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STUDIES OF ALTERED VITROPHYRE FOR THE PREDICTION OF NUCLEAR WASTE REPOSITORY-INDUCED THERMAL ALTERATION AT YUCCA MOUNTAIN, NEVADA

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ABSTRACT

Nuclear waste emplacement in devitrified volcanic tuff at Yucca Mountain will raise the temperature of surrounding rock for a geologically significant period of time. This study evaluates the susceptibility of an underlying 50 ft-thick vitrophyre to thermal alteration by examining alteration that occurred in the rock as it cooled after deposition. A 10°C temperature rise should have no mineralogical effects on the vitrophyre, but an increase of 60° or more is likely to result in alteration. Expected mineralogic changes in the vitrophyre caused by this amount of thermal loading include crystallization of zeolites and smectite. Alteration will be concentrated in a thin interval near the top of the vitrophyre and along fractures. Adsorbed water and water in pre-existing hydrous minerals and in glass may contribute to hydrothermal alteration of underlying vitrophyre. Bulk porosity change would be slight and local porosity increase would probably be restricted to the upper part of the vitrophyre. Although some fracture filling could occur, such a minor sealing effect would be balanced by development of secondary porosity. Zeolites and smectite, newly-crystallized along fluid flow paths below the waste repository, could provide an enhanced sorptive barrier to radionuclide migration.

1.

INTRODUCTION

Nuclear waste emplacement in an underground repository will raise the temperature of surrounding rock for a geologically significant period of time [1,2], making it necessary to establish that thermal loading will not reduce the capacity of the geologic medium to isolate the nuclear waste. This study, as part of the Nevada Nuclear Waste Storage Investigations managed by the Nevada Operations Office, US Department of Energy, is concerned with thermal effects at Yucca Mountain in southern Nevada (Fig. 1). The geologic setting is a thick sequence of silicic volcaniclastic rocks, with the candidate host rock for repository construction in densely welded devitrified tuff [3]. A densely to moderately welded glassy tuff (vitrrophyre) beneath the devitrified tuff may be especially susceptible to thermal alteration. The alteration that originally occurred in this rock as it cooled following deposition can be used to predict repository-related thermal alteration. This approach eliminates the necessity for extrapolation of conditions and effects from different lithologies and locations, a problem common to many natural analogue studies.

METHODS OF STUDY

Core samples for this study come from drill holes at Yucca Mountain and vicinity (Fig. 1). The area of coverage is large enough to permit investigation of possible effects of lateral variation in present subsurface

FIG. 1. Location map for samples in this study.

temperature. Rock units were also examined in outcrop. Sample mineralogy and texture were studied by petrographic microscope, x-ray diffraction, and electron microprobe. Oxygen isotope determinations for secondary minerals to help elucidate alteration conditions are in progress.

GEOLOGY OF THE YUCCA MOUNTAIN AREA

The Paintbrush Tuff is the uppermost sequence of Tertiary volcaniclastic rocks distributed throughout Yucca Mountain. Within this sequence, the oldest and thickest unit is the Topopah Spring Member; the Member is about 1200 ft thick at central Yucca Mountain, becoming slightly thinner to the south [4,5]. The area of Yucca Mountain under study as a possible nuclear waste repository is described by Bish et al [6]. A generalized stratigraphic column of the lower portion of the Topopah Spring unit (Fig. 2) shows the distribution of glassy and devitrified tuff. The proposed position for the nuclear waste repository, as indicated in the figure, lies within a segment of devitrified densely welded tuff several

FIG. 2. Generalized stratigraphic and lithologic section of lower Topopah Spring vitrophyre.

FIG. 3. Fracture-controlled alteration of vitrophyre, sample GU-3-1195. Above: photomicrograph, plane-polarized light, long dimension is 2.5 mm. Below: identification of features in photomicrograph. Inset shows location of photomicrograph relative to fracture and bordering devitrified zone.

hundred feet thick, overlying a 50-ft thick vitrophyre. The typical transition downward from devitrified to glassy tuff, as seen in outcrop, is a 20-ft thick interval of interfingering glassy and devitrified tuff. At the base of this interval and extending downward into the vitrophyre are numerous fractures bordered by devitrified zones up to 1 ft thick.

DESCRIPTION OF NATURAL ALTERATION

Two types of alteration are considered here: (1) the crystallization of glassy rock to anhydrous silica and feldspar minerals (devitrification) and (2) the crystallization of glass to hydrous minerals, principally the zeolite heulandite and the smectite clay beidellite. Devitrified tuff overlying the vitrophyre and some devitrified zones bordering fractures within the vitrophyre are mixtures of microcrystalline silica (cristobalite and quartz) and feldspar [7]. A much finer grained devitrification texture consisting of cryptocrystalline (crystals not visible under 400x magnification) cristobalite-feldspar mixtures is common throughout the vitrophyre but especially in the upper part; it is usually related to fractures [7,8].

