

Fluid circulation, progressive deformation and mass-transfer processes in the upper crust: the example of basement–cover relationships in the External Crystalline Massifs, Switzerland

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(Received 8 September 1991; accepted in revised form 22 March 1992)

Abstract—Basement and cover rocks of the external zone in the Swiss Alps are affected by Tertiary ductile deformation under greenschist facies conditions (300–450°C; 3–4.5 kbar). The analyses of stable isotope systematics of veins and shear zones, as well as variation profiles of chemical elements across major shear zones, distinguish two types of fluid–rock interactions. (i) Closed systems: most syntectonic veins within the Helvetic carbonate cover have $\delta^{18}\text{O}$ compositions depending on the adjacent wall rock compositions and varying with respect to the initial chemical heterogeneity of each sedimentary layer. Within the granitic basement, chemical profiles across minor granite shear zones show equal volume gains and losses for each oxide Na_2O or K_2O . (ii) Open systems: variation profiles in major cover thrusts show a variable increase of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios combined with a strong decrease in $\delta^{18}\text{O}$ approaching an isotopic equilibrium with the basement rocks. Within the major basement shear zones, decrease in CaO and increase in MgO content are observed with progressive deformation increase. A two-step tectonic and geochemical model is proposed to explain the coexistence of open and closed systems. Fluid sources and transport mechanisms of chemical elements are discussed.

INTRODUCTION

FLUIDS are recognized to play a major role in many geological processes (e.g. Fyfe *et al.* 1978, Oliver 1986). There is an overwhelming number of recent publications dealing with the theoretical aspects of fluid flow, fluid sources, fluid–rock interaction and the role of fluids on the rheology of rocks (Bickle & McKenzie 1987, Rumble 1989, McCaig *et al.* 1990, Ferry & Dipple 1991). Regional studies at various scales have documented the presence of fluids during deformation, and their importance as a medium for mass transfer and the alteration of rocks. In granites, for example, retrograde metamorphic reactions and chemical mass transfer often accompany shear zone development and are strong evidence for fluid flow channelled in these narrow deformation zones (Drury 1974, Beach 1976, Kerrich *et al.* 1977, 1980, 1984, Roy 1977, Bossière 1980, Williams & Dixon 1982, Dixon & Williams 1983, Etheridge *et al.* 1983, McCaig 1984, Kerrich & Hyndman 1986, Sinha *et al.* 1986). In some situations, shear zones show isochemical behaviour (Kerrich *et al.* 1980, Marquer 1989). This varying behaviour of shear zones is interpreted as a selective chemical disequilibrium generated by local mechanical instabilities in which new metamorphic conditions allow the crystallization of mineralogical assemblages different from the pre-existing ones. The type of behaviour of mobile and immobile elements is controlled by the types of metamorphic reactions involved within shear zones (Marquer 1989).

The most commonly used tools to study fluid–rock interactions are chemical analyses of major and trace elements, as well as stable and radiogenic isotope systematics (Etheridge *et al.* 1984, Kerrich *et al.* 1984, McCaig 1984, Winsor 1984, Brodie & Rutter 1985, Knipe & Wintsch 1985, Sturchio & Mühlenbachs 1985,

Kerrich & Hyndman 1986, Reynold & Lister 1987). These methods are limited in their application and either method usually infers that a certain minimum amount of externally derived fluid, of an assumed chemical composition, is necessary to explain the observed chemical transformations of the rocks investigated. Only theoretical considerations, however, allow one to constrain the precise fluid source, fluid flow direction and regime (Ferry & Dipple 1991).

Here, we present a synthesis of combined structural and geochemical studies which focuses on the fluid regime during Tertiary deformation of both basement and cover units from the external Swiss Alps (Kerrich *et al.* 1978, 1984, Hoernes & Friedrichsen 1980, Dietrich *et al.* 1983, Marquer 1987, Burkhard & Kerrich 1988, 1990). The study of basement–cover relationships is promising because (i) there is a strong chemical contrast between the two domains, and (ii) major thrusts in the cover are connected to shear zones in the basement. The aim of this paper is to incorporate previously published data (Marquer *et al.* 1985, Burkhard & Kerrich 1988, 1990, Fourcade *et al.* 1989, Marquer 1989) in a general fluid circulation framework during the deformation of the External Crystalline Massifs and associated carbonate cover. Different potential fluid sources are critically reviewed and discussed to explain the chemical and isotopic changes observed within deformed rocks. In particular, we try to investigate fluid regimes associated with chemical and mechanical instabilities at a crustal scale.

GEOLOGICAL SETTING

The central Alps of Switzerland offer insight into more than 20 km of cross-section of the upper crust

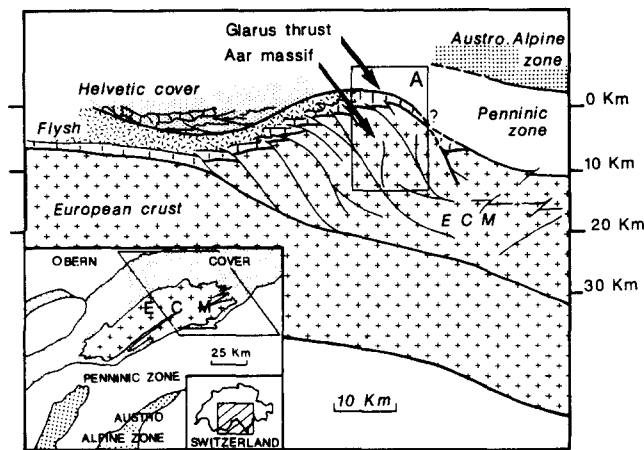


Fig. 1. General cross-section of the northern part of the Alps in eastern Switzerland before late Tertiary uplift (modified after Pfiffner *et al.* 1990a). E.C.M.: External Crystalline Massifs (Aar Tavetsch and Gotthard). Black frame A is shown in Fig. 2.

