Radon emanation coefficients of several minerals: How they vary with physical and mineralogical properties

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\textbf{ABSTRACT}

The escape rates of radon gas from rocks and minerals are of great relevance to many branches of geosciences, and it is, thus, important to understand the physical and mineralogical properties that control radon emanation rates. Mechanisms of radon loss from minerals have direct bearing on the reliability of U-Pb and U-Th-He geochronology. Fourteen minerals from three different mineral groups and with localities spanning three continents were selected for this study. The radon emanation coefficients (REC) for each mineral were measured as a function of grain size, temperature, \textsuperscript{238}U and \textsuperscript{232}Th activities, total absorbed \(\alpha\)-dose, density, and mineral melting temperature. The measured \textsuperscript{238}U and \textsuperscript{232}Th activities ranged from 0.01 to 6487 Bq/g and from below detection limit to 776 Bq/g, respectively. The REC values for unheated, pulverized samples ranged from 0.083 to 7.0\%, which is comparable to previously reported ranges (except for zircon). An inverse correlation between grain size and REC was observed. Full annealing of fission tracks resulted in an overall decrease in REC values, suggesting that nuclear tracks could possibly act as conduits for radon release. While activity, \(\alpha\) dose, density, and melting temperatures are not strongly correlated with REC values, it was observed that minerals with high melting points (\(\geq 1400\,{\text{\textdegree}}\text{C}\)) have lower REC values, most likely due to inhibition of radon release by compact crystal-lattice structures. This is the first attempt, to our knowledge, to correlate REC values with melting temperature, and this study reports six minerals for which no REC values have been previously reported.

\textbf{Keywords:} Radon emanation, REC, metamict minerals, nuclear track annealing, uranium, Invited Centennial article

\textbf{INTRODUCTION}

The studies of radon emanation rates from soils, rocks, and minerals have a wide range of applications in many branches of geosciences. The escape of radon from rocks and minerals is of importance to geological dating, where it can limit the reliability of U-Pb geochronology (e.g., Heaman and LeCheminant 2000; Corfù 2012; Goa et al. 2014). Radon concentration gradients observed in air (both interstitial soil and atmospheric air in planetary boundary layers and above) and groundwater are widely used as tracers to predict earthquakes, locate subsurface uranium ore and hydrocarbon deposits, and study atmospheric transport (Garver and Baskaran 2004; Nazaroff 1992; Levinson et al. 1982; Fleischer and Mogrocampero 1985; Wakita et al. 1991; Fleischer and Turner 1984; Tanner 1964, 1980; Liu et al. 1984; Kritz et al. 1993; Baskaran 2016). Radon is also used as a tracer for quantifying the rate of gas exchange across the air-sea interface (Broecker et al. 1967; Baskaran 2016). Furthermore, the inhalation of radon and its progeny poses a radiation health hazard, as radon was classified as a human carcinogen (in the same carcinogen group as tobacco smoke, asbestos, and benzene) in 1988 by the International Commission on Radiation Protection (IARC; WHO 2009). For these reasons, it is important to understand the physical and geological factors that affect radon release rates from rocks, minerals, and soils.

During the \(\alpha\) decay of \textsuperscript{\textit{226}}Ra, both an \(\alpha\)-particle with a range of \(\sim 10000\,\text{nm}\) in solids and an energetic \textsuperscript{\textit{222}}Rn recoil nucleus are produced. With a recoil energy of \(85\,\text{keV}\) and a recoil distance of \(\sim 40\,\text{nm}\) (e.g., Amin 1986: 35 nm in clays, 95 nm in water and \(64000\,\text{nm}\) in air), the recoil nucleus of \textsuperscript{222}Rn can collide with other atoms in a mineral’s crystal lattice structure and alter their arrangement (Semkow 1990). In minerals where the radiation dose exceeds \(\sim 10^{16}\,\alpha\)-decay events per mg, the mineral is reported to undergo a radiation-induced transition from the crystalline to amorphous state (Murakami et al. 1991; Weber et al. 1994). The degree of internal radiation damage to the mineral structure by recoil and fission tracks can affect a mineral’s radon emanation coefficient (REC, also known as coefficient of emanation, escape ratio, escape-to-production ratio, and percent emanation), which is the ratio of radon emitted to radon produced within the mineral. These nuclear tracks can become interconnected and increase the internal surface area of the mineral, promoting further escape of radon atoms. Radon atoms, upon formation from the decay of \textsuperscript{\textit{222}}Ra and located primarily within recoil distance, migrate either (1) from the edge of a mineral grain into pore space or pore water, (2) deeper into the mineral grain, (3) into an adjacent grain, or (4) into pore space by indirect or penetration recoil (Semkow 1991). The 40 nm recoil distance of radon atoms implies that only...