Evolution of fluid compartmentalization in a detachment fold complex

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ABSTRACT

Oxygen, carbon, and strontium isotope variations in vein-filling calcite and quartz cements and their host rocks are used to elucidate the origin, spatial and temporal evolution, and migration pathways of fluids in the detachment Nuncios fold complex, northeastern Mexico. The folded Mesozoic sedimentary sequence contains two regional paleohydrostratigraphic units separated by a unit of low permeability. Two main generations of cements are present in both paleohydrostratigraphic units. Distinct differences exist between δ^{18} O, δ¹³C, ⁸⁷Sr/⁸⁶Sr, and fluid-inclusion temperatures of early vein-filling cements in the lower and the upper units. These differences, together with a strong correspondence between early cement and host-rock δ^{18} O and δ^{13} C values, suggest that early diagenetic fluids were compartmentalized between the two units. Late vein-filling cements have isotopic compositions and fluid-inclusion temperatures that converge to similar values, indicating a change to open fluid flow between the lower and upper units. We hypothesize that the fluid history of the Nuncios fold complex evolved in two main stages: (1) burial diagenesis and early folding, during which fluids were confined within individual units, and (2) latestage folding, during which increased deformation associated with fold tightening caused the expulsion of fluid from the lower unit into the upper unit.

Keywords: fluid migration, evaporite, isotopes, detachment fold, Sierra Madre Oriental.

INTRODUCTION

During the past two decades the importance of regional scale integrated geological and geochemical investigations of the paleohydrology of orogenic regimes has become widely recognized. Such studies can help in understanding the origin, evolution and movement of crustal fluids and their structural. economic, and environmental significance (Cathles, 1990). Despite the widely recognized importance of folds in localizing hydrocarbons and/or ore deposits, the topic of fluid migration during folding has received little attention. Previous studies have focused on foreland fold-thrust belts such as the Appalachians (Evans and Battles, 1999), Ouachita Mountains (Richards et al., 2002), Canadian Rockies (Nesbitt and Muehlenbachs, 1994), Lachlan fold belt (Gray et al., 1991), and Alps (Kirschner et al., 1999).

Detachment folds, a major structural feature in many orogenic belts, form in stratigraphic packages with large thickness and competency contrasts among lithologic units (Mitra, 2003). Because faulting in detachment fold belts is usually minor, particularly in the early stages of deformation, the general hydrostratigraphic structure and synorogenic fluid migration in detachment fold belts can be very different from those described in classic fold-thrust belts.

In this study we present isotope data from veins and host rocks of the detachment Nuncios fold complex, which is the Laramide-age frontal structure of the Monterrey salient of the Sierra Madre Oriental, northeastern Mexico. The stratigraphy of the fold complex is not complicated by significant thrust ramps, duplexes, or faulting (Fischer and Jackson, 1999). The unique geochemical signature of rocks and vein minerals allows us to trace the spatial and temporal evolution of the fluidrock system associated with fold development. Specifically we present evidence that: (1) fluid flow in the fold belt was compartmentalized within individual hydrostratigraphic units during burial diagenesis and early stages of folding; (2) deformation associated with fold

tightening during late stages of folding was accompanied by fold rupture and upward fluid migration from the lower, overpressured unit into the upper unit; (3) oxygen and strontium isotope signatures of vein minerals are powerful tracers of fluids that equilibrated with the basal evaporite.