involved in the Alpine continent–continent collision (e.g. Pfiffner *et al.* 1990a). Structural relationships between basement and cover are well exposed (Fig. 1) (Rohr 1926, Kammer 1989, Pfiffner *et al.* 1990b). The external zone of the Alps is composed of a relatively thin (1–3 km) sedimentary cover overlying the so-called External Crystalline Massifs (ECM: Aar, Tavetsch and Gotthard). This basement is mainly composed of old crystalline rocks and of late Variscan granites (Jäger & Niggli 1964, Wüthrich 1965, Arnold 1970, Labhart 1977, Schaltegger 1989). During the Tertiary Alpine collision, this basement has been affected by heterogeneous ductile deformation under greenschist facies conditions (Steck 1966, 1968, 1984, Choukroune & Gapais 1983, Marquer & Gapais 1985, Marquer 1987, 1990). The ECM are overlain by an incomplete series of (para-) autochthonous cover rocks of Permian to Tertiary age. A stack of Helvetic cover nappes has been thrust to the northwest from more internal positions and lies now structurally above and in front of the ECM. Structural relationships are well exposed due to the plunge of large-scale folds at both lateral extremities of the Aar Massif culmination. The situation of the Glarus nappe cross-section in Eastern Switzerland is sketched on Fig. 1. The Glarus thrust is a well-exposed ductile thrust, where more than 35 km of thrust translation is apparently accommodated by a metre-thick calc-mylonite layer, the so-called ‘Lochseitenkalk’ (Schmid 1975).

An estimate of the metamorphic conditions prevailing during the main deformation studied in the Aar granites corresponds to the upper greenschist facies grade, with progressive increasing of pressure and temperature from the north (400°C, 3 kbar) to the south (450–500°C, 3.5–4 kbar) (Steck & Burri 1971, Frey *et al.* 1974, 1980, Steck 1976, Bernotat & Bambauer 1980, Hoernes & Friedrichsen 1980, Bambauer & Bernotat 1982, Fourcade *et al.* 1989). Metamorphic conditions during thrusting of the Glarus nappe are lower greenschist facies in the southern part of the thrust and in the hangingwall Verrucano, and ‘Anchizone’ determined from northern localities and Tertiary flysch in the footwall (e.g. Frey

1988). Stable isotope thermometry yields values from 350 to 200°C, which are interpreted as decreasing temperatures during progressive thrusting deformation (Burkhard *et al.* 1991).

STRUCTURAL SETTING

In the sedimentary rocks, two types of structures are of interest for the study of fluid–rock interaction (Fig. 2): (i) veins and stylolites which are clear evidence for the presence of fluids during deformation; and (ii) thrust faults as potential conduits for channelled fluid flow. Within the Helvetic nappes, the structural style is strongly dependent on the metamorphic grade (e.g. Ramsay & Huber 1983, 1987, Burkhard 1986, 1990, Dietrich 1990). Northern and higher Helvetic nappes were deformed under diagenetic to anchizone conditions (around 200°C). These units are characterized by pressure-solution cleavage, brittle fractures and abundant veins. Major thrusts, usually following shaly horizons, are extremely narrow deformation zones which show a combination of brittle fracturing, dissolution–crystallization and ductile deformation mechanisms. Southern and lower Helvetic nappes were deformed under epizonal conditions (around 300°C). They show intense, ductile, intracrystalline deformation leading to well-developed schistosity and lineations. The vein formation in these units is restricted to competent layers or can be attributed to early, pre-peak-metamorphic or late-tectonic deformation phases. Extreme strain localization occurs along major thrust planes with predominantly intracrystalline deformation mechanisms and syntectonic recrystallization. A particular example of very asymmetric strain distribution occurs in the Western Helvetic Morcles and Doldenhorn nappes. A highly attenuated inverted limb, decreasing in thickness from the northwest to the southeast to less than 100 m, accommodates some 10–15 km of thrusting distance, whereas the underlying footwall seems to be virtually undeformed (Ramsay & Huber 1983, Dietrich & Dur-

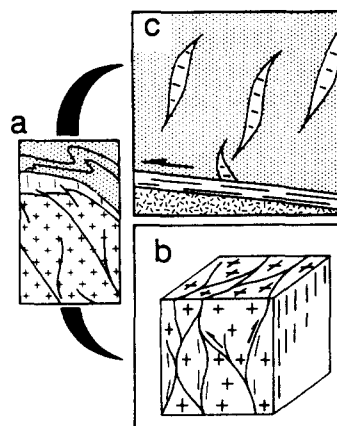


Fig. 2. Sketch of major structures analysed to trace chemical mass transfers and fluid circulations in the crystalline basement and the sedimentary cover. (a) Black frame located in Fig. 1. (b) Shear zones in the crystalline Aar Massif basement. (c) Veins and thrusts in Helvetic cover rocks, i.e. above the Glarus thrust.

ney 1986). Stratigraphic evidence indicates minimum thrusting distances which are typically around 10 km for the major Helvetic nappes but in excess of 35 km in the case of the Glarus thrust.

In the basement and particularly in the granite rocks, the Alpine deformation leads to the formation of ductile shear zones which show anastomosed patterns at different scales (Fig. 2b). In the Aar granites, this deformation is very heterogeneous, with ductile shear zones surrounding lenses of weakly deformed rocks (Choukroune & Gapais 1983, Gapais *et al.* 1987, Marquer 1987). The geometry of structures and the evolution of microstructures across the shear zones are independent of the shear zone size (Marquer 1989). Both grain size and mineralogy change systematically and progressively across ductile shear zones, leading to syntectonic mineralogical assemblages in equilibrium with synkinematic metamorphic conditions and with a grain size of about 30–100 μm in ultramylonites (Marquer 1987). Two shear zones were chosen in the Grimsel granodiorite (north of Grimsel Pass) to analyse the chemical behaviour of stable isotopes, and major and trace elements (Marquer *et al.* 1985, Fourcade *et al.* 1989): the width of the two shear zones is about 3 and 80 m. In some samples, the presence of deformed mafic inclusions allows an estimate of finite strain intensity from the geometry of these xenoliths. A correlation is found between strain intensity and abundance of fine-grained recrystallized material (matrix) which developed in the rocks as a consequence of deformation (see Marquer *et al.* 1985). Thus, the amount of matrix measured in inclusion-free rocks has been used as a quantitative estimate of finite strain intensity in each shear zone in order to compare chemical modifications along concentration profiles related to strain intensity (Marquer *et al.* 1985, Fourcade *et al.* 1989).

