coexisting quartz-illite, ESLINGER & SAVIN (1973) determined a temperature of 350-400 °C for a complete conversion of $1M_d$ to $2M_1$ polytype. These data agree quite well with our present results since from sample KAW 1760 (100% $2M_1$) we estimate metamorphic temperatures of about 350-380 °C necessary to transform the $1M_d$ polytype completely.

From the mineralogical data, the systematic occurence of $1M_d$-$2M_1$ polytype as well as from the results obtained by FREY (1969) on a equivalent rock formation in the Glarus Alps we may suggest the following illite formation reaction:

mixed-layer illite/montmorillonite $\rightarrow$ phengitic illite + chlorite + quartz + H_2O

4.2. Conventional K-Ar systematics

The K-Ar data are given in Table 2. In Figure 4 all K-Ar ages on white K-mica of Mesozoic cover rocks (including «schistes lustrés») available along this cross-section are plotted versus metamorphic grade as was reported by FRANK (1979).

The results of the pesent study and related work (FRANK 1979) may be summarized as follows:

- All white K-micas give Tertiary ages between 8 and 39 my.
- The K-Ar ages of low-grade illites ($1M_d$-$2M_1$) decrease from 39 my to 8 my as their metamorphic grade increases. At the same time we observe a continous decrease in the amount of $1M_d$ polytype from about 50% to 0%, when the $2M_1$ polytype becomes completely dominant. Different grain size fractions of the same sample show a positive correlation between K-Ar ages and grain size.
- Higher grade muscovites ($2M_1$) give rather uniform K-Ar ages of 8-13 my over the whole metamorphic range from medium greenschist facies (Ausser-

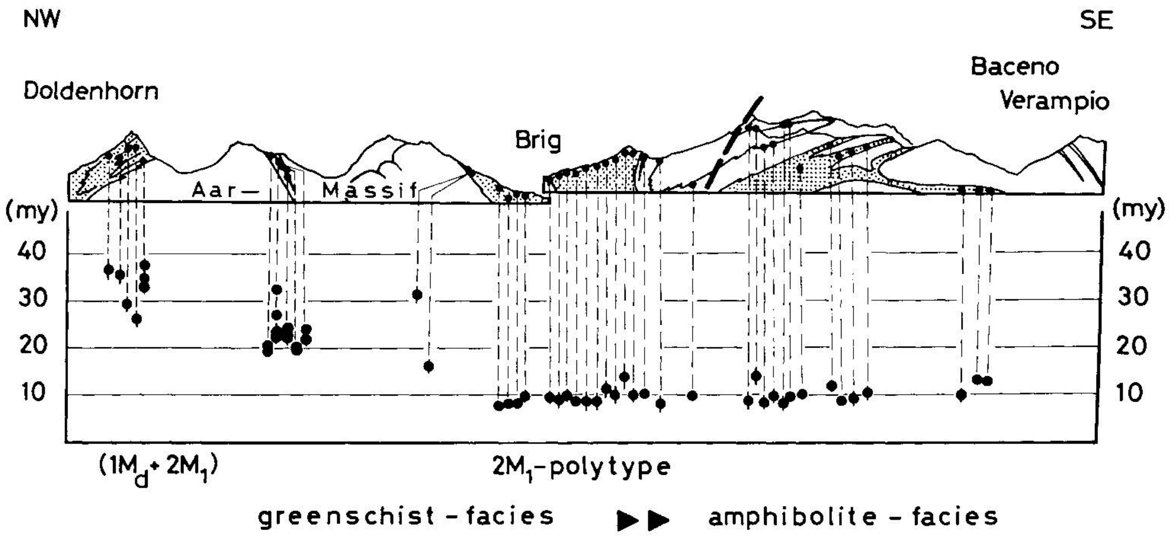

Fig. 4: Regional distribution of K-Ar ages of white K-mica in Mesozoic rocks (stippled area), plotted versus metamorphic grade (FRANK 1979). Dashed line indicates steeply dipping staurolite-isograd in the Simplon region according to STRECKEISEN and WENK (1974).

berg-Brig) up to higher amphibolite facies (Verampio-Baceno). No grain size dependency is observed since fine grained muscovite fractions (μ-size) have identical K-Ar ages as mm-size mica flakes.