GEOLOGIC SETTING

The Sierra Madre Oriental, a Laramide-age thin-skinned fold-thrust belt, is located along the eastern edge of northeastern Mexico. In the Monterrey salient, folds are arcuate in plan view with wavelengths from 5 to 8 km and amplitudes from 2 to 3 km. Many can be traced 20-70 km along strike (Fig. 1). The folds are generally symmetric, upright to slightly north-vergent, kink-style box folds. The Nuncios fold complex is the westwardplunging frontal structure of the Monterrey salient that involves a 3000-m-thick sedimentary section detached above the Minas Viejas evaporitic sequence, the initial thickness of which is estimated as 1000 m (Goldhammer et al., 1991). The sedimentary succession consists of Upper Jurassic (Minas Viejas, Zuloaga, and La Casita Formations) and Lower Cretaceous (Taraises, Cupido, La Peña, and Aurora Formations) units. The Upper Cretaceous Parras Formation and Tertiary Difunta Group crop out north of the study area, and are presumed to have once overlain these formations. Gray et al. (2001) suggested that in the Monterrey salient the Upper Cretaceous and Tertiary clastic units were as thick as 2000 m, limiting maximum burial depths of Lower Cretaceous rocks to 7 km.



Figure 1. Schematic regional tectonic map of northeastern Mexico (modified after Fischer and Jackson, 1999).

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SAMPLING AND ANALYTICAL TECHNIQUES

Samples of veins and their sedimentary host rocks were collected from each lithostratigraphic unit throughout the Nuncios fold complex. Vein mineral separation involved handpicking and acid dissolution to remove calcite from quartz samples. Stable isotope ratios were measured at Northern Illinois University on a Finnigan MAT 250 mass spectrometer. Oxygen isotope ratios of 1-2 mg of vein quartz were determined by a laser fluorination technique using BrF₅ as the oxidizing reagent. NBS-28 quartz standards, analyzed at the same time as the samples, yielded $\delta^{18}O_{VSMOW}$ $= 9.59\% \pm 0.15\%$ (*n* = 23; VSMOW—Vienna standard mean ocean water). The stable isotope ratios of carbonate samples were determined by reaction with H₃PO₄ at 25 °C according to the method of McCrea (1950). Isotope data are reported in permil (%) relative to the Vienna Peedee belemnite (VPDB) standard for carbon and VSMOW for oxygen. The reproducibility is estimated as $\pm 0.2\%$ (1 σ) for quartz and $\pm 0.15\%$ (1 σ) for carbonates. Strontium isotope analyses were conducted at the University of Texas at Austin using analytical techniques described by Banner and Kaufman (1994). NBS-987 yielded an 87Sr/ ⁸⁶Sr ratio of 0.710249 \pm 0.000009(1 σ) during the interval when the samples were analyzed. The geochemical results are available in Data Repository Tables DR1 and DR2.1

ISOTOPIC VARIATIONS IN VEINS AND HOST ROCKS

Vein mineral assemblages closely mirror the host-rock lithology throughout the study area. Thus, quartz is a major vein mineral in predominantly siliciclastic units (La Casita, Taraises) and calcite is the dominant vein mineral in carbonate units (Zuloaga, Cupido, Aurora).

In siliciclastic units, the δ^{18} O values of vein quartz display a distinctly bimodal distribution (Fig. 2). Taraises veins are characterized by quartz with high δ^{18} O values (27% ± 0.4%, n = 12), and La Casita veins are characterized by quartz with distinctly low δ^{18} O values (20.2‰ ± 0.5‰, n = 46). Uniform δ^{18} O values of vein quartz occur at all scales, from hand specimen to outcrop. Furthermore, veins from forelimb and backlimb of the fold display similar ranges of δ^{18} O values (cf. Figs. 2B and 2C). For comparison, Gray et al. (1991) and Richards et al. (2002) reported similar restricted ranges of δ^{18} O in quartz



Figure 2. Stable isotope data of quartz and calcite veins from siliciclastic units related to their structural location. Vein minerals from the Taraises Formation (A) display isotopic signatures distinct from those of La Casita Formation (B). Isotopic values of vein minerals within each formation are relatively uniform; similar ranges of values for (B) fold forelimb and (C) backlimb. See Figure 1 for location of cross section in regional setting. (Cross section modified after Fischer et al., 2002.) SL—sea level.

veins of other terrains of low metamorphic grade, independent of their orientation or timing. However, the strongly bimodal δ^{18} O signature of vein quartz (with a difference of ~7‰) in the two adjacent units of the Nuncios fold complex that was observed in samples only meters apart is a distinctive feature that, to our knowledge, has not been encountered elsewhere.