GEOCHEMICAL DATA

Basement

With increasing strain intensity, the Grimsel granodiorite changes progressively to an albite-bearing mylonite. The progressive mineralogical and chemical transformations are described as follows (Marquer *et al.* 1985).

(i) From weakly deformed rocks (50% oligoclase, 15% K-feldspar, 20% quartz, 13% biotite) to orthogneiss, the initial magmatic feldspars are replaced by albite and epidote metamorphic minerals. At higher strains, the newly recrystallized albite appears to be unstable. From orthogneiss to ultramylonites, all the feldspathic phases (clasts and blasts) partly disappear and are progressively replaced by phengite and quartz. Epidote is not stable within the ultramylonites (28% albite, 30% phengite, 27% quartz, 13% biotite).

(ii) The associated chemical variations differ from those of magmatic trends (Marquer 1989). Comparison between weakly deformed rocks and ultramylonites show that the immobility of some elements (Si, Al, Fe,

Ti, P, V, Zr, Y) is compatible with isovolumetric transfer (Fig. 3a).

(iii) Plots of concentration strain intensity reveal two types of behaviour of mobile elements (Ca, Mg, Na, K) with increasing strain (Fig. 3b): Ca and Mg decrease and increase, respectively, as a monotonous function whereas K and Na show antithetic variations in such profiles. For example, the Na content increases from weakly deformed rocks to orthogneiss, corresponding to syntectonic crystallization of albite. The Na content decreases in the ultramylonitic stages corresponding to the destabilization of the feldspathic phases. The chemical tendency is opposite for K (Marquer 1989).

(iv) Stable isotopes $\delta^{18}\text{O}$ display similar variations as Ca and Mg profiles (Fourcade *et al.* 1989) and show positive correlation with respect to MgO and negative correlation with respect to CaO. In other terms, mylonitic and ultramylonitic rocks are significantly ^{18}O enriched. Most of the weakly deformed rocks have an isotopic ratio around 7.5‰ which may represent initial magmatic ratios (Fig. 3c). With increasing strain, the isotopic composition increases up to values of 9.65 and 8.65‰ in the ultramylonites of the large and small shear zone, respectively (Fig. 3c).

Changes in mobile elements and stable isotopes become dramatic when finite strain intensity reaches a critical value $\bar{\epsilon}_s$ of about 2 (Marquer *et al.* 1985) (Figs. 3b & c). This chemical boundary can be related to the structural and microstructural evolution observed across the shear zones: a high degree of connection between shear bands is also reached around $\bar{\epsilon}_s=2$ (Fig. 4) (Marquer 1989).

Cover

The Helvetic cover consists largely of carbonates (limestones and marls). Geochemical studies (Kerrich *et al.* 1978, Dietrich *et al.* 1983, Burkhard & Kerrich 1988, 1990) have focused on variations in the stable isotopic composition of these limestones and marls, of secondary calcite precipitated within different generations of veins and pressure shadows, as well as of calc-mylonites from major thrust contacts. Veins are a widespread feature within all Helvetic nappes. Any vein can be easily classified as pre-, syn- or post-tectonic from its orientation with respect to the local structural frame (schistosity, bedding, fold axis) and from the degree of subsequent deformation. Veins are quite important deformation features and represent locally up to 15% of the rock volume. This attests to the presence of a fluid phase during deformation in order to allow the redistribution of ionic species through dissolution-crystallization processes. Most veins display very close isotopic compliance with the surrounding wall rocks (Fig. 5). The local variations of initial isotopic compositions compiled in a schematic stratigraphic profile, show that ^{18}O -rich pure limestones are interlayered with relatively ^{18}O - and ^{13}C -depleted marls with total variations of up to 5‰ in $\delta^{18}\text{O}$ and 3‰ in $\delta^{13}\text{C}$. Despite this heterogeneity, a majority of individual vein to wall-rock

