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Transient radon signals driven by fluid pressure pulse, micro-crack closure, and failure during granite deformation experiments

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ABSTRACT

In seismically active fault zones, various crustal fluids including gases are released at the surface. Radon-222, a radioactive gas naturally produced in rocks, is used in volcanic and tectonic contexts to illuminate crustal deformation or earthquake mechanisms. At some locations, intriguing radon signals have been recorded before, during, or after tectonic events, but such observations remain controversial, mainly because physical characterization of potential radon anomalies from the upper crust is lacking.

Here we conducted several month-long deformation experiments under controlled dry upper crustal conditions with a triaxial cell to continuously monitor radon emission from crustal rocks affected by three main effects: a fluid pressure pulse, micro-crack closure, and differential stress increase to macroscopic failure. We found that these effects are systematically associated with a variety of radon signals that can be explained using a first-order advective model of radon transport. First, connection to a source of deep fluid pressure (a fluid pressure pulse) is associated with a large transient radon emission increase (factor of 3–7) compared with the background level. We reason that peak amplitude is governed by the accumulation time and the radon source term, and that peak duration is controlled by radioactive decay, permeability, and advective losses of radon. Second, increasing isostatic compression is first accompanied by an increase in radon emission followed by a decrease beyond a critical pressure representing the depth below which crack closure hampers radon emission (150–250 MPa, ca. 5.5–9.5 km depth in our experiments). Third, the increase of differential stress, and associated shear and volumetric deformation, systematically triggers significant radon peaks (ca. 25–350% above background level) before macroscopic failure, by connecting isolated cracks, which dramatically enhances permeability.

The detection of transient radon signals before rupture indicates that connection of initially isolated cracks in crustal rocks may occur before rupture and potentially lead to radon transients measurable at the surface in tectonically active regions. This study offers thus an experimental and physical basis for understanding predicted or reported radon anomalies.

1. Introduction

Various fluids produced in the Earth's crust migrate through fault zones and are released at the surface in seismically active regions worldwide (e.g., Irwin and Barnes, 1980; Sato et al., 1986; Sugisaki et al., 1983; Kennedy et al., 1997; Perrier et al., 2009; Walia et al., 2010). One of those fluids, radon-222, is a radioactive noble gas of half-life of 3.82 days and is naturally produced in rocks. An alpha-decay product of radium-226 in the uranium-238 decay chain, radon-222 becomes detectable when it reaches connected pores or cracks and then exits the porous network. The ability of a rock to emit radon depends thus on the bulk radium concentration in the grains and on the emanation factor (Tanner, 1964; Sakoda et al., 2011), which accounts for radium distribution and geometry of the porous network. Radon transport from its source to the surface is governed by diffusion, advection and/or convection (Nazaroff, 1992). Due to its relatively short half-life, advection is best able to carry radon-222 from crustal layers to the surface fast enough to be measured before its decay. Therefore, radon-222 is generally used to track fluid migration in tectonic and volcanic geosystems characterized by high permeability and shallow processes (e.g., Trique et al., 1999; Cigolini et al., 2007; Girault et al., 2014). As a chemical probe to look for physical phenomena in the upper crust, radon-222 is sometimes consid-

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ered as a potential earthquake precursor (Virk and Singh, 1994; Igarashi et al., 1995). However, such intriguing observations remain controversial (Geller, 2011), mainly because physical characterization of potential radon signals from the upper crust is lacking.

Many transient signals of radon-222 concentration in soil-gas or in water have been recorded in the field. Nevertheless, only few of them are accepted by the scientific community as likely related to short time scale deformation processes such as lake-level variations, volcanic activity and earthquakes (Cox et al., 1980; Wakita et al., 1980, 1991; Igarashi et al., 1993, 1995; Virk and Singh, 1994; Trique et al., 1999; Richon et al., 2003; Steinitz et al., 2003; Wang and Manga, 2010). All of these signals show a relatively narrow peak of duration between 1 day and 1.5 week and amplitudes between 1.5 to 28 times the background levels. This peak is frequently characterized by a rapid increase and a more progressive decrease. However, at these generally poorly controlled natural sites, the source of the radon signals remains difficult to constrain. As the number of natural sites instrumented with radon sensors installed near the surface is increasing worldwide, a better understanding of the radon concentration time-series is required. Relevant rock deformation experiments have been conducted in the laboratory since the late 1970s (Holub and Brady, 1981 and references herein; King and Luo, 1990; Tuccimei et al., 2010; Mollo et al., 2011; Nicolas et al., 2014; Koike et al., 2015). These studies proposed formation and growth of micro-cracks, formation of new grain surfaces or connection of isolated micro-pores as the main effects leading to anomalous radon signals. However, little physical characterization has been attempted. For instance, the potential role of permeability changes (Manning and Ingebritsen, 1999; Manga et al., 2012) has never been investigated. In addition, all but one (Nicolas et al., 2014) of the experimental studies was performed under uniaxial compression (i.e., different from natural conditions better represented using triaxial systems). Some studies were carried out on poorly characterized samples (Holub and Brady, 1981; Belikov et al., 2014) or with only intermittent (Tuccimei et al., 2010; Mollo et al., 2011) to daily (Scarlato et al., 2013; Nicolas et al., 2014) radon monitoring. Higher time resolution is needed to capture and physically characterize the short time scales of such transient signals.