- Microscopic observations indicate a syn- to late-kinematic recrystallization of muscovite.
- Phengite-rich illite and aluminium-rich muscovite of the same metamorphic zone show no differences in K-Ar age.
- Argon isotope analyses of fissure quartz from the low-grade zone (Doldenhorn-Brig) yielded locally large amounts of inherited $^{40}Ar_K$ (up to 14.0 x 10^{-6} cm^3 STP/g) incorporated in inclusions, indicating that radiogenic argon had not been expelled from the rock system. In the higher grade zone (Brig-Verampio) no radiogenic argon could be detected in quartz segregations (FRANK 1979). The problem of inherited or excess argon was studied in detail on fissure minerals from the Swiss Alps by PURDY & STALDER who showed that inherited $^{40}Ar_K$ is a very common phenomenon in low grade zones.

Based on conventional K-Ar data only, a geological interpretation of the low-grade illite ages is not possible unequivocally. In principle there are several possible reasons for the observed age distribution:

a) Presence of excess or inherited $^{40}Ar_r$ causing high ages.
b) Presence of different mica generations which are related to distinct thermal phases of the Alpine orogeny.
c) Heterogenous structure of the illite crystals containing sites of different retentivity for in situ produced radiogenic ^{40}Ar.
d) Homogeneous illite structure but varying metamorphic conditions at different sites.

To distinguish between these possibilities the ^{39}Ar-^{40}Ar incremental heating method was applied to a suite of key samples from the metamorphic profile.

4.3. ^{39}Ar-^{40}Ar systematics

The results are listed in Table 3, while Figure 5 relates the argon release patterns to the corresponding sample locations. From Table 2 we can see that 96-100% pure illite and muscovite samples were used, except sample KAW 1840 with 13% chlorite in the illite concentrate. As shown in the last column of Table 3, we find good agreement between conventional K-Ar ages and the total