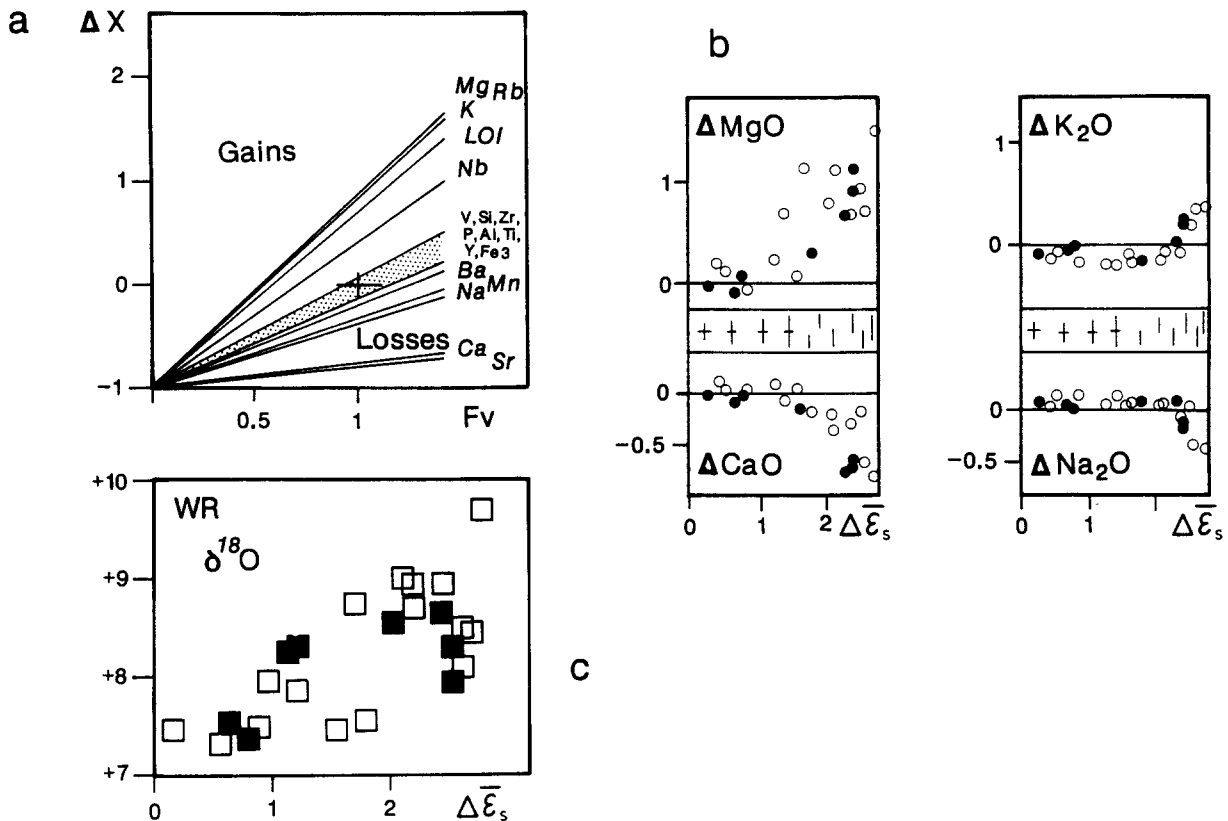


Fig. 3. Geochemical and isotopic variations associated with deformation in shear zones of the Grimsel granodiorite. (a) Volume-composition relationships: graphical resolution of mass balance equation for the comparison of weakly deformed granodiorite and ultramylonitic rock. For each oxide n , the transition between the two states of deformation is expressed by: $\Delta X = Fv \cdot (d_{11}/d_1) \cdot (C_n^{II}/C_n^I) - 1$, where ΔX is the relative gain or loss of mass, C_n^I and C_n^{II} are the weight per cent of the oxide in the initial and the modified rocks, respectively, d_1 and d_{11} the density of these rocks (equivalent in the present case) and Fv volume factor given by the volume ratio between the transformed rock and initial one (Potdevin & Marquer 1987). Dotted area refers to the magmatic fluctuation ranges (immobile elements) and solid lines to mobile elements. (b) Variation profiles of mobile elements vs finite strain intensity for two shear zones (open circles: large (80 m width) shear zone; black dots: small shear zone (3 m width)). Chemical variations (Δ oxide) are normalized to initial concentrations of each oxide in the weakly deformed rock (reference rock: point (0,0)). $\Delta \bar{\epsilon}_s$ is the deviation of finite strain intensity with respect to the reference rock. $\bar{\epsilon}_s = 1/\sqrt{3} (\log a^2 + \log b^2 + \log ab^2)^{1/2}$ (Nadai 1963) where a^2 and b^2 are λ_1/λ_2 and λ_2/λ_3 , respectively ($\lambda_1 > \lambda_2 > \lambda_3$, principal quadratic stretches). Symbol size is greater than analytic errors (data from Marquer *et al.* 1985). (c) Whole rock $\delta^{18}O$ composition vs relative finite strain intensity $\Delta \bar{\epsilon}_s$. Open squares: large shear zone; black squares: small shear zone (data from Fourcade *et al.* 1989).

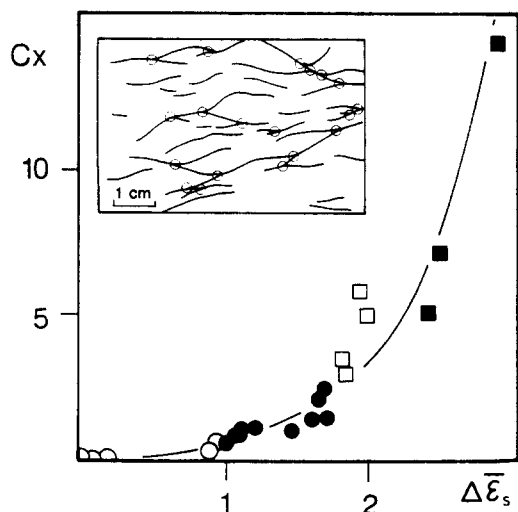


Fig. 4. Degree of shear band interconnection for variously deformed samples from the Grimsel granodiorite (Cx = number of intersections per cm^2) vs relative finite strain intensity $\Delta \bar{\epsilon}_s$. The window shows an example of intersecting shear bands (open circles) in a sample of Grimsel granodiorite orthogneiss. Open circles: isotropic granodiorite; black circles: orthogneiss; white squares: mylonites; black squares: ultramylonites.

pairs show discrepancies of less than 1‰ in both $\delta^{18}O$ and $\delta^{13}C$. This suggests a closed system behaviour (Burkhard & Kerrich 1988, compare with Dietrich *et al.* 1983). Any important fluid circulation across the stratigraphic pile would have certainly led to larger discrepancies between compositions of veins and wall rocks. To be rigorous, the isotopic data do not permit us to rule out (nor to assume) channelled, layer-parallel fluid flow, where the fluid would have been in isotopic equilibrium with each individual stratigraphic layer but not with its neighbour. The extent to which such fluid flow could have occurred is limited, however, because the carbonate-rich Helvetic nappes are isotopically very distinct from any other tectonic unit in the Alps. Carbonate rocks, with a mean $\delta^{18}O$ composition of 26‰ are very sensitive to exchange with any externally derived fluid. All major crystalline units within the Alps have average crustal $\delta^{18}O$ compositions of around 8–10‰ (Frey & Hoefs 1976, Hoernes & Friedrichsen 1980, Fourcade *et al.* 1989).

