American Mineralogist, Volume 58, pages 555–557, 1973

Compositions of Natural Sillimanites from Volcanic Inclusions and Metamorphic Rocks: A Discussion

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Kwak (1971) presented the results of electron microprobe analyses of sillimanite from 32 rocks showing a variety of parageneses. Among these were four samples from the inclusion-rich biotite-cordierite-labradorite dacite at Cerro del Hoyazo, SE Spain, *viz.*, Nos. 26–29 of Table 1 in Kwak (1971). Of these four analyses, three are of sillimanite found in 5–50 cm large inclusions of Al-rich rock in the lava.[1] Each of the three sillimanites is from a different type of Al-rich rock inclusion. Thus, *No. 26* is from a foliated cordierite-almandine-biotite-sillimanite rock (for description see Zeck, 1968, pp. 65–72; the analyzed material probably represents sample Z 78 Ho, and not Z 73 Ho as indicated by Kwak, 1971); *No. 27* is from a foliated almandine-sillimanite-biotite-quartz-cordierite rock (No. Z 46 Ho ?, Zeck, 1968, pp. 72–75); and *No. 29* is from a non-foliated spinel-cordierite-almandine-andalusite-sillimanite rock (No. Z 9 Ho, Zeck, 1968, pp. 75–80). The fourth analytical figure *(No. 28)* combines the analytical results of two sillimanite fragments immersed in the lava's glassy base, *viz.* 1) a relatively small fragment of fibrolite felt, the like of which are of common occurrence in the glassy base, and were interpreted (Zeck, 1968, pp. 57, 127) as being derived from rocks similar to the three types of Al-rich rock inclusions just mentioned; and 2) a "large" sillimanite crystal immersed in the lava glass base. The size of the sillimanite crystal was not given by Dr. Kwak, but assuming that this crystal is considerably larger than the abundant acicular sillimanite crystals in the glass base of the lava, I assume it to be a "mono-crystal" inclusion derived from the kind of rocks which form the Al-rich inclusions.

Kwak (1971) classified the 32 analyzed sillimanites into two "geological environments": (a) middle almandine amphibolite facies (600°C, ± 5 kbar minimum ?) to granulite facies rocks, and (b) inclusions in dacite and olivine basalt lavas (about 1150°C, ± 0.5 kbar ?). The analyzed sillimanites of the four samples from Cerro del Hoyazo were put in group (b).

Dr. Kwak set out to test the possibility of solid solutions between natural mullite and sillimanite, as proposed by Aramaki (1961) and later defended for artificial material by Hariya, Dollase, and Kennedy (1969). In this connection Kwak (1971, p. 1751) noted that "analysis of sillimanite in inclusions from low pressure-high temperature environments such as lavas . . . should show the solid solution to the degree Aramaki's work suggests, especially if corundum occurs as a buffering phase." I feel that this statement is not quite to the point. The crucial point seems not whether the analyzed "sillimanite" phase occurs in a sample which passed previously through a HT-LP environment in which the supposed solid solution would be stable (in terms of Gibbs free energy), but rather if it *formed* under these conditions, meaning that also the kinetical factors should permit the reaction.

As for the "sillimanite" crystals analyzed by Aramaki (1961), there appears to be little doubt as to their pyrometamorphic (HT-LP, sanidinite facies conditions of) origin. The "sillimanite" crystals occur in accidental inclusions (in an andesite) which appear to be fragments of pyrometamorphic equivalents of non-metamorphic or very low-grade regional metamorphic rocks, which immediately underlie the present volcano. Consequently, the sillimanite, corundum, cordierite, *etc,* in these inclusions, is due to pyrometamorphism.

In the case of the inclusions in the Cerro del Hoyazo dacite, the situation is not as simple. In this dacite and its inclusions, "sillimanite" is found in three types of occurrence (Zeck, 1968, 1970): 1) acicular, isolated crystals in the glass base of the lava; 2) acicular crystals, usually assembled in fibrolite aggregates (but locally coarser grained crystals occur), in the Al-rich rock inclusions and in

[1] Note that in Nos. 26–29 of Table 1 (Kwak, 1971), "C" (indicating the presence of corundum) should be read as "Co" (for cordierite).

small fragments derived from these *rock* inclusions; and 3) acicular crystals, usually assembled in rather loose aggregates, in accidental inclusions of fine-grained schists to quartzites. A detailed description of the three types of "sillimanite" and a discussion of their origin was given in the earlier papers (Zeck, 1968, 1970). As it might be cumbersome for the reader to extract the pertinent data from the extensive 1968 paper, some data which have bearing on the current discussion are briefly related here.

The type 1) "sillimanite" crystals are idiomorphic, very small (often 30–60 μ long and 1–2 μ wide) and are found in considerable numbers isolated in the glass base of the fresh lava. Therefore it was concluded that these crystals "probably are precipitation products of the magma" (Zeck, 1968, p. 128).

