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Comment on "Multipath Ar transport in K-feldspar deduced from isothermal heating experiments" by Igor Villa

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Villa [1] interpreted ^{39}Ar loss experiments performed with a low-temperature, igneous K-feldspar (ga1) as being inconsistent with multi-diffusion domain model (MDD; see [2]). The applicability of the MDD model for ga1 was dismissed in [1] exclusively on the basis of two features of the experiments: (1) initial steps at lower temperature produced more gas loss than initial steps at higher temperature; and (2) a sharp drop in $\log(D/r^2)$ occurred in successive isothermal steps. However, failure of these experiments to reproduce previously determined ^{39}Ar loss systematics for the same material [19] and improper characterization of the domain distribution of ga1 indicate that the basis for the conclusion is flawed.

Three aliquots of ga1 were step-heated, with each experiment beginning with isothermal steps at a different temperature. While more Ar is normally expected to be released at higher temperatures, the first 580°C step of ga1c liberated nearly twice as much ^{39}Ar ($\sim$ 7%) as the first 670°C step of ga1b ($\sim$ 4%), and about the same amount as the first 750°C step of ga1a ($\sim$ 7%). All heating durations were 10 min (see table 1 of [1]). The generality of the conclusions arising from these isothermal experiments are contradicted by every K-feldspar step-heating experiment in which $\log(D/r^2)$ monotonically rises with $1/T$ through the temperature interval 500–$\sim$ 750°C. This includes the vast majority of K-feldspar results reported by a diverse group of geochronologists (e.g., [2–18,20–23]).

In the previous analysis of ga1 (GA-1-N, see [19]), the first four heating steps (550, 650, 700 and 750°C) released 3.1%, 6.1%, 5.5% and 4.3% of ^{39}Ar, respectively. Although heating durations were not reported [19], we interpolate the curve fits to GA-1-N presented in [1] (dashed, solid and dot lines in fig. 1a) to estimate $\log(D/r^2)$ of these steps. These calculations (Fig. 1) require that the heating durations be only $\sim$ 2 min. The monotonically increasing values of $\log(D/r^2)$ with temperature exhibited by GA-1-N are difficult to reconcile with the complex pattern shown by the three isothermally treated aliquots (ga1a, ga1b and ga1c) without concluding that problems exist with the experimental approach. Note that the highly unusual heating schedule required to reconcile the two data sets (2 min at 550°C followed by 50 min at 650°C) would not reproduce the MDD model results shown in fig. 1a of [1]. Moreover, the multipath behavior invoked in [1] to account for the isothermal results lacks merit in that the previous results [19] cannot be accounted for.

The small variation in $\log(D/r^2)$ for MH-10 between 7% and 16% of ^{39}Ar released, shown in fig. 1a of [1] is correct and consistent with our earlier conclusion that this degassing interval is dominated by a particular domain size in this sample (see p. 2065 of [3]). Characterization of the MDD model as predicting a uniform value of D/r^2 with increasing

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F (see caption of fig. 1 in [1]) is incorrect because that is precisely the behavior expected for a sample containing but a single diffusion domain size. A sharp decrease in D/r^2 with increasing F as indicated in fig. 1a of [1] is, in fact, typical of K-feldspars that contain a relatively small volume fraction of the smallest domain (e.g., FA-1; [8]). The significant drops in $\log(D/r)^2$ between the subsequent initial steps in the isothermal experiments and the apparent low activation energy shown in Fig. 1 (~ 27 kcal/mol) suggest that the smallest diffusion domains of ga1 comprise ~ 5% or less of the total gas. Note that, in this case, the GA-1-N step-heating data would not be capable of revealing the domain distribution, because ~ 15% of the gas was released in the first three steps. Therefore, aside from possible experimental problems, any apparent inconsistency with the MDD model viewed by Villa [1] (i.e., a small decrease in D/r^2 with increasing F) is probably due to an incorrect estimate of the distribution parameters for ga1 using data resulting from an inadequate step-heating schedule. In addition, the inconsistencies observed between the two data sets (GA-1-N step-heating and ga1 isothermal heating) preclude the domain distribution obtained from GA-1-N from being used to interpret the isothermal results [1] in terms of the applicability of the MDD model.