Clear evidence for open system fluid advection is found in some calc-mylonites from major thrust contacts

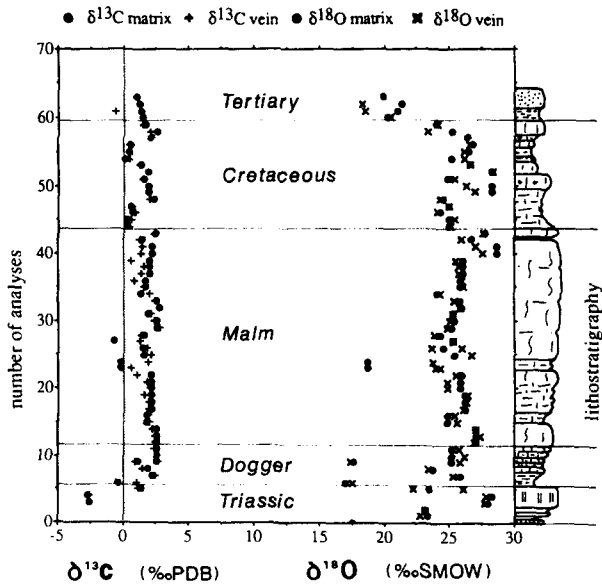


Fig. 5. Stable isotopic carbon and oxygen compositions of syntectonic veins and their adjacent wall-rocks for a series of vein–matrix pairs compiled from the different Helvetic nappes (data from Burkhard & Kerrich *et al.* 1978, Dietrich *et al.* 1983, Burkhard & Kerrich 1988). Data are plotted in a schematic lithostratigraphic profile (the vertical scale is given by the number of isotopic analyses and does not reflect the stratigraphic thickness). Error bars of individual measurements are $\pm 0.2\%$; i.e. smaller than the symbols drawn.

(Doldenhorn and Glarus thrusts; Burkhard & Kerrich 1988, 1990). Calc-mylonites from the Glarus thrust are systematically depleted in both ^{18}O and ^{13}C with respect to precursor marine carbonates (compare $\delta^{18}\text{O}$ in Figs. 5 and 6b). These mylonites show also variable $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (Fig. 6b). However, except for slight enrichments in Sr, S and As (Burkhard & Kerrich 1990), no geochemical anomaly in major or trace element compo-

sitions with respect to marine carbonates could be detected. Stable isotopic data allow us to calculate minimum molar water/rock ratios varying between 3 and up to 30 and more, which is strong evidence for fluid circulation in an open system.

DISCUSSION

Fluid sources

Three main potential fluid sources have to be considered: (1) formation water present in fractures and pores prior to Alpine tectonic activity; (2) dehydration fluids released from rocks undergoing prograde metamorphism; and (3) direct input of surface-derived, meteoric waters.

(1) *Formation waters.* Both the crystalline basement and overlying cover rocks of the present-day Alpine foreland can be regarded as close analogues of the Aar Massif and Helvetic nappes, respectively, prior to their involvement in Alpine tectonics. From thorough hydrogeological studies of both basement and cover rocks in northern Switzerland by NAGRA (Swiss nuclear waste disposal project), there is now a wealth of hydrogeological and geochemical data available for this area (e.g. Matter *et al.* 1988, Pearson *et al.* 1991). Total porosities, i.e. the total amount of stored water in basement rocks, vary from 0.2 to 8% according to the degree of fracturing and hydrothermal alteration of the rocks. From these NAGRA studies, it seems that the top (1–2 km) of the crystalline basement has been considerably fractured and hydrothermally altered during Permian time. Total average open porosities of

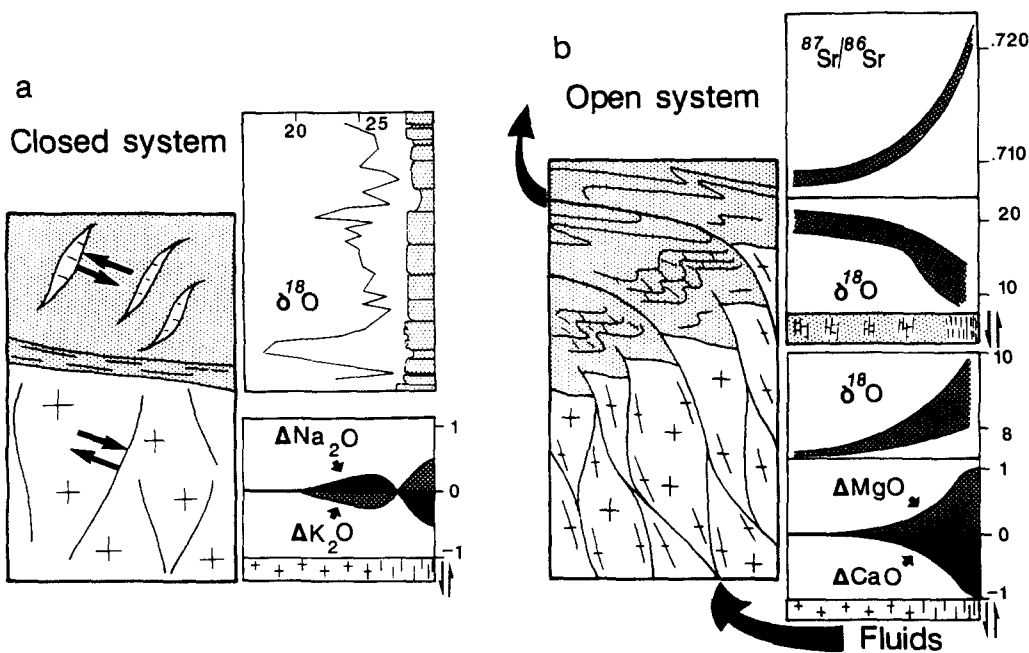


Fig. 6. Sketch of chemical mass transfer evolution, fluid circulation, progressive deformation and basement–cover relationships in the upper crust. Summary of chemical data used are shown on the left-side windows. (a) Initiation of spatially separated mechanical instabilities in basement and cover. (b) Propagation of deformation and creation of an anastomosed network of discontinuities allowing for channelled fluid flow between basement and cover.

2.4% are reported for 'fresh' biotite granites from the Schafisheim drill hole, whereas cataclastic granites have up to 5% open porosity (Matter *et al.* 1988). These average values are comparable with the porosities encountered in lithified, unmetamorphosed Mesozoic sediments dominated by micritic limestones, marls and minor shales (e.g. Matter *et al.* 1988). Potentially more porous sandstones (up to 20% absolute porosity) are scarce within the typical Helvetic series and are found within the (mainly north Helvetic) Tertiary flysch sequence only.