Stable isotope behavior in coexisting quartz and calcite pairs mirrors the bimodal distribution observed in vein quartz; vein calcites of La Casita Formation display homogeneous but lower δ^{18} O and δ^{13} C values compared to those of Taraises vein calcites (Fig. 2). Furthermore, although the δ^{18} O values of vein quartz in the Taraises and La Casita Formations are distinctly different, the isotope fractionation factor between coexisting quartz and calcite, Δ^{18} O[qz-cc], is very similar for both the Taraises (2.06‰ ± 0.2‰) and La Casita Formations (1.94‰ ± 0.3‰).

Oxygen isotope ratios of carbonate host rocks plot in two distinct groups, regardless of their lithologies, with little overlap between them (Fig. 3A). The first group, composed of the upper formations in the stratigraphic succession (Taraises, Cupido, and Aurora), has relatively high δ^{18} O values of 22‰–27‰. The second group, composed of the Zuloaga and

La Casita Formations, has significantly lower δ^{18} O values of 18%-22%. All carbonate rocks have δ^{18} O values lower than their estimated original marine value of $\sim 28\%$ (Veizer et al., 1999). In contrast to δ^{18} O patterns, δ^{13} C values of both groups are broadly similar, the low δ^{18} O group displaying a relatively wider range.

The isotopic composition of vein calcite from upper carbonate units (Cupido, La Peña, and Aurora) displays a unique pattern. Whereas early vein-filling calcites have isotopic compositions similar to their host rock, latestage vein-filling calcites are depleted in ¹⁸O by as much as 8‰ compared to the adjacent host rocks (Fig. 3B). The late-stage veins have variable dimensions (millimeters to decimeters) and highly variable orientation (Fischer and Jackson, 1999). One example of a vein sample from a shear zone in the Cupido Formation is presented in Figure 4. This handsized sample is composed of networks of fractures with three generations of calcite fillings recognized by their texture, color, and isotopic signature, demonstrating repeated episodes of fracturing and fluid flow in the shear zone. The δ^{18} O value of the oldest generation of vein calcite (type 1: 23.9‰) is very similar to that of the adjacent host rock (24.5‰). Later generations of vein-filling calcites have dis-

 $^{^1\}text{GSA}$ Data Repository item 2005009, Table DR1, $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ data from carbonate host rocks and calcite veins of Nuncios fold complex, and Table DR2, oxygen isotope data of vein quartz, is available online at www.geosociety.org/pubs/ft2005.htm, or on request from editing@geosociety.org or Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301-9140, USA.



Figure 3. A: δ^{18} O and δ^{13} C values of carbonate rocks from Nuncios fold complex. Geochemical signatures of host rocks define distinct populations that are correlated with upper and lower paleohydrostratigraphic units (UHU and LHU). B: Whole-rock calcite δ^{18} O. s. vein calcite δ^{18} O. Early-stage veins throughout sequence have δ^{18} O values similar to their host rocks. Late-stage veins from LHU have lower δ^{18} O values compared to their host rock. SMOW—standard mean ocean water; PDB—Peedee belemnite.

tinctly lower δ^{18} O values of 18.6‰ for type 2 and 16.8‰ for type 3 vein fillings. The low δ^{18} O values of late-stage vein fillings are coupled with lower ⁸⁷Sr/⁸⁶Sr ratios (type 1: 0.7079; type 2: 0.7077; type 3: 0.7076). For comparison, ⁸⁷Sr/⁸⁶Sr ratios of vein calcites from the La Casita and Zuloaga units are relatively uniform, with values lower than 0.7073. The ⁸⁷Sr/⁸⁶Sr ratio of Minas Viejas evaporite, the oldest exposed unit in the sequence, is 0.7069.