Heulandite and smectite alteration products in the vitrophyre exhibit a variety of textures [8], but are almost always spatially associated with cryptocrystalline devitrification. Pore-filling heulandite (and minor smectite) is the main exception. The photomicrograph in Fig. 3 shows one type of heulandite and smectite occurrence related to a fracture. Silica and feldspar in the devitrified zone bordering the fracture become finer grained away from the fracture. Within the outer cryptocrystalline part of the devitrified zone are small pockets of heulandite. Similar zeolite pockets are present within the cryptocrystalline devitrified material surrounding larger lithic inclusions in the vitrophyre; heulandite also fills fractures in the rock. Smectite is most abundant just beyond the cryptocrystalline outer part of the devitrified zone, but is present throughout the vitrophyre. Additional alteration is evident in the same area; glass shards in the vitrophyre are partly dissolved, creating a small amount of secondary porosity.

Hydrous mineral content in altered vitrophyres varies from 6 wt% for the sample illustrated in Fig. 3 up to 15% heulandite and 60% smectite [7,8]. In highly altered core samples, the interrelation of fractures, devitrification, and hydrous mineral crystallization is not always apparent.

DISTRIBUTION OF NATURAL ALTERATION

An interval about 2 ft thick of partly devitrified tuff with secondary hydrous minerals is nearly ubiquitous along the interface between devitrified tuff and vitrophyre throughout Yucca Mountain and vicinity. The amount and nature of alteration deeper in the vitrophyre is highly variable. At southern Yucca Mountain and nearby Busted Butte, there is little or no alteration below the transition zone; in a drill hole at eastern Yucca Mountain the entire vitrophyre section is highly altered. Causes of this variation are under study; they may relate to factors influenced by thickness of the volcanoclastic units or proximity to the volcanic source. Some variation in amount of alteration probably reflects local conditions such as fracture abundance.

NATURAL ALTERATION CONDITIONS

No definite conclusions have been reached regarding the origin of heulandite and smectite in the Topopah Spring vitrophyre. The possibility that these minerals are alteration products formed during a late stage in the cooling of a volcanoclastic deposit is being investigated by the following means: (1) textural relations such as those described in the previous two sections; (2) comparison with other zeolitized rocks at Yucca

Mountain, especially in regard to the position of the static water level; (3) comparison with similar alteration in known thermal environments; and (4) oxygen isotope studies of smectites and silica fracture fillings texturally associated with heulandite.

Zeolitization has been widespread in stratigraphic units below the vitrophyre and is interpreted as diagenetic alteration of glassy tuffs at or below the static water level [9,10]. In a generalized way, the distribution of zeolitized and nonzeolitized tuff is related to present or former elevations of the static water level [8]. The vertical zonation of zeolite minerals follows trends recognized by Iijima and Ohwa in diagenetic regimes (low-temperature burial metamorphism in which temperature increases with depth) [11].

The distribution of heulandite and smectite in the vitrophyre seems to be controlled mainly by the location of the devitrified tuff-glassy tuff transition zone in the Topopah Spring tuff and bears no relationship to past or present static water levels. Vitrophyre east and west of Yucca Mountain is presently within the saturated zone, but preliminary study shows that it is not consistently more altered than vitrophyre in central Yucca Mountain that has possibly never been below the water table [12].

According to Iijima and Ohwa, heulandite is a possible product of burial diagenesis [11], but it should tend to form at equal or higher temperatures than the zeolite clinoptilolite [13]. However, the clinoptilolite-rich zeolitic rocks at Yucca Mountain are all below the heulandite-bearing altered vitrophyre, again indicating that the occurrence of heulandite in the Topopah Spring vitrophyre cannot be easily fitted into a burial diagenetic regime.

Heulandite and smectite have been reported as alteration products of glassy silicic tuff in an active hydrothermal system over the temperature range 85 to 150°C [14]. Alteration textures are very similar to some of the textures in samples from Yucca Mountain. The similarities are suggestive but not conclusive, for heulandite is not a reliable temperature-diagnostic mineral. Silica fracture-filling and smectite known by textural relations to have formed about the same time as the heulandite may provide temperature information through oxygen isotope study in progress. Preliminary results for smectite have not defined any temperature limits or alteration. This study is not expected to yield precise temperatures because of uncertainties regarding partition coefficient values and isotopic composition of water from which secondary minerals precipitated.

Thermal data from drill holes indicate that temperatures in the vitrophyre beneath Crater Flat are about 10°C higher (around 37°) than in the same unit at central Yucca Mountain [15]. These thermal conditions may not pertain to the alteration regime in which heulandite and smectite crystallized, but they provide the basis for examining effects of a small temperature rise in a repository. The degree of vitrophyre alteration in Crater Flat drill holes, as already mentioned, is no greater than at Yucca Mountain, and there are no differences in the nature of alteration. A 10°C temperature increase should therefore be insufficient to produce any significant alteration of vitrophyre below a nuclear repository.