In conclusion, overall porosities of 1–5% were certainly present in both crystalline basement and Mesozoic cover rocks after compaction and lithification but prior to their involvement in Alpine deformation. During deformation (additional compaction induced by burial below higher advancing nappes; Oliver 1986) and greenschist facies metamorphism (recrystallization), a reduction of these porosities to less than 1% (as measured in rocks from the Helvetic region: e.g. Durney 1972, Bernauer & Geiger 1986) can be expected. The amount of fluids thereby released can thus be estimated at 1–4 vol.%. Assuming that all these fluids escaped upwards, the integrated total fluid flux from a typical north Helvetic 5 km thick pile of rocks (2 km of basement plus 3 km of Mesozoic cover) would be of 50–200 tons of fluid per m² section. Accordingly, volumetric water/rock ratios as high as 200 could be recorded by an appropriate 1 m thick horizon located at the top of the pile. A critical question associated with this fluid source is the timing of the main fluid release. Depending on the size and the degree of interconnection of the fracture porosity, a large amount of the formation water may be lost during early stages of compressional deformation, at still relatively low temperatures. Therefore, this may have left no easily detectable signs within the rocks involved.

Apart from the topmost few km of crust, where the presence of formation waters is well documented, small amounts of a free fluid phase might be present throughout the entire thickness of the continental European crust. Several independent lines of evidence for this exist. (i) At the Kola drill hole, formation fluids were encountered at depth below 10 km within old, stable continental crust. (ii) Earthquakes in the northern Alpine foreland of Switzerland occur down to depths of about 30 km within an estimated temperature range where normally no brittle failure should occur (Deichmann *in press*). The presence of fluids is postulated by Deichmann (*in press*) to explain this anomaly. (iii) Mid-crustal reflectors seem to be a general feature of continental crust as revealed by many deep seismic reflection profiles including the Alps (Pfiffner *et al.* 1990b). Among other hypotheses to explain this reflectivity, the presence of fluids has been proposed (Hall 1986). During the involvement in tectonic activity, such as downward flexure below the advancing orogen or simply due to changes in stress state (e.g. an increase in horizontal stress during Alpine collision), these deep-seated formation fluids may be rendered unstable and

expelled in a general upward direction. Thereby, these fluids can interact with rocks along their passage through the crust.

(2) *Dehydration reactions.* Prograde metamorphic reactions, involving sheet silicate minerals are a great potential for the release of 'metamorphic water' (Fyfe *et al.* 1978, Connolly & Thompson 1989). In the context of the external Alps, probably the most important amount of fluid release is related to the successive transformation of smectite to illite and muscovite. Smectite and illite are present in small amounts (1–10%) throughout the Mesozoic stratigraphic pile (e.g. Persoz 1982). More important quantities are found only in relatively thin and scarce shale horizons (Triassic Quartenschiefer, Aalenian black shale, Tertiary Globigerina shale) and Tertiary flysch series. Clay minerals (sericite, illite, smectites, etc.) are also present in basement rocks, due to Permian hydrothermal alteration (e.g. Peters 1987). Given the small quantities of hydrated clay minerals present in both sediments (from 1 to <10%) and basement (<1%), and assuming an average H₂O content of 4 wt% for these minerals (Fyfe *et al.* 1978), the overall fluid production potential from dehydration reactions can be estimated at <0.04 to <0.4 wt%. Assuming again an upward escape, the integrated total fluid flux from a 5 km thick rock pile of rocks would be between 2 and 20 tons of water per m², clearly an order of magnitude smaller than for that of formation waters discussed above. Such dehydration waters are expected to be released during prograde metamorphism from 100°C onward where smectites are destabilized (e.g. Hower *et al.* 1976). However, this holds true only for relatively porous series (molasse, flysch), whereas the disappearance of smectites within limestones, marls and shales seems to be considerably retarded (Persoz 1982). Some smectites are still present even within epizonal Malm limestones (Huon *et al.* 1988). The transformation from illite to muscovite is progressive too and seems to take place during the diagenesis and anchizone (e.g. Hunziker *et al.* 1986). In conclusion, moderate amounts of fluids from dehydration reactions (<<1 vol.%) may be produced from the Helvetic sedimentary pile and underlying altered crystalline basement from *ca* 100°C onward, to be completed only during greenschist metamorphism, at more than 300°C.

Lower parts of the northern Alpine chain may have been buried deep enough to undergo higher grade metamorphic reactions (Connolly & Thompson 1989). In contrast to the wet metapelitic model crust considered by Connolly & Thompson (1989) however, in this part of the Alps, only crystalline units without any significant amount of sedimentary cover are buried deep enough to reach amphibolite grade metamorphic conditions. These basement rocks have either already undergone a cycle of Hercynian high-grade metamorphism or are late Hercynian intrusives and thus the pristine basement is essentially 'dry' and would not produce any significant amount of metamorphic fluid

from dehydration reactions unless heated to granulite facies conditions. It is questionable whether parts of the lower European crust (deep below the Aar Massif) underwent granulite facies metamorphism; one would rather expect these sections of crust to have experienced high-pressure, eclogite facies conditions (e.g. Laubscher 1990). This argument can even be reversed: the pristine Hercynian basement, when undergoing Alpine deformation and greenschist facies metamorphism, is expected to undergo fluid consuming, retrograde mineral reactions rather than liberate any fluids.

(3) *Surface water.* During the formation of the present-day high topographic relief and favoured by the establishment of a longitudinal extensional regime with deep reaching brittle fractures within parts of the chain during Miocene times (e.g. Mancktelow 1985, Ratschbacher *et al.* 1989), deep infiltration of meteoric water has to be expected down to depths of several kilometres. More or less surficial circulation systems are clearly active today, as expressed by numerous thermal springs within the Alps and its foreland (e.g. Arthaud & Dazy 1989, Minissale 1991). Geochemical evidence for deep paleo-fluid circulation is present in the late-tectonic tensile veins where calcite precipitated most probably from surface-derived and extremely ^{18}O -depleted meteoric waters. These must have penetrated down to more than 5 km depth during uplift of the External Crystalline Massifs and overlying Helvetic nappes (Burkhard & Kerrich 1988).

