Chemical Geology 367 (2014) 13–22

Contents lists available at ScienceDirect

Chemical Geology

journal homepage: www.elsevier.com/locate/chemgeo

# Zircon radiation damage ages

R.T. Pidgeon

*Department of Applied Geology, Curtin University, Perth, Western Australia, Australia*

## ARTICLE INFO

*Article history:*
Received 10 September 2013
Received in revised form 14 December 2013
Accepted 16 December 2013
Available online 22 December 2013

Editor: K. Mezger

*Keywords:*
Zircon radiation damage ages
Radiation damage annealing
Zircon dating
Annealing temperatures
Yilgarn Craton

## ABSTRACT

The suggestion that the accumulation of radiation damage in zircons could be used as a dating method was first proposed in the 1950s. In the original technique radiation damage was determined by X-ray diffraction however, this is not suitable for the small sample analysis needed in modern geochronology. It is now possible to measure radiation damage and U and Th contents on micron-sized areas on the polished surface of zircon grains using Raman spectroscopy and SIMS mass spectrometry. This opens the way for a reassessment of the potential of radiation damage ages and the purpose of this contribution is to re-examine the technique through its application to zircons from two granitoids from the Archean Yilgarn Craton and ancient detrital zircons from the Jack Hills in Western Australia. The three examples show internally consistent radiation damage ages that are also in accord with independent geochronological evidence. The 420 ± 110 Ma radiation damage age for the Darling Range granite is coincident with 500–400 Ma biotite Rb–Sr ages in the same region of the Yilgarn Craton. Likewise a tonalite from the Youanmi Terrane in the Craton with a 1650 ± 150 Ma zircon radiation damage age, lies within the domain of a 1600 Ma event recorded by biotite Rb–Sr ages. The Jack Hills zircons have a 1120 ± 130 Ma radiation damage age that is explained by a complex damage accumulation and annealing history culminating in a mild heating event indicated by biotite Ar–Ar ages of about 1140 Ma. The positive results for the three case histories suggest that radiation damage ages could play a useful role in dating low temperature thermal events.

## 1. Introduction

The atomic-scale structure of zircon is gradually broken down due to the radioactive decay of trace amounts of U and Th (e.g. Pabst, 1952) resulting in progressive decreases in density, refractive index, birefringence and an expansion of the zircon unit cell (e.g. Holland and Gottfried, 1955). The possibility of using these properties to measure geological age was first suggested by Holland and Kulp (1950) followed by Kulp et al. (1952), Hurley and Fairbairn (1953), and more recently by Pidgeon et al. (1998).

In their classic paper, Holland and Gottfried (1955) demonstrated a systematic correlation between measured properties of density and unit cell, determined by XRD, and alpha dose determined from the measured U and Th contents of the zircons and the then known age of Sri Lankan zircons of 570 Ma. The authors envisaged that the observed systematic relationships could be used as a calibration curve for determining radiation damage ages. However, discrepancies between the unit cell measurements of the Sri Lankan zircon and zircons from other localities led Holland and Gottfried (1955) to conclude that “the Sri Lankan zircons have suffered slight thermal annealing since their formation”. The recognition that radiation damage in zircons could be annealed added a degree of complexity that had not been considered by the early pioneers and placed in doubt the validity of the calibration curve so comprehensively determined by Holland and Gottfried (1955). Further research has confirmed that radiation damage in zircon can be thermally annealed (e.g. Bursill and Mc Laren, 1966; Pidgeon et al., 1973; Meldrum et al., 1998; Geisler et al., 2001). However, in the 1950s the implications of radiation damage annealing for the determination and interpretation of radiation damage ages were not pursued as, with the invention of precise isotope dilution techniques for determining zircon U–Pb ages (Tilton et al., 1955; Silver and Deutsch, 1963), the idea of zircon radiation damage ages was essentially abandoned as a possible geochronological technique.