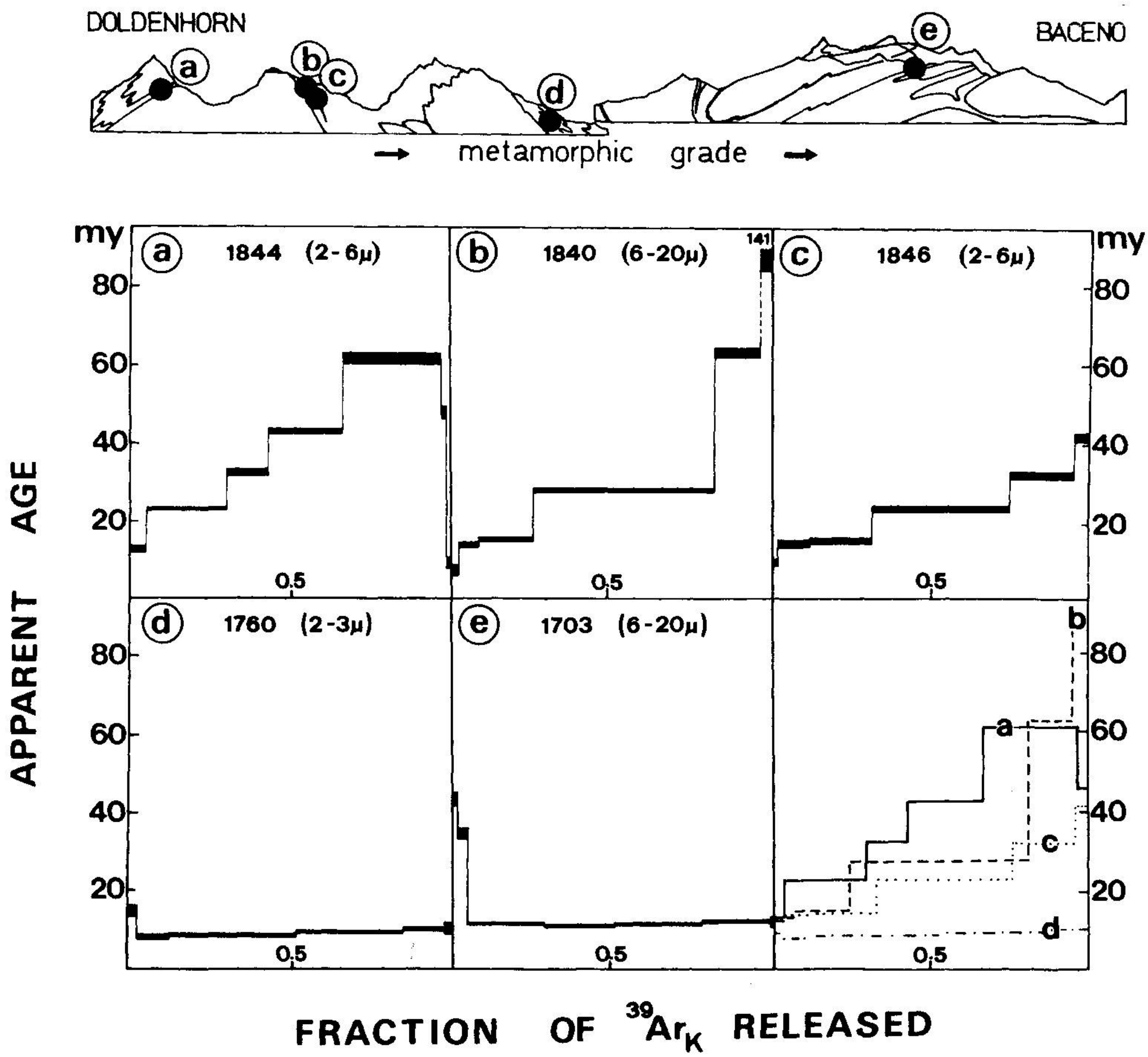

Fig. 5: Locations of four illites and one muscovite sample in the Alpine cross section and corresponding ^{39}Ar-^{40}Ar release patterns. Note the progressive modification of the release curves with increase of metamorphic grade from the Doldenhorn area to Baceno.

Table 3: Argon results from stepwise heating of neutron activated samples. Ar^{40} concentrations determined from ion-beam intensities.