DISCUSSION

Paleohydrostratigraphic Units

The bimodal distribution of δ^{18} O values in vein quartz (Fig. 2) and host rocks (Fig. 3A) provides evidence for the existence of two geochemically distinct paleohydrostratigraphic units. The lower paleohydrostratigraphic unit is composed of the Zuloaga and La Casita Formations. The unit directly overlies the lower detachment in the Minas Viejas evaporite and underlies the low-permeability rocks of the Taraises Formation. The upper paleohyFigure 4. A: Photograph of vein sample from shear zone. Three generations of calcite vein fillings are distinguished by their texture, color, and isotopic signature. B: Strontium and oxygen isotope values of vein calcites from shear zone. Low δ^{18} O values of late-stage vein fill-



ings (type 2 and 3) are coupled with lower 87Sr/86Sr ratios.

drostratigraphic unit comprises the Taraises, Cupido, La Peña, and Aurora Formations (Fig. 2). The upper paleohydrostratigraphic unit is confined beneath the Parras shale, which also constituted the regional upper detachment in the Monterrey salient (Wilson et al., 1999).

The isotopic pattern of rocks and early veins within each paleohydrostratigraphic unit is continuous across the study area. Their distinct geochemical signatures suggest that during burial diagenesis and early stages of folding, a regionally continuous hydrologic system existed within each paleohydrostratigraphic unit with little or no fluid transfer between the two paleohydrostratigraphic units. The overall compartmentalization of fluids can be related to the particular lithostratigraphic package of the folded sedimentary sequence. Low-permeability units such as the Taraises and Parras Formations acted as aquitards during burial diagenesis and most of the folding processes, allowing the formation of two regional aquifers. The fossil system we infer for the Nuncios fold complex has a close parallel in the stratified fluids of modern and fossil systems seen in the U.S. Gulf Coast (e.g., Hunt, 1996; Stover et al., 2001), Appalachian region (Evans and Battles, 1999), and Alberta Basin (Bachu, 1999).

Origin of Fluids

The oxygen isotope composition of veinforming fluid provides constraints on its source. Meteoric water and water expelled during dehydration reactions of hydrous minerals (clay minerals, opal, or gypsum) are possible sources of vein fluid during diagenesis and low-temperature metamorphism of sedimentary rocks. In the Nuncios fold complex, fluid-inclusion data indicate average temperatures of vein-mineral precipitation of 180 °C for upper and 220 °C for lower paleohydrostratigraphic unit (Fischer et al., 2002). Using the δ^{18} O values of vein quartz, the fluid-inclusion homogenization temperatures, and the quartz-water fractionation equation of Matsuhisa et al. (1979), the calculated average δ^{18} O values of vein-forming fluids are 10‰ for lower and 14‰ for upper paleohydrostratigraphic unit, much greater than would be consistent with a meteoric water source. In sedimentary basins, typical diagenetic processes

result in progressive increases in δ^{18} O values of formation waters with increasing burial depth (Kharaka and Thordsen, 1992). The calculated compositions of the vein-forming fluids are evidence that the vein minerals precipitated from basinal fluids enriched in ¹⁸O relative to Late Jurassic or Cretaceous seawater (~-1‰; Veizer et al., 1999).

In the lower paleohydrostratigraphic unit, the 1000-m-thick Minas Viejas evaporite was a possible source of fluid. During the conversion of gypsum to anhydrite, each cubic meter of gypsum releases 480 kg of water. The actual depth at which this transition occurs can be highly variable and depends on a complex set of local conditions, including geothermal gradient, fluid pressure, lithostatic pressure, activity of water, and heat capacity of overlying lithologies (Hanshaw and Bredehoeft, 1968; Jowett et al., 1993). Water of hydration in gypsum is enriched in ¹⁸O by $\sim 4\%$ compared to water from which it precipitates (Gonfiantini and Fontes, 1963), i.e., evaporated Jurassic seawater. The low δ^{18} O values of carbonate rocks from the Zuloaga carbonate and La Casita siliciclastic formations, depleted in ¹⁸O by 6‰-10‰ relative to marine values (Fig. 3A), imply high water-rock ratios during diagenesis. Furthermore, the similarities in low δ^{18} O values of coexisting veins and host rocks from lower paleohydrostratigraphic unit (Fig. 3B) can be explained by a fluid-dominated system. The 87Sr/ ⁸⁶Sr ratio of Late Jurassic evaporite (0.7069) is near the Phanerozoic minimum for the Sr isotope ratio in seawater (Jones et al., 1994). Because younger formations of the sedimentary sequence will have higher 87Sr/86Sr ratios, Minas Viejas evaporite is a likely source of low radiogenic Sr in the vein-forming fluid. These observations, together with the low $\delta^{18}O$ and low 87Sr/86Sr values of calcite veins from lower paleohydrostratigraphic unit, suggest that dehydration of the basal Minas Viejas evaporite was a possible source of lower paleohydrostratigraphic unit water.