In this paper, we present several month-long deformation experiments performed on granitic rock samples with a triaxial oil-medium cell. We studied the effects of a fluid pressure pulse, micro-crack closure, and failure on radon emission from crustal rocks. Physical characterization of the radon concentration time-series is proposed as a basis to clarify predicted or reported transient radon signals, in particular those of a pre-seismic nature.

2. Radon-222 source term, sample description, and experimental system

The radon-222 source term of a sample, called the effective radium-226 concentration (EC_{Ra}), is defined by the product of the bulk radium-226 concentration C_{Ra} and the emanation factor E. It accounts for the ability of radon-222 to be produced within the sample and to reach the connected pore space, which is a prerequisite to escape from the porous network (Sakoda et al., 2011). To measure EC_{Ra} from intact and fractured samples, we applied the classical accumulation method (Ferry et al., 2002), placing the sample in a cylindrical container attached to an ionization chamber (AlphaGUARD™, Saphymo, Germany) and correcting for possible leakage in the system (Nicolas et al., 2014). Uncertainties include experimental uncertainty related to the accumulation curve (<1%) and repeatability (5%), and common overall uncertainty of the instrument (4%). The emanation factor E is then inferred from EC_{Ra}/C_{Ra}, C_{Ra} being determined using gamma spectrometry.

Table 1
Characteristics of the PL and AL samples including major elements concentrations, ^{238}U, ^{226}Ra, and ^{232}Th activity concentrations, radon-222 source term and emanation, porosity and permeability.

Sample characteristics	Portugal leucogranite (PL)	Allaire leucogranite (AL)
Concentrations of major elements (wt.%)[a]		
SiO_2	74.19	72.91
Al_2O_3	13.52	14.76
Fe_2O_3(T)	1.8	1.5
MnO	0.05	0.02
MgO	0.29	0.33
CaO	0.89	0.58
Na_2O	3.2	3.18
K_2O	4.5	4.5
TiO_2	0.196	0.197
P_2O_5	0.18	0.26
LOI	0.6	1.41
Total	99.41	99.64
Activity concentrations (Bq kg^{-1})[b]		
U-238	368 ± 13	160 ± 17
Ra-226	432 ± 125	210 ± 76
Th-232	67.0 ± 5.7	43 ± 10
Radon-222 source term and emanation		
Effective radium-226 concentration EC_{Ra} (Bq kg^{-1})	4.9 ± 0.2	11.2 ± 0.8 (heat-treated)
Emanation factor E (%)	1.1 ± 0.3	5.3 ± 2.0 (heat-treated)
Physical parameters		
Connected porosity ε	<0.02	<0.02 (heat-treated)
Axial permeability k (m^2) at confining pressure of 10 MPa	(1.5 ± 0.1) × 10^{-18}	(6.1 ± 0.3) × 10^{-18} (heat-treated)

[a] Determined by Fusion-Inductively Coupled Plasma (FUS-ICP).
[b] Determined by gamma spectrometry, Ra-226 activity being determined by its own ray.

In this study, we used two granitic rock samples typical of the upper crust, a leucogranite from Portugal (PL) and a leucogranite from Allaire, France (AL). The main characteristics of the granite samples are summarized in Table 1. Their ^{238}U, ^{232}Th, and ^{226}Ra activity concentrations are 368 ± 13, 67.0 ± 5.7, and 432 ± 125 Bq kg^{-1} for PL, and 160 ± 17, 43 ± 10, and 210 ± 76 Bq kg^{-1} for AL, respectively. To artificially increase its initial permeability, AL sample was slowly heated to 575 °C in order to induce thermal cracks (Darot et al., 1992). The radon source term EC_{Ra} and emanation factor E of the samples are 4.9 ± 0.2 Bq kg^{-1} and 1.1 ± 0.3% for PL, and 11.2 ± 0.8 Bq kg^{-1} and 5.3 ± 2.0% for AL, respectively. The PL sample has larger ^{226}Ra activity concentration, smaller EC_{Ra} and smaller permeability than the heat-treated AL sample. All experiments were conducted on cylindrical cores of diameter 4 cm and length 8 cm, stored at 40 °C for at least a week before the start of an experiment and jacketed in a neoprene sleeve during experiments.

Month-long experiments were conducted under controlled dry upper crustal conditions using a triaxial oil-medium cell designed for long-term experiments (Schubnel et al., 2005; Fortin et al., 2005). The system (Fig. 1) allows placing the sample under natural conditions, controlling confining pressure (i.e., setting depth) and axial stress using two independent volumetric servo-pumps, and controlling pore pressure using a servo-controlled delivery system of pressurized argon. The pore fluid system can be set either to maintain a constant pore fluid pressure or to inject a pore fluid volume at a constant rate. In our experiments with relatively low permeability granite samples, the pore pressure was set constant, and thus the injected volume of argon varied. Room temperature was maintained quasi-constant (±1 °C) during the experiment.

Axial and radial deformation was continuously recorded using two pairs of strain gauges glued to the sample surface. The total