Temp. (°C)	Ar^{40} (10^{-8} cm^3 STP/g)	Ar^{36}/Ar^{38}	Ar^{38}_{Cl}/Ar^{37} (x 10^{-2})	Ar^{39}_K/Ar^{37}	Ar^{40}/Ar^{36}	Ar^{40}/Ar^{39}_K	Ar^{40}_r/Ar^{40}	Apparent * Age (my)
Illite KAW 1844 (2 - 6 μ) / 79.11 mg								
475	87.7 ± 4.5	4.35 ± .50	5.0 ± 2.8	13.24 ± .25	332.2 ± 1.5	36.01 ± .20	0.110	11.50 ± .50
660	148.2 ± 8.0	1.57 ± .70	5.0 ± 2.5	13.07 ± .30	928 ± 50	11.66 ± .14	0.682	22.92 ± .80
700	116.2 ± 6.0	1.02 ± .45	4.9 ± 2.3	11.32 ± .18	1430 ± 45	14.32 ± .10	0.793	32.68 ± .80
740	213 ± 11	0.83 ± .45	5.5 ± 3.0	12.80 ± .25	2580 ± 140	16.99 ± .16	0.885	43.2 ± 1.1
845	380 ± 19	1.3 ± 1.3	5.7 ± 5.7	28.4 ± 1.5	5120 ± 150	22.94 ± .19	0.942	61.7 ± 1.5
965	24.1 ± 1.2	1.25 ± .25	6.5 ± 1.4	5.67 ± .30	1105 ± 30	23.11 ± .13	0.733	48.5 ± 1.2
1070	6.65 ± .35	1.84 ± .14	5.55 ± .60	1.860 ± .090	331.0 ± 4.0	30.20 ± .35	0.107	9.4 ± 1.0
1620	8.35 ± .40	2.05 ± .35	10.6 ± 2.0	9.50 ± .25	343.9 ± 3.5	12.570 ± .070	0.141	5.13 ± .40
Total	985	1.80	5.4	14.18	1355	18.13	0.782	40.68 (38.8)
Illite KAW 1840 (6 - 20 μ) / 90.33 mg								
460	85.5 ± 4.0	5.17 ± .35	1.5 ± 1.5	13.52 ± .70	307.8 ± 1.5	68.05 ± .40	0.040	7.9 ± 1.1
540	30.5 ± 1.5	4.9 ± 1.5	1.0 ± 1.0	17.95 ± .90	394.5 ± 5.5	19.78 ± .12	0.251	14.37 ± .70
710	102.4 ± 5.1	2.9 ± 1.4	3.5 ± 2.5	16.08 ± .25	543.7 ± 8.0	11.83 ± .11	0.456	15.60 ± .45
780	307 ± 15	3.2 ± 3.2	1.5 ± 1.5	27.70 ± .80	1810 ± 80	11.790 ± .070	0.837	28.42 ± .70
870	160.5 ± 8.0	2.8 ± 2.8	3.0 ± 3.0	29.3 ± 2.0	3140 ± 140	24.57 ± .14	0.906	63.5 ± 1.5
945	68.6 ± 3.5	2.00 ± .80	3.3 ± 1.6	7.22 ± .25	2970 ± 130	56.35 ± .35	0.900	141.6 ± 3.5
1050	12.25 ± .80	1.170 ± .045	2.85 ± .20	0.370 ± .070	411 ± 14	114 ± 20	0.280	90 ± 18
1620	9.65 ± .50	5.0 ± 3.0	-	2.43 ± .80	426 ± 13	34.9 ± 1.9	0.306	30.8 ± 2.5
Total	776	3.5	2.4	17.26	924.6	17.02	0.680	33.30 (32.0)
Illite KAW 1846 (2 - 6 μ) / 62.24 mg								
490	27.1 ± 1.4	4.45 ± .50	2.2 ± 1.3	5.15 ± .14	323.7 ± 1.9	38.07 ± .20	0.087	9.60 ± .70
600	51.3 ± 2.5	3.7 ± 1.2	1.4 ± 1.2	5.233 ± .080	439.3 ± 2.0	15.540 ± .080	0.327	14.70 ± .40
700	80.3 ± 4.5	0.87 ± .13	2.10 ± .30	1.346 ± .020	531 ± 16	11.41 ± .15	0.443	14.63 ± .70
750	157 ± 15	0.55 ± .13	3.30 ± .80	2.95 ± .20	1165 ± 90	11.00 ± .15	0.746	23.65 ± .90
