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## Potassium-Argon and Argon-Argon Dating of Crustal Rocks and the Problem of Excess Argon

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According to the assumptions foundational to potassium-argon (K-Ar) and argon-argon (Ar-Ar) dating of rocks, there should not be any daughter radiogenic argon ( $^{40}\text{Ar}^*$ ) in rocks when they form. When measured, all  $^{40}\text{Ar}^*$  in a rock is assumed to have been produced by *in situ* radioactive decay of  $^{40}\text{K}$  within the rock since it formed. However, it is well established that volcanic rocks (e.g. basalt) contain excess  $^{40}\text{Ar}^*$ , that is,  $^{40}\text{Ar}$  which cannot be attributed to either atmospheric contamination or *in situ* radioactive decay of  $^{40}\text{K}$ .<sup>1</sup> This excess  $^{40}\text{Ar}^*$  represents primordial Ar carried from source areas in the earth's mantle by the parent magmas, is inherited by the resultant volcanic rocks, and thus has no age significance.

However, are all other rocks in the earth's crust also susceptible to "contamination" by excess  $^{40}\text{Ar}^*$  emanating from the mantle? If so, then the K-Ar and Ar-Ar "dating" of crustal rocks would be similarly questionable.

When muscovite (a common mineral in crustal rocks) is heated to 740°–860°C under high Ar pressures for periods of 3 to 10.5 hours it absorbs significant quantities of Ar, producing K-Ar "ages" of up to 5 billion years, and the absorbed Ar is indistinguishable from radiogenic argon ( $^{40}\text{Ar}^*$ ).<sup>2</sup> In other experiments muscovite was synthesized from a colloidal gel under similar temperatures and Ar pressures, the resultant muscovite retaining up to 0.5 wt% Ar at 640°C and a vapor pressure of 4,000 atmospheres.<sup>3</sup> This is approximately 2,500 times as much Ar as is found in natural muscovite. Thus under certain conditions Ar can be incorporated into minerals which are supposed to exclude Ar when they crystallize.

Patterson et al. envisage noble gases from the mantle (and the atmosphere) migrating and circulating through the crust, so there should be evidence of excess  $^{40}\text{Ar}^*$  in crustal rocks.<sup>4</sup> Noble gases in  $\text{CO}_2$ -rich natural gas wells confirm such migration and circulation—the isotopic signatures clearly indicate a mantle

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origin for the noble gases, including amounts of excess  $^{40}\text{Ar}^*$  in some  $\text{CO}_2$ -rich natural gas wells exceeding those in mantle-derived mid-ocean ridge basalts.<sup>5</sup> In fact, the quantities of excess  $^{40}\text{Ar}^*$  in the continental crust can be as much as five times that found in such mantle-derived mid-ocean ridge basalts, strongly implying that excess  $^{40}\text{Ar}^*$  in crustal rocks and their constituent minerals could well be the norm rather than the exception.

Dalrymple, referring to metamorphism and melting of rocks in the crust, has commented: "If the rock is heated or melted at some later time, then some or all the  $^{40}\text{Ar}$  may escape and the K-Ar clock is partially or totally reset."<sup>6</sup> Thus  $^{40}\text{Ar}^*$  escapes to migrate in the crust to be incorporated in other minerals as excess  $^{40}\text{Ar}^*$ , just as  $^{40}\text{Ar}^*$  degassing from the mantle does. Excess  $^{40}\text{Ar}^*$  has been recorded in many minerals (some with essentially no  $^{40}\text{K}$ ) in crustal rocks—quartz, plagioclase, pyroxene, hornblende, biotite, olivine, beryl, cordierite, tourmaline, albite, and spodumene.<sup>7</sup> The Ar-Ar method has also been used to confirm the presence of excess  $^{40}\text{Ar}^*$  in feldspars and pyroxenes.<sup>8</sup> In a recent study 128 Ar isotopic analyses were obtained from ten profiles across biotite grains in high-grade metamorphic rocks, and apparent Ar-Ar "ages" within individual grains ranged from 161 Ma–514 Ma.<sup>9</sup> This cannot be solely due to radiogenic build-up of  $^{40}\text{Ar}^*$ , but due to incorporation by diffusion of excess  $^{40}\text{Ar}^*$  from an external source, namely,  $^{40}\text{Ar}^*$  from the mantle and other crustal rocks and minerals. Indeed, a well-defined law has been calculated for  $^{40}\text{Ar}$  diffusion from hornblende in a gabbro due to heating.<sup>10</sup> Excess  $^{40}\text{Ar}^*$ , which accumulated locally in the intergranular regions of the gabbro, reached partial pressures in some places of at least  $10^{-2}$  atm.