In the upper paleohydrostratigraphic unit, the calculated fluid $\delta^{18}O$ value of 14‰ indicates that Lower Cretaceous carbonates dominated the oxygen isotope composition of the fluid-rock system involved in early vein formation. The low $\delta^{18}O$ signatures of late-stage vein fillings (Fig. 3B) are evidence of open-

system behavior (Gregory and Criss, 1986). To precipitate the low $\delta^{18}O$ calcite from internally derived upper paleohydrostratigraphic unit waters would require unreasonably high temperatures of >350 °C. Thus, an external fluid source with low δ^{18} O and low 87 Sr/ 86 Sr values is required. The overlap in δ^{18} O values of lower paleohydrostratigraphic unit vein calcite and late-stage upper paleohydrostratigraphic unit vein calcite suggests a possible common source of the vein-forming fluids. The δ^{18} O and 87 Sr/ 86 Sr trends, with lower values in late-stage upper paleohydrostratigraphic unit veins, provide further evidence that the origin of fluid in late-stage upper unit veins was the lower paleohydrostratigraphic unit fluid. These fluids were probably expelled upsection during active deformation in the late stages of fold tightening.

Implications for Fluid Flow During Folding

We suggest that the evolution of the foldrelated fluid-rock system can be divided into two main stages. Stage I was characterized by burial diagenesis, compressional tectonics, fracturing of strata, and episodic fluid flow in response to tectonic stresses. During this stage, the Taraises Shale acted as an aquitard between the two main paleohydrostratigraphic units, and vein minerals within each paleohydrostratigraphic unit were distinctive in their O and Sr isotope composition. Fracture permeability within the paleohydrostratigraphic units undergoing compression was, in general, limited and discontinuous, and the fluid regime was isolated with little or no vertical movement. The similar $\delta^{18}O$ signatures of veins and adjacent host rocks suggest that during early folding the fluids were mainly locally controlled within the layered paleohydrostratigraphic units. Stage II was characterized by fold tightening, extensive fracturing, and a major change in the hydrologic system both within each paleohydrostratigraphic unit and overall in the fold complex. Active deformation associated with late stages of folding created new fractures and thus greatly enhanced the overall permeability. During this stage, geochemically distinct fluids from the lower paleohydrostratigraphic unit were expelled into the upper paleohydrostratigraphic unit. Overall, the initiation, style, and evolution of the fold complex may have been influenced by the early stratification of the hydrologic system, as well as the later upward expulsion of fluids.

CONCLUSIONS

The data presented here provide new evidence for a complex evolution of the fluid-rock system in the Nuncios fold complex. Two novel conclusions of our study are: (1) the fluid-rock interaction in the fold complex was compartmentalized within two major hydrostratigraphic units, independent of their lithologies, during most of the folding process. Compositionally distinct fluids evolved in each compartment. The basal, 1000-m-thick evaporite sequence was a principal source of geochemically distinct fluids in the lower unit. Carbonates dominated fluid geochemistry in the upper unit. (2) The migration of high-pressure fluids from the lower unit into the upper unit was possible only during increased deformation associated with late-stage fold tightening. The spatial and temporal evolution of the fluid-rock system in detachment folds, such as presented for the Nuncios fold complex, proves to be different from the evolution described in typical fold-and-thrust structures.

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