Transport mechanism

Many authors (Fletcher & Hoffmann 1974, Bickle & McKenzie 1987, Ferry & Dipple 1991) have discussed different models of fluid flow and transfer mechanisms in which physico-chemical parameters such as diffusion and permeability coefficients, temperature, activity of chemical species, load and stress gradients, but also the size of the closed system and the width of channelized fluid pathways are strongly influential (Gratier 1984). In recent works (Baumgartner & Rumble 1988, Rumble 1989, McCaig & Knipe 1990, Ferry & Dipple 1991), two basically different transport processes have been invoked to explain intergranular mass transfer assisted by a fluid medium (Fletcher & Hoffmann 1974): (i) ionic diffusion in a stationary fluid phase, so called diffusion: or (ii) element transport by fluid infiltration (e.g. convection or single path) through a porous solid medium, so called infiltration. As a summary of these theoretical investigations and in agreement with many authors (Korschinskii 1970, Fletcher & Hoffmann 1974, Frantz & Weisbrod 1974, Beach 1976, Fonteilles 1978, Fyfe *et al.* 1978, Etheridge *et al.* 1984, Gratier 1984, Fyfe & Kerrich 1985, Wood & Graham 1986, Bickle & McKenzie 1987, Rumble 1989, Ferry & Dipple 1991), only infiltration processes seem to be able to generate the observed chemical transformations over large scales (>several metres) and within the disposable geological times of several million years. The co-

existence of diffusion and infiltration processes is emphasized by the results of our investigations on mass transfer at different scales. The presence of a fluid phase during deformation in both basement and cover rocks is obvious from the formation of pressure shadows and veins which apparently formed in closed systems of relatively small dimensions (centimetres to some metres). These phenomena could be explained by either a stationary pore fluid with diffusion processes leading to material redistribution, or by a mechanism of fluid recycling where fluids are alternatively sucked into veins or shear zones and expelled into the surrounding undeformed and porous wall rock, following local pressure gradients in response to tectonic activity, but without leaving the system (Fig. 6a). Veins from the Helvetic nappes typically formed by a crack-seal mechanism (e.g. Durney 1972) are compatible with such a fluid recycling model (see also Mullis 1976).

On the other hand, once the network of deformed zones (shear zones and thrusts) is sufficiently anastomosed, large-scale fluid infiltration can induce chemical mass transfer and isotopic modifications (Fig. 6b). Geochemical reactions between basement and cover rocks can be generated at this stage of progressive deformation with an equilibration of $\delta^{18}\text{O}$ values at around 10‰ for both basement shear zones and major thrusts in sedimentary rocks (Fig. 6b). As suggested by McCaig & Knipe (1990), the chemical mass transfer observed in both basement and cover rocks, could be interpreted as a geological example for diffusion and infiltration processes operating simultaneously at different scales, but also evolving in space according to local heterogeneous deformations and basement-cover relationships. In this interpretation $\delta^{18}\text{O}$ values of veins in cover series or K/Na variation profiles across basement shear zones express the first stages of chemical mass transfer during progressive deformation in a closed system (Fig. 6a).

Fluid circulations during progressive deformation

The foregoing discussion of fluid sources and fluid regimes during deformation is schematically summarized in Fig. 7. The present-day cross-section of the

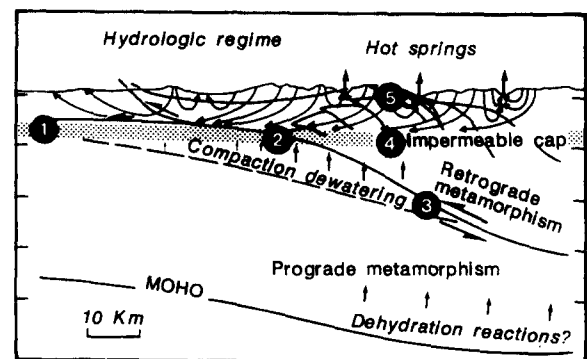


Fig. 7. Sketch of potential fluid sources and fluid regimes at the crustal scale. A present-day cross-section of the eastern Swiss Alps (compare with Fig. 1) is used to discuss the relative position of the Aar Massif in space and time during its tectonic evolution from late Eocene (1) to its present situation (5) (see text for explanation).

external Alps (compare with Fig. 1) is roughly regarded as a typical cross-section for the Oligocene–Miocene evolution of the northern part of the Alps. We further assume that a present-day large basal crustal-scale thrust below the External Crystalline Massifs was the main locus of the last 15 Ma (and maybe still ongoing?) thrust deformations. This thrust separates an upper plate undergoing fluid consuming retrograde metamorphism whereas the lower plate is being buried, compacted and possibly undergoing prograde, fluid releasing metamorphism. During the evolution of the Alpine chain, one or several similar large-scale thrusts must have developed: from top to bottom and succeeding each other in time, the main thrusts are the Austroalpine, the Pennine and the Helvetic (e.g. Glarus) thrusts. The tectonic evolution of the Aar Massif or of the Helvetic nappes above the Glarus thrust can thus be sketched within this single cross-section: numbers (1)–(5) correspond to the approximate position of the Aar Massif through time from Eocene (1) to Oligocene ‘peak’ metamorphism (3) and to its present-day elevated position (5).

(1)–(2): early stages of expulsion of formation fluids due to tectonic burial and compaction are extremely difficult to document. The present-day hydrogeologic regime of the upper few kilometres within the Alps and their foreland is dominated by recent and often quite old (e.g. Pearson *et al.* 1991) meteoric waters circulating in a network of fractures and/or sedimentary aquifers. Any contribution of expelled formation or metamorphic waters from region (2) to (3) into this regime must be diluted beyond recognition (but compare Arthaud & Dazy 1989, Minissale 1989, Pearson *et al.* 1991). The identification of this stage of paleo-circulation by means of geochemical analyses of rocks is extremely difficult (compare Ayalon & Longstaffe 1988) and it has not been documented so far in the Alps. Despite the potentially large volumes of fluids involved in this stage, very little geochemical imprint on the rocks is to be expected because of the prevailing low temperatures. Furthermore, any geochemical signature in the rock would tend to be subsequently obscured by deformation and alteration under greenschist facies metamorphism. However, Dietrich *et al.* (1983) interpreted small disequilibrium in $\delta^{13}\text{C}$ signatures between early veins and wall rocks as evidence for an early stage of open system fluid circulation within the Helvetic cover rocks.