In contrast with the demise of radiation damage as a potential geochronological technique the formation of fission tracks in minerals by the natural fission of $^{238}$U, proposed as a dating technique in a pioneering paper by Fleischer et al. (1975), has proved to be extremely successful in its application to geological problems. Structural damage is produced by kinetic energy of fission fragments rather than alpha particles and nuclide recoil. The fission track dating technique (FT) has led to the development of specialist laboratories and has been successfully applied (e.g. Gallagher et al., 1998) to numerous problems using apatite, zircon, titanite and natural glasses (obsidian, pitchstone). Fission tracks are thermally unstable and where conditions exceed the fission track annealing temperature they will be partially or even totally annealed. The FT age is therefore a cooling age, giving the time when the mineral cooled below the temperature of track annealing. Apatite has been particularly successful in low-temperature thermochronology where it has been applied in numerous fields including tectonic modelling,

---

*E-mail address:* r.pidgeon@curtin.edu.au.

0009-2541/$ – see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.chemgeo.2013.12.010

landscape development, tectonic geomorphology, dating processes of mountain building, hydrocarbon exploration, sedimentary burial history and the low temperature history of the Yilgarn Craton (Kohn et al., 2002: Weber et al., 2005). Zircon fission track dating has also been applied to provenance studies, the thermal revolution of basins, low temperature metamorphism and exhumation studies (e.g. Bernet and Garver, 2005; Hurford, 1986). The successful application of fission track dating raises the possibility that zircon radiation damage ages might also have a useful role to play in geochronology and the purpose of this contribution is to re-examine the potential of the zircon radiation damage age technique, as applied in the modern geochronological environment, using three examples of zircons from rocks from the Archean Yilgarn Craton of Western Australia (Fig. 1).

## 2. Sri Lankan zircons and the calibration of the method

In the 1950s the degree of radiation damage was determined using X-ray diffraction measurements on milligramme samples of zircon powder (Holland and Kulp, 1950; Kulp et al., 1952; Holland and Gottfried, 1955). However, using secondary Ion Mass spectrometry (SIMS), isotopic analyses can now be made on 20 μm diameter spots on the polished surfaces of individual zircon crystals. Also, SIMS analyses have revealed complexities in the U–Pb systems and inhomogeneity in U and Th within zircon grains and micro-imagery, using cathodoluminescence and other electron microscopic techniques, has revealed complexities of zircon zoning and reactions on a micro scale. The early X-ray diffraction methods are clearly inadequate for investigating radiation damage on a micro scale and for radiation damage ages to be useful in the modern age a new technology was needed that could determine the degree of radiation damage of an area on a zircon smaller than a SIMS analytical spot. A solution to this problem was presented in a pioneering paper by Nasdala et al. (1995) who demonstrated that, using Raman spectroscopy, it was possible to make quantitative measurements of the radiation damage on 1 μm diameter areas on the polished surface of a zircon grain. They found that zircon vibrational bands progressively broaden, decrease in intensity and increase in wavelength with increasing radiation damage until, as the zircon becomes essentially amorphous, the zircon Raman bands disappear entirely. This new technology provided a means of making quantitative measurements of the degree of radiation damage on the same SIMS analytical areas used to determine the U–Pb age and the U and Th concentrations.