850	98.4 ± 5.0	1.02 ± .55	3.2 ± 1.7	7.22 ± .12	1900 ± 25	13.460 ± .070	0.844	32.70 ± .80
960	18.00 ± .90	1.64 ± .25	1.65 ± .25	1.108 ± .020	705 ± 12	25.40 ± .20	0.581	43.3 ± 1.1
1100	9.07 ± .45	2.688 ± .090	2.63 ± .16	0.355 ± .012	301.5 ± 3.0	124.3 ± 3.5	0.020	7.1 ± 3.5
1620	8.90 ± .50	3.57 ± .15	1.77 ± .20	0.459 ± .011	312.7 ± 9.0	132.5 ± 4.0	0.055	21 ± 10
Total	450	1.22	2.60	2.580	723.2	13.43	0.591	22.92 (24.5)
Illite KAW 1760 (2 - 3 μ) / 59.90 mg								
300	5.20 ± .25	-	-	-	336 ± 14	2700 ± 2000	0.121	750 ± 500
460	187 ± 10	5.28 ± .30	2.8 ± 1.4	7.12 ± .20	312.1 ± 1.4	96.90 ± .70	0.053	14.9 ± 1.5
560	91.2 ± 4.5	5.0 ± 1.1	4.1 ± 2.0	54.5 ± 3.0	333.1 ± 2.0	23.40 ± .13	0.113	7.65 ± .45
690	284 ± 16	5.1 ± 3.0	0.45 ± .20	19.55 ± .35	402.8 ± 5.0	9.97 ± .18	0.266	7.70 ± .35
760	134.7 ± 7.0	4.2 ± 4.2	2.1 ± 1.1	37.53 ± .70	595.5 ± 5.0	6.574 ± .060	0.504	9.58 ± .25
840	56.2 ± 3.0	3.5 ± 3.5	3.0 ± 1.5	39.6 ± 1.3	710.0 ± 7.0	5.980 ± .050	0.584	10.11 ± .25
940	15.55 ± .80	5.0 ± 1.3	1.30 ± .70	15.4 ± 3.0	355.3 ± 6.0	21.24 ± .50	0.168	10.27 ± .80
1050	6.14 ± .30	5.6 ± 1.5	-	2.98 ± .25	352 ± 13	33.8 ± 3.5	0.162	15.8 ± 3.5
1620	9.27 ± .50	5.55 ± .70	-	12 ± 12	317.4 ± 7.0	223 ± 20	0.070	44 ± 14
Total	790	5.0	1.1	24.25	396.8	12.11	0.255	8.95 (8.2)
Muskovite KAW 1703 (6 - 20 μ) / 60.88 mg								
460	37.4 ± 1.9	3.98 ± .30	6.0 ± 2.0	4.95 ± .40	421.8 ± 4.0	81.3 ± 1.3	0.300	69.3 ± 2.5
515	22.8 ± 1.2	4.60 ± .80	1.6 ± 1.6	6.44 ± .45	446.5 ± 6.0	38.70 ± .35	0.338	37.6 ± 1.4
570	27.0 ± 1.4	2.50 ± .50	6.0 ± 2.0	5.60 ± .40	650 ± 11	29.80 ± .20	0.545	46.6 ± 1.3
615	31.3 ± 1.6	3.18 ± .70	3.2 ± 1.4	5.5 ± 1.3	579.0 ± 8.0	23.45 ± .19	0.490	33.00 ± .90
760	172.0 ± 8.0	4.5 ± 3.5	0.7 ± 0.7	12.75 ± .60	515.0 ± 3.5	9.418 ± .060	0.426	11.62 ± .30
840	125.3 ± 6.5	3.7 ± 3.0	2.2 ± 2.0	26.93 ± .50	680.0 ± 8.0	6.660 ± .070	0.566	10.90 ± .30
940	147.7 ± 7.5	4.0 ± 4.0	1.5 ± 1.0	24.00 ± .40	709.5 ± 5.0	6.833 ± .045	0.583	11.54 ± .30
1050	175.3 ± 9.0	4.6 ± 2.5	1.0 ± 1.0	12.85 ± .25	470.0 ± 2.5	11.760 ± .060	0.371	12.63 ± .35
1240	17.65 ± .90	6.3 ± 1.8	-	1.592 ± .040	359.0 ± 5.0	24.22 ± .25	0.177	12.40 ± .90
1620	9.57 ± .50	10.0 ± 5.0	-	1.60 ± .90	316.3 ± 7.0	1040 ± 250	0.065	188 ± 70
Total	766	4.2	1.4	14.88	541.7	9.867	0.455	12.97 (12.1)