This crustal migration of  $^{40}\text{Ar}^*$  is known to cause grave problems in regional geochronology studies. For example, in the Middle Proterozoic Musgrave Block (northern South Australia), a wide scatter of K-Ar mineral "ages" was found, ranging from 343 Ma to 4493 Ma due to inherited (excess)  $^{40}\text{Ar}^*$ , so no meaningful interpretation could be drawn from the rocks.<sup>11</sup> Of the diabase dikes which gave anomalous "ages," it was concluded that the basic magmas probably formed in or passed through zones containing a high partial pressure of  $^{40}\text{Ar}^*$ , permitting inclusion of the gas in the crystallizing minerals. Likewise, when Ar "dating" was attempted on Proterozoic granulite-facies rocks in the Fraser Range (western Australia) and Strangways Range (central Australia), it was found that garnet, sapphirine, and quartz contained excess  $^{40}\text{Ar}^*$  that rendered the Ar dating useless because of "ages" higher than expected.<sup>12</sup> The excess  $^{40}\text{Ar}^*$  was probably incorporated at the time of the formation of the minerals, and calculations suggested a partial pressure of  $\sim 0.1$  atm Ar in the Proterozoic lower crust of Australia, which extends over half the continent.

An Ar-Ar "dating" study of high-grade metamorphic rocks in the Broken Hill region (New South Wales) found widely distributed excess  $^{40}\text{Ar}^*$ .<sup>13</sup> Plagioclase and hornblende were most affected, step heating Ar-Ar "age" spectra yielding results up to 9.588 Ga. Such unacceptable "ages" were produced by excess  $^{40}\text{Ar}^*$  release, usually at 350–650°C and/or 930–1380°C, suggesting excess  $^{40}\text{Ar}^*$  is held in sites within respective mineral lattices with different heating requirements for its release. Thus at crustal temperatures, which are less than 930°C, some excess  $^{40}\text{Ar}^*$  will always be retained in those trapping sites in minerals where it is "held" more tightly. A viable interpretation of these Broken Hill data was only produced because assumptions were made about the age of the rocks and of a presumed subsequent heating

event (based on Pb-Pb and Rb-Sr dating), when it is conjectured that accumulated  $^{40}\text{Ar}^*$  was released from minerals causing a significant regional Ar partial pressure of  $\sim 3 \times 10^{-4}\text{atm}$ .

Domains within the mantle and crust have been identified and the interaction between them described, all of which is relevant to the migration and circulation of Ar (and thus excess  $^{40}\text{Ar}^*$ ) from the lower mantle through the crust.<sup>14</sup> The six domains are physically distinct units which exhibit wide differences in average physical and chemical properties, as well as structure and tectonic behavior. They are the lower mantle (below 670 km), upper mantle, continental mantle lithosphere, oceanic mantle lithosphere, continental crust and oceanic crust, the latter four constituting the earth's crust. Each is a distinct geochemical reservoir.

A steady-state upper mantle model has been proposed for mass transfer of rare gases, including Ar.<sup>15</sup> Rare gases in the upper mantle are derived from mixing of rare gases from the lower mantle, subducted rare gases, and radiogenic nuclides produced *in situ*. Assuming a 4.5 Ga earth, it is claimed, "The lower mantle is assumed to have evolved isotopically approximately as a closed system with the *in situ* decay of  $^{129}\text{I}$ ,  $^{244}\text{Pu}$ ,  $^{238}\text{U}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  adding to the complement of initial rare gases." Thus some of the  $^{40}\text{Ar}^*$  must be primordial (*not* derived from radioactive  $^{40}\text{K}$ ), but how much is unknown. It is also claimed that  $^{40}\text{K}$  decay in the upper mantle further increases the radiogenic  $^{40}\text{Ar}$  there by a factor of  $\sim 3$  compared with the lower mantle, but this also presupposes a 4.5 Ga earth and doesn't allow for primordial  $^{40}\text{Ar}^*$  already in the upper mantle. The bulk of the  $^{40}\text{Ar}^*$  in the lower and upper mantles could be primordial, but there is no way of knowing, as primordial  $^{40}\text{Ar}$  is indistinguishable from  $^{40}\text{Ar}^*$ .

Because it is known that excess  $^{40}\text{Ar}^*$  is carried from the mantle by plumes of mafic magmas up into the earth's crust, it is equally likely that much of the excess  $^{40}\text{Ar}^*$  in crustal rocks could be primordial  $^{40}\text{Ar}$ . Thus, we have no way of knowing if any of the  $^{40}\text{Ar}^*$  measured in crustal rocks has any age significance. Additional to the primordial  $^{40}\text{Ar}$  from the mantle is  $^{40}\text{Ar}^*$  released from minerals and rocks during diagenesis and metamorphism, so that there is continual migration and circulation of both primordial  $^{40}\text{Ar}$  and  $^{40}\text{Ar}^*$  in the crust which is reflected in their presence in  $\text{CO}_2$ -rich natural gases. Therefore, when samples of crustal rocks are analyzed for K-Ar and Ar-Ar "dating," one can never be sure that whatever  $^{40}\text{Ar}^*$  is in the rocks is from *in situ* radioactive decay of  $^{40}\text{K}$  since their formation, or if some or all of it came from the mantle or from other crustal rocks and minerals. Thus all K-Ar and Ar-Ar "dates" of crustal rocks are questionable, as well as fossil "dates" calibrated by them.

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**Notes:** "Ma" represents a million years (Mega-annum); "Ga" represents a billion years (Giga-annum).

That portion of elemental  $^{40}\text{Ar}$  derived from radioactive decay is denoted by  $^{40}\text{Ar}^*$ . The remainder has no radiogenic source. The two are identical. ❄



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