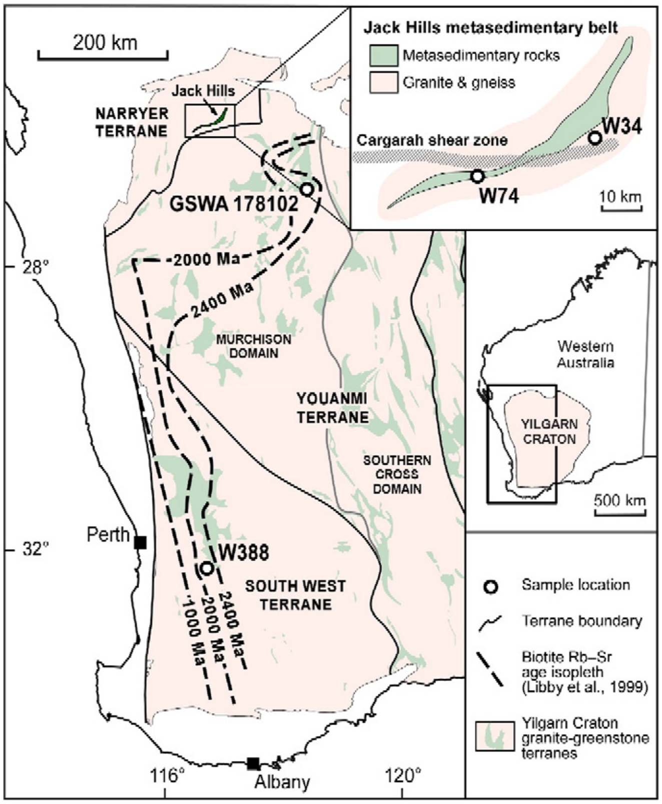

**Fig. 1.** Sketch map of the western Yilgarn Craton showing sample locations and the distribution of Rb–Sr biotite ages.

To determine radiation damage ages on small areas on zircon grains was now technically feasible but before this could be done the problem of calibrating the system had to be solved. To this end a population of zircons with a known age and spread in U and Th, and hence radiation damage, was needed that had not experienced any annealing of radiation damage over its geological history. The first question was how seriously annealed were the Sri Lankan zircons investigated by Holland and Gottfried (1955)? This question was answered by Raman measurements made by Nasdala et al. (1998) on zircons from Permian rhyolites from Saxony in Germany (referred to as the Saxonian zircons). Nasdala et al. (2001) showed that these zircons had significantly greater radiation damage for a given α-dose than Sri Lankan zircons confirming conclusions that the Sri Lankan zircons have been annealed (Nasdala et al., 2001, 2004). This relationship further suggested that the Saxonian zircons represented a zircon population that had retained all its radiation damage. This is supported by the geological setting and history of the rhyolite which would have cooled quickly after extrusion and remained at near surface temperatures since the Permian. The relationship between radiation damage (FWHM) and α-dose for the Saxonian zircons should, therefore, provide a suitable basis for the formulation of a calibration line for the determination of radiation damage ages. However, the maximum FWHM of grains determined to date of about 12 (Nasdala et al., 1998), is only sufficient for zircons with low degrees of radiation damage. Nasdala et al. (2001) extended this calibration to an FWHM of 25 $cm^{-1}$ by adding additional data points from lunar zircons determined by Wopenka et al. (1996). The assumption that the lunar data points of Wopenka et al. (1996) have never been annealed is not convincingly demonstrated, however Palenik et al. (2003) found that the Sri Lankan zircon data points fell on this calibration line when the α-dose of zircons was calculated with an age of 375 Ma. Using a calibration line determined from the Saxonian rhyolite zircon data alone the age becomes 380–400 Ma. Within the uncertainties, which are not specified, these ages are not significantly different and in the present contribution 375 Ma is adopted as the radiation damage age of the Sri Lankan zircons.

The 375 Ma age is significantly younger than the known age of the Sri Lankan zircons of ~560 Ma and was interpreted by Palenik et al. (2003) as indicating "that although the Sri Lankan zircons have an age of 560 Ma they have only accumulated radiation damage equivalent to an age of ~375 Ma suggesting that up to one third of the damaged volume has been annealed". The question of whether this age has geological significance needs to be assessed in the light of independent geological evidence. However, on a practical level, the coincidence of Sri Lankan data points with α-doses recalculated with an age of 375 Ma provides a basis for formulating a model calibration line to cover the full range of radiation damage in zircons. From previous work (e.g. Holland and Gottfried, 1955; Weber, 1990; Palenik et al., 2003) it is known that radiation damage increases exponentially with a-dose and we have used the model curve described by Palenik et al. (2003), based on fitting an exponential curve of the form given by Weber (1990) to α-doses of Sri Lankan zircons normalized to an age of 375 Ma, as a calibration curve for determining radiation damage ages, where the radiation damage age is the time needed to produce the observed radiation damage This equation (Eq. (4) of Palenik et al., 2003) is as follows —

$$FWHM = A\left[1-exp^{(-B_{FWHM}D_{ed})}\right] \qquad (1)$$