Finally, we emphasize that the ability to model ^{39}Ar release data alone is *not* the virtue of the MDD model, since the ^{39}Ar data account for only half the story. The best evidence that the diffusion mechanisms and boundaries that define natural Ar retention in K-feldspars are the same as those interrogated during laboratory heating is the correlated relationship between the age spectrum and its associated $\log(r/r_0)$ plot (see [2–8,13]). The main virtue of the model is that the existence of a distribution of diffu-

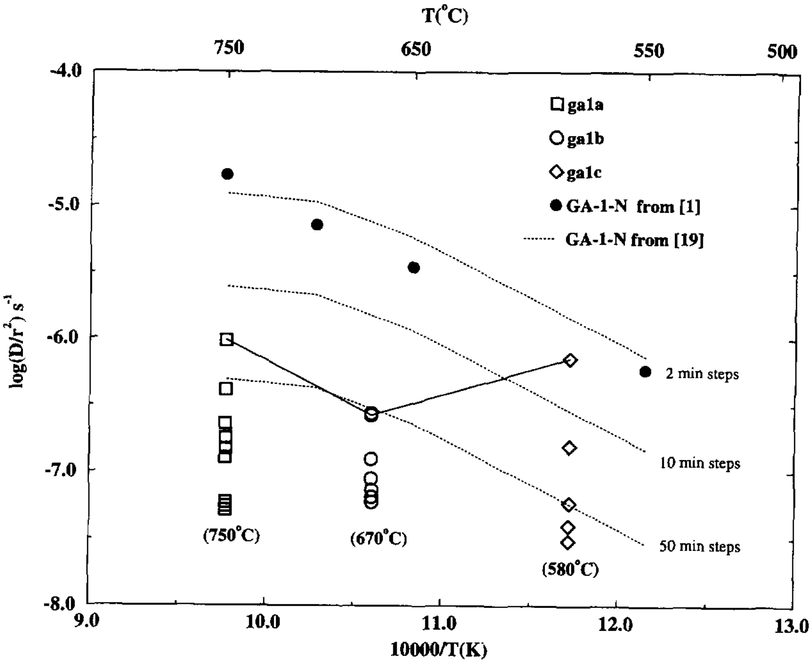

Fig. 1. Arrhenius diagram of data reported in [1] from three step-heating experiments performed on ga1 feldspar. Also plotted are estimated $\log(D/r^2)$ values from a previous step-heating experiment performed with the same material (GA-1-N in [19]). Because heating times were not provided in [19], $\log(D/r^2)$ values were obtained by interpolation of the curves fits of GA-1-N shown in fig. 1 of [1]. Note that the resulting values require a step time of only ~ 2 min in the GA-1-N step-heating sequence. (Dashed curves show calculated values from the GA-1-N ^{39}Ar released [19] assuming 2, 10, or 50 min durations for each step.) Moreover, the > 600°C steps of GA-1-N plot about an order of magnitude higher than the ga1a (750°C) and ga1b (670°C) data. Finally, note that the highly unusual heating schedule required to reconcile the two data sets (2 min at 550°C followed by 50 min at 650°C) would not reproduce the model results shown in fig. 1a of [1].

sion domain sizes explains correlations between inflections observed in both the age spectrum (reflecting processes operating over millions of years) and associated Arrhenius plots (created in the laboratory in a matter of hours). We are not aware of any other proposal that explains this relationship. Unfortunately, both the low and high temperature portions of gas release from ga1 K-feldspar [19] are contaminated by excess Ar, precluding assessment of this key relationship. Thus, Villa [1] bases his conclusions upon inconsistent observations, contradicted by the vast majority of the literature (e.g., [2–18,20–23]), of only one-tenth (i.e., between 7% and 16% ^{39}Ar release) of one half of the available evidence. The other half — that recorded in the age spectrum — is ignored. *[FA]*

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