* Total ages in parentheses correspond to K-Ar ages (cf. section 4.2)

^{39}Ar-^{40}Ar gas ages. A comparison of the concentrations of $^{39}Ar_K$ and K as obtained by the different methods (Table 2 and3) shows that neutron irradiated samples suffered some gas losses during and/or after irradiation. Similar argon losses have been observed previously when fine grained powder of lunar basalts was investigated by the ^{39}Ar-^{40}Ar technique (TURNER et al., 1974). However, it is important to note that the ^{39}Ar-^{40}Ar total degassing ages of our illite and muscovite samples apparently remained unchanged regardless of some argon loss.

From Figure 5 we can observe a progressive modification of the release patterns of our samples (a-e) from NW (Doldenhorn) to SE (Baceno-Verampio), indicating that these spectra are *controlled by metamorphic grade* along our profile.

Higher grade samples KAW 1760 ($2M_1$ illite) and KAW 1703 ($2M_1$ muscovite) both exhibit curves which can be either interpreted to reflect plateau (concordant ages) at 9.0 ± 1.3 my and 11.8 ± 1.0 my (95% ^{39}Ar released) respectively, or slight saddle-shaped spectra with minima at 7.6 ± 0.4 my and 10.9 ±0.3 my respectively. The high apparent ages at initial temperatures may indicate a previous preferential loss of ^{39}Ar relative to ^{40}Ar from grain surfaces by ^{39}Ar recoil (TURNER et al., 1974, HUNEKE et al., 1976) or, also by recoil, a transfer of ^{39}Ar from low retentive sample sites to sites of higher retentivity. If released during extraction over an appreciable temperature range, the displaced ^{39}Ar may not be visible by a sharp drop in the age curve. The slight increase of apparent ages toward higher temperatures might indicate that the samples were able to retain a very small amount of $^{40}Ar_r$ when subjected to a severe outgassing process. The superposition of both effects would yield slight saddle-shaped release patterns.

Low grade illites (KAW 1844, 1840, 1846) show disturbed release curves typical for samples which have lost $^{40}Ar_r$ by a degassing process since their initial crystallization: incremental ages becoming progressively older from low to high release temperatures ranging up to 141 my in KAW 1840. Diffusion calculations predict similar patterns for argon release from an assemblage of spherical grains if grain size follows a log-normal distribution and if the sample has lost most of its $^{40}Ar_r$ in a post-crystallization event (TURNER 1969).

An analogous calculation for sheet geometry (probably more realistic in case of illite and muscovite) instead of spherical geometry does not significantly change the corresponding release patterns. Hence, assuming a simple two-stage model one can estimate the age of the outgassing event or the age of the closure of the argon system after a more extended period of outgassing from the ordinate intercept of a smooth curve fitting the ^{39}Ar-^{40}Ar release pattern. However, such an intercept age may be relatively uncertain since ages at initial temperatures can appear too high due to ^{39}Ar recoil (see sample 1760 and 1703) or too low when a small diffusion loss of $^{40}Ar_r$ occured past the major outgassing. Accounting for these difficulties we assess a relatively high error of ± 3 my to the estimated average age of 11 my for the partial resetting of the argon clock. The clearly noticeable high-temperature drop in the age curve of sample 1844 indicates release of ^{39}Ar recoiled into retentive sites. Also at high temperatures (typically in the interval 950-1100 °C) all illite samples show maxima in their $^{37}Ar/^{39}Ar$ and $^{38}Ar_{Cl}/^{39}Ar$ ratio. The corresponding peak of the muscovite $^{37}Ar/^{39}Ar$ ratio appears above 1200 °C. Obviously the Ca and Cl bearing sites are most retentive for argon release. Such behaviour suggests that Ca may be concentrated in the octahedral domains of illite. At high temperatures above 950 °C we assume that the disruption of the remaining illite structure occurs. The shift towards higher release temperatures for the $2M_1$ muscovite (1703) may be due to the higher degree of crystal lattice order, the higher enthalpy and to the aluminium-rich chemistry of this mica compared with the more disordered structure and the Fe-Mg rich composition of the illite samples.

If we relate the ^{39}Ar-^{40}Ar results to the conventional K-Ar data and to the geological context (Table 2 and Figures 4 and 5) we may conclude that the argon clock of all illites in the lower greenschist facies has been partially reset during Alpine metamorphism 8-14 my ago, whereas a complete reset can be recognized in the higher-grade samples. The observable regional distribution of K-Ar ages (Fig. 4) can be attributed to total outgassing of high-grade samples and to a varying degree of partial $^{40}Ar_r$ losses in samples from the low-grade zone (Ausserberg to Doldenhorn). We believe the higher grade muscovites ($2M_1$) have completely recrystallized during Alpine metamorphism, while the low-grade illites still reflect a pre-Alpine origin of their crystal-lattice, despite the fact that crystallinity as well as mineralogy indicate a formation of these illites during Alpine times. This suggests that K, Al, Mg and Fe were rearranged within preserved crystal lattices of presumably diagenetic origin. In the low-grade zone the kinetics of metamorphic mineral reactions may be too slow, leading to an incomplete «recrystallization» due to a very slow reaction rate.