The type 2) sillimanite crystals form an important constituent of all Al-rich rock inclusions, which further consist mainly of biotite, cordierite, almandine, plagioclase, quartz and spinel. The rocks show a (relict) foliation, chiefly sustained by fibrolite-biotite streaks. Except for sillimanite—which is mainly in the variety fibrolite—the major constituents have a grain size between 1 mm and several cm. Many cordierite crystals reach a size of 3–10 cm. In thin section the rocks render definite proof that these cordierite crystals postdate the fibrolite (Zeck, 1968, pp. 128, 129, 131, 132; see also Zeck, 1970, pp. 230–231, Fig. 2). As the size of the cordierite crystals precludes a pyrometamorphic origin, the fibrolite must also be formed before the eruption of the magma. The fibrolite fits in with the other, medium to (very) coarse mineral grains, defining rocks formed under comparatively deep-seated conditions (amphibolite facies and hornblende hornfels facies) (Zeck, 1968, 1970).

In sharp contrast with this main assemblage in the Al-rich inclusions stands the occurrence of very small amounts of glass containing minute crystals mainly of spinel, sanidine, plagioclase and hypersthene ($\varnothing \simeq$ 5–50 μ). The vesicular glass with its immersed crystals occurs in irregular veins, often at grain boundaries of crystals of the much coarser grained assemblage, or fills embayments and rounded holes in these crystals. This textural relationship is taken to indicate that the glass and immersed crystals were formed from, or in connection with, a melt phase which originated at the expense of crystals of the older assemblage (Zeck, 1968, pp. 129–130, 131, 134). Based on these textural relations it is possible to distinguish consistently and unequivocally between the older assemblage and the newer one. This sharp difference is interpreted to reflect a major break in conditions of origin: the older, medium to (very) coarse-grained assemblage would have been formed under comparatively deep-seated conditions (see above), whereas the very fine-grained, glass-immersed grains would have been formed in relation to the eruption of the dacitic magma. Though of minor interest in the present discussion, it might be worth noting that the Al-rich inclusions, amounting to about 10 percent of the total dacite, were not interpreted as "normal" pelitic rock fragments nor as "normal" accidental inclusions, but as anatectic restite material, syngenetical with and complementary to the magmatic melt. (A list of arguments sustaining this conclusion is given in Zeck, 1970, pp. 238–240.)

The third type of "sillimanite" occurs in a quite different type of inclusion which makes up less than 0.1 percent of the total dacite, and which is readily and consistently distinguishable from the Al-rich inclusions. These inclusions consist of rather fine-grained, low to medium grade pelitic schists (to quartzites) little different from the common pelitic schists which (presumably) form the country rock of the upper part of the volcanic vent, except that the schist fragments enclosed in the dacite show a pyrometamorphic imprint which resulted in the formation of glass, spinel, sanidine and "sillimanite." These inclusions were interpreted as accidental (enallogene) (Zeck, 1970, p. 240). Also here, as in the Al-rich inclusions discussed above, the regional metamorphic paragenesis can on textural grounds be clearly distinguished from the pyrometamorphic one. The "sillimanite" in this type of inclusion forms rather loosely built, schistosity-parallel aggregates, which in thin section cut parallel to the schistosity of the rock, and show a radial pattern; glass occurs in beween the "sillimanite" needles. One could imagine that these aggregates have originated from decomposition of colorless mica (Zeck, 1968, p. 137), which is strikingly absent in the schist inclusions, whereas it is a major constituent in schists in the basement.

The Cerro del Hoyazo sillimanites Dr. Kwak analysed came from the Al-rich inclusions, and thus belong to the second type of occurrence. The petrographic evidence cited above suggests that these sillimanite crystals have not been formed under shallow, HT-LP conditions. "Sillimanite" crystals formed under HT-LP conditions are present in the Cerro del Hoyazo (the third and probably also the first type of occurrence mentioned above), but analytical results regarding these were not reported by Kwak (1971).

After finding that the analysed crystalline phases showed a stoichiometric sillimanite composition (within the error of measurement), Dr. Kwak (1971, p. 1758) concluded: "no reason is known for the differing conclusions of Aramaki (1961) and the present study, but it appears that possible mullite-sillimanite solid solutions at values greater than the 3 wt percent (the maximum error of measurement) in nearly all geological materials need not worry the petrologist." In view of the above this conclusion appears not sufficiently well founded. This will be even more the case if it should appear that the same objections as related above for the Cerro del Hoyazo samples can also be made regarding the samples from the second, and only other, volcanic occurrence studied by Dr. Kwak (Finkenberg, Germany). In fact, the coarse grain size of the sillimanite in the Finkenberg samples (to 2 cm long, Kwak, 1971, p. 1758) favors such a supposition.

It may be worth stressing that the issue raised in this discussion is not whether the conclusion of Aramaki (1961), that mullite-sillimanite solid solutions may be formed under unusual HT-LP conditions, is correct or not, but rather whether the arguments which Dr. Kwak employed to reject Aramaki's conclusions are conclusive. In fact, recently published results of Cameron and Ashworth (1972) on material from xenoliths from Asama volcano (the same material as studied by Aramaki, 1961) cast doubt upon Aramaki's assertion. Also results of earlier work (*e.g.* Agrell and Smith, 1960; Aramaki and Roy, 1963; Zeck, 1968, p. 94) and preliminary results of current research of the present author point in that direction.

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