VOLCANICLASTIC AND REPOSITORY-INDUCED THERMAL REGIMES

If natural alteration in the vitrophyre occurred at temperatures likely to be duplicated by a repository-induced thermal regime, then it will be helpful to design a model for cooling conditions in the Topopah Spring tuff to compare with thermal models for the repository. During evolution of the vertical temperature profiles under volcaniclastic and repository conditions, there may be periods of close similarity. Correlation of natural thermal conditions and the resulting alteration provides the basis for prediction of repository-induced alteration.

Cooling behavior of silicic volcaniclastic rocks has been modeled by Riehle for deposits up to 40 m (131 ft) thick (before compaction) [16].

Although the ash flow containing the Topopah Spring vitrophyre was thicker than 131 ft, the vertical temperature profile should have been similar to Riehle's model. At the time of deposition, there was a uniform high temperature throughout the deposit. A thermal gradient developed during cooling, with cooler temperatures at the top and base of the flow and the highest temperature slightly below the center of the flow. Based on degree of welding and bulk density values [17], the vitrophyre and underlying moderately welded tuff represent the lower to lower central portion of the ash flow below the area of maximum temperature.

Initial thermal conditions following nuclear waste emplacement will differ from conditions in newly deposited ash flows because temperatures will be increasing. Repository thermal loading should eventually produce a maximum temperature around the waste emplacement area and decreasing temperatures downward into the vitrophyre; the highest temperatures reached within the vitrophyre during this stage may persist several thousand years [12]. These conditions may correspond to the natural thermal regime about 10^2 to 10^3 years after pyroclastic deposition when the temperature within the vitrophyre was probably 100 to 200°C or less. The vitrophyre should have remained around this temperature hundreds to thousands of years [16].

The clear role of fractures in controlling natural alteration suggests that convective heat transport along fractures may be equally or more important in this respect than the conductive heat transfer emphasized by Riehle. Other modeling factors of possible importance include moisture content, effects of mineralogical reactions, and the mass of rock involved in such reactions.

POSSIBLE REPOSITORY-INDUCED ALTERATION AND ITS EFFECTS

Thermal effects of nuclear waste emplacement on surrounding rock should not be permitted to reduce the capacity of the geologic repository to isolate waste [18]. Unfavorable effects include any changes that would decrease the travel time of contaminated fluids from the repository to the accessible environment. Increased rock permeability and degradation of minerals with radionuclide sorptive capacity are examples of possible unfavorable effects.

Mineralogic effects of natural thermal alteration in Topopah Spring vitrophyre include (1) glass dissolution without in situ crystallization of secondary minerals, (2) in situ crystallization of glass to feldspar and cristobalite, (3) in situ crystallization of glass to smectite and/or heulandite, and (4) crystallization of heulandite and smectite as fracture filling.

Glass dissolution increases porosity locally and may affect permeability. In situ crystallization of glass to anhydrous or hydrous minerals seems to have little local effect on the porosity, whereas crystallization of these minerals in fractures should reduce porosity and permeability. Calculated porosity values (3 to 4%) for several vitrophyre core samples show no differences among more or less altered samples [17], suggesting that glass dissolution and new mineral precipitation tend to compensate each other. Few data are available regarding permeability; one sample of altered moderately welded vitric tuff (below vitrophyre) has about the same low permeability as densely welded devitrified Topopah Spring tuff [19]. As can be seen in Fig. 3, new pores formed during alteration tend not to be interconnected and therefore should not significantly increase the permeability.

Two issues regarding possible changes in mineral properties require investigation: the effect of heating on pre-existent sorptive minerals (smectite and heulandite) and the sorptive behavior of these minerals, pre-existent or newly-crystallized, at elevated temperature. Little is presently known about the behavior of smectite and heulandite from altered Topopah Spring vitrophyre upon heating. The effects of water content and

heating on bulk rock properties are unknown. D. Bish has determined the amount of crystal lattice contraction for cation-exchanged clinoptilolite upon heating at room humidity [20] and will investigate the effects of heating under controlled humidity.

Availability of water may affect the likelihood of alteration. In addition to loosely adsorbed water, the vitrophyre contains at least 3 wt% bound water [21]; highly altered rocks probably contain more bound water. The movement of hot water away from the repository along fractures may be important in initiating alteration.

If repository-induced alteration follows the pattern of natural thermal alteration in the vitrophyre, new alteration products should crystallize along fractures in glass near the top of the vitrophyre. Some glass alteration or precipitation of new minerals may occur along or within fractures deeper in the vitrophyre. These minerals, newly crystallized along fluid flow paths beneath the nuclear waste repository, could provide an enhanced sorptive barrier to radionuclide migration [19].

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