The present K-Ar and ^{39}Ar-^{40}Ar data of our low-grade illites show that diffusion was an important mechanism for the argon loss: the shape of the release spectra, its relation to metamorphic grade, the grain size dependency as well as the systematic pattern of conventional K-Ar ages supports this interpretation. Assuming a formation of the $1M_d$-illites 180 my ago (diagenetic stage) and a subsequent metamorphic overprint during Alpine orogeny untill 11 my ago, we can calculate the fraction of $^{40}Ar_{radiogenic}$ lost in nature during this event (Table 4). Independently the release of pile-produced $^{39}Ar_K$ up to the arbitrarily chosen temperature of 700 °C during sample extraction (last column of Table 4) characterizes the sample retentivity. If we compare illite samples 1840 and 1846 from the same locality, we can calculate the ratio of average grain sizes $a_{1840}/a_{1846} = 1.6$ assuming volume diffusion of $^{39}Ar_K$ from sheet-shaped grains. This is in reasonable accord with the grain-size ranges known from sample preparation. The same calculation applied to $^{40}Ar_r$ lost in nature (Tab. 4) yields a ratio

Table 4: Compilation of fractional $^{40}Ar_r$ and $^{39}Ar_K$ losses. The corresponding values for $^{40}Ar_r$ are calculated from the total $^{40}Ar_r/^{39}Ar_K$ ratios (Table 3) assuming an original crystallization age of 180 my and apartial$^{40}Ar_r$-loss-event 11 my ago.

Sample No.	Range of grain size (µm)	Fraction of $^{40}Ar_K$ lost in nature	Fraction of (*) $^{39}Ar_K$ released up to 700 °C.
1844	2-6	0.829	0.541
1840	6-20	0.866	0.436
1846	2-6	0.892	0.672
1760	2-3	0.990	0.627

*This last column gives the fraction of pile-produced $^{39}Ar_K$ released up to 700 °C (including those $^{39}Ar_K$ lost during neutron irradiation) as a measure of argon retentivity.

of 1.06, indicating either that diffusion of ^{40}Ar in nature was essentially independent of grain size or occurs from grains of comparable sizes in both samples. The latter would illustrate the fact that the grainsize distribution obtained by separation procedure may deviate from the original grain size distribution in the rock.

4.4. Geological implications and conclusions

The present study demonstrates that conventional K-Ar data on low-grade illites are not reliable for timing metamorphic events. However, important informations can be deduced from ^{39}Ar-^{40}Ar stepwise heating experiments: in the present case, the average of all ^{39}Ar-^{40}Ar ordinate intercept ages of the low-grade illites is 11 ± 3 my and therefore concordant with the conventional K-Ar ages of samples from the higher grade part of the profile (Brig-Verampio). This means that the K-Ar system of all white K-mica studied here was closed at about the same time, despite the significant change in metamorphic grade along this section (260-300 °C/1-2 kb in the north, 580-600 °C/5-7 kb at Verampio in the south). Our age pattern clearly *postdates* the high temperature conditions of the Alpine metamorphism in this area, which could be dated previously by different authors: Köppel & Grünenfelder (1978) deduced from concordant U-Pb monazite ages of 20-30 my in the Verampio-Domodossola area, that high temperature conditions prevailed until this time. Further, Jäger (1967,1973, 1976) and Hunziker (1969, 1974) have argued that Rb-Sr muscovite ages from the external Pennine areas date the climax of the last phase of Alpine metamorphism with about 35-40 my.