(3)–(4): deeper burial by higher units led to the Oligocene syntectonic metamorphism. During this stage, remaining formation pore-fluids as well as moderate amounts of dehydration water from clay minerals are expected to be chemically re-equilibrated with the surrounding rock masses and are henceforward to be considered as ‘metamorphic fluids’ (Sheppard 1986). Given that formation fluids were in part of *D*-depleted meteoric (rather than marine) origin, and that rocks have very low hydrogen content, δD signatures will not easily change during fluid–rock interaction and may therefore still betray the origin of fluids (e.g. Kerrich 1987). Whatever their ultimate source, these metamorphic fluids play an important role in the formation of

veins and chemical alteration in shear zones. In small-scale structures, with moderate associated deformation, there is no geochemical evidence for larger scale transport of these fluids and solutes. Observed geochemical trends can be explained by local closed systems where diffusion processes in a ‘stationary’ fluid phase and fluid migration on a small scale (<metric) occur. With increasing deformation, fluids were apparently channelled in deformation zones, leading to significant geochemical alteration of the rocks involved. This occurred especially along larger shear zones and major thrust faults. Even though part of the deformation along basement shear zones took place in a prograde regime (increasing metamorphic grade, situation 3 in Fig. 7) this led to seemingly retrograde fluid consuming mineral reactions because the pristine basement are granites and higher grade metamorphic rocks.

Apart from the fact that ^{18}O -depleted and ^{87}Sr -enriched basement-derived fluids are required to explain the isotopic signatures found in some of the largest Helvetic thrusts, the general fluid flow direction remains unknown. Theoretical arguments, rather than geochemical data, suggest that fluid migrations are generally upward and foreland directed (e.g. Oliver 1986, Connolly & Thompson 1989). Indeed: (i) the only potential fluid sources are located within the downgoing plate; (ii) fluids at higher than hydrostatic pressure (as expected during metamorphism and thrusting deformation; Hubbert & Rubey 1959, Etheridge *et al.* 1983, 1984), are gravitationally unstable and tend to migrate upward; (iii) fluid-consuming retrograde reactions take place within the upper and uplifting plate; and (iv) the direct access of meteoric waters down to greater depth (down more than 10 km) is extremely difficult to imagine; it would imply a general flow direction against the pressure gradient, from brittle rocks with larger porosities and permeabilities down into rocks undergoing active metamorphism, recrystallization and thereby losing most of their porosity, hence decreasing their permeability. Note however, that very deep infiltration of surface-derived fluids against the regional, topographically induced pressure gradients has been postulated in a geologic context very similar to that of the northern Alps (McCaig *et al.* 1990, fig. 3, see also Wickham & Taylor 1987). McCaig *et al.* (1990) invoked a large-scale seismic pumping mechanism (strongly modified from Sibson 1990) in order to solve the apparent paradox of fluids flowing against pressure gradients.

(4)–(5): at some stage during uplift, the rocks are uplifted to within the region of brittle deformation with a supposedly hydrostatic fluid pressure regime. There is no direct evidence for the precise depth of this transition zone labelled ‘impermeable cap’ (after Etheridge *et al.* 1983). Thermal springs within the Alps, with temperatures reaching 60°C (e.g. Loèche les Bains) provide evidence for the infiltration of waters to depth of at least 3 km. Much deeper infiltration, however, can be anticipated from the topographic relief of the Alps (Kimmerer *et al.* 1985). So far, very detailed studies of the deep fluid circulation within the crystalline basement of the Alpine

foreland (Matter *et al.* 1986, Pearson *et al.* 1991, p.409ff.) show that very old (>>1000 to <30,000 years) and possibly older meteoric water is present within this basement. The recharge area is thought to be the topographic high of the 'Forêt Noire' to the north. Hydrogeologic and geochemical data, however, do not rule out an Alpine origin (Oxburgh & England 1980, fig. 7), a possibility also supported by hydrogeologic modelling of the northern flank of the Alps (Kimmeier *et al.* 1985).

CONCLUSIONS

During Alpine Tertiary events, strongly deformed rocks of the basement and the cover of the Aar Massif have suffered modifications of their bulk whole-rock chemistry due to fluid-rock interactions. Fluid circulation seems to be localized in narrow pathways rather than to occur as pervasive fluid flux. The initiation of fluid-rock interaction was closely related to local mechanical instability development (such as micro-cracks) which allowed introduction of fluids in these heterogeneously deformed domains. If a chemical instability generated by local metamorphic conditions exists in the basement shear zones or cover thrusts, geochemical and isotopic mass transfer occur. The scale of this chemical instability and of the associated mass transfer is controlled by the geometry of structures in the system. Isolated vein systems in the cover of the Aar Massif are typical examples of small-scale closed systems with small amount and small-scale fluid circulation, whereas anastomosed networks of thrusts and shear zones allow for larger fluid fluxes and mass transfer to occur in open systems at the upper crust scale. In the analysed basement-cover association, the fluids during the main Alpine deformation are thought to be of deep origin (expelled formation fluids and dehydration reactions), the general fluid migration being upward and foreland directed.

Acknowledgements—Financial support by the Fonds National Suisse projects Nos 20-26 313.89 and 20-27597.89 are gratefully acknowledged. We thank our colleagues in the CAESS Rennes and R. Kerrich (Sakatooon) for their logistical support of all aspects of chemical and stable isotope analyses, respectively, and many constructive discussions. F. Persoz and J. P. Schaer provided incisive critiques of a provisional draft and two anonymous reviewers are thanked for their careful revision of this paper.

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