If temperature is the most critical parameter for closing the K-Ar system of our white K-mica, then we would date the time at which the cooling rocks passed this critical temperature range. The K-Ar age distribution (Fig. 4) in our cross-section would indicate a more or less horizontal cooling isotherme cutting the steeply inclined metamorphic isograd-surfaces observable in this area (CHATTERJEE 1961, STRECKEISEN & WENK 1974, FRANK 1979). This would also imply a differential temperature-time regime of Alpine metamorphism, beginning with faster cooling-rates (or uplift) in the SE and a much slower cooling in the NW, the Aarmassif and the Helvetic nappe zone. The steeply dipping isograd-pattern observable today in the Simplonregion may so be considered at least partially as a result of this post-metamorphic updoming of the deepest tectonic units (Verampio). In the low-grade part of our profile (Doldenhorn) maximum temperatures reached during Alpine metamorphism are about 260-300 °C. Our results clearly demonstrate that low grade illites have suffered differential partial argon loss at these low temperatures until 11 my, leading to mixed ages. With increasing metamorphic grade complete convertion of $1M_d$ illite to $2M_1$ white K-mica indicates the beginning of total outgassing of all preexistent radiogenic argon.

If we compare our results to Rb-Sr and K-Ar mica ages from the polymetamorphic basement rocks, reported so far in this profile section (JÄGER 1967, 1973, 1976, HUNZIKER 1969, PURDY & JÄGER 1976), the following relations can be found:

- In the Brig-Verampio area, our results are consistent with the Rb-Sr biotite age pattern, showing throughout uniform young ages of 9-13 my. From this simple pattern, cutting zones of Alpine metamorphism and tectonic lines JÄGER (1967, 1973) concluded that these ages reflect cooling ages indicating the time when the rocks cooled to a temperature where the loss of radiogenic strontium ceased. The model of JÄGER postulated very similar «closing temperatures» for retention of radiogenic strontium in biotite (300 ± 50 °C) and for retention of radiogenic argon in muscovite. Our results clearly confirm this assumption.
- Coarse grained muscovite from Alpine fissures in the Simplontunnel give identical K-Ar ages of 9-10 my (PURDY & STALDER 1973). K-Ar muscovite ages of 12-14 my are reported from granitic basement rocks in the Verampio area (PURDY & JÄGER 1976), which are identical to our age results from the cover-rocks. However, much higher apparent muscovite K-Ar ages (phengite-rich muscovites), ranging up to 21 my, are reported from the frontal part of the Pennine nappes in basement-rocks. This discrepancy to our results is not clearly understood at the moment. In principle several factors may be responsible for it:

  a) Higher ages may be due to the presence of excess argon, or these micas were apparently not completely reset during the last metamorphic event, preserving still some pre-existent radiogenic argon. This has recently been found in alpine phengites from the Suretta nappe , where phengites always gives higher K-Ar than Rb-Sr ages (STEINITZ & JÄGER pers. comm.).

b) Apparent ages may have been governed to some extent by the grade of recrystallization: the degree of Alpine recrystallization can strongly depend on the resistance of the rock, leading to different mica-ages in different rock-types within the same locallity as was shown by ARNOLD & JÄGER 1965.

Deformation induced recrystallization may have still continued in phyllitic rocks and may have ceased much earlier in more resistant rock-systems as granitic gneisses. Furthermore the presence of metamorphic fluids in the phyllites may have strongly favoured mineral-reactions by increasing the reaction rates. We will examine these possibilities by further ^{39}Ar-^{40}Ar analyses.

CONCLUSIONS

From the K-Ar and ^{39}Ar-^{40}Ar systematics of white K-mica as deduced in this study by following one single type of rock, we can recognize two important mechanisms of mineral isotope system behaviour:

- in low grade terranes the kinetics of opening and partial resetting of the K-Ar isotope system of illite, mainly controlled by the replacement reaction of $1M_d$ to $2M_1$ illite (continous reaction) and by diffusion processes, both obviously dependent on grain size.
- in the higher grade samples the mechanism of closing of the isotope system (argon-retention ages, obviously independent of grain size). From the mineralogical data it can be deduced that interlayer cations (K-Na) in muscovite exchanged at high temperature conditions near the thermal peak of Lepontine metamorphism (~30 my) whereas argon-retention ages record some later stage at about 10 my during cooling.

The interference of both mechanisms leads to the regional age pattern found in our metamorphic profile. From this pattern we conclude that in the case studied here complete recrystallization of white K-mica occurs at higher temperatures than those critical for argon retention. That means that conventional K-Ar ages do not date any geological event when applied to illite in low grade metamorphic zones.

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