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Geocronologia por traço de fissão em apatitas de rochas alcalinas: Comparação dos diferentes métodos de datação e calibração da dosimetria de nêutrons.

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Apresentação

Esta tese é apresentada em forma de artigo, nos quais compõem seus capítulos. Em linhas gerais estão sendo abordadas discussões a respeito da metodologia de traço de fissão com o intuito de entender o comportamento dos traços frente a procedimentos experimentais. Para isso amostras de rochas alcalinas e apatitas de Durango foram utilizadas. Em principio, acredita-se que estas rochas possuem uma evolução geológica relativamente simples e com poucas variáveis termotectônicas que possam complicar a interpretação dos dados. Abaixo são apresentados brevemente os capítulos que compõem esta tese.

O primeiro capítulo intitulado: "FURTHER INVESTIGATION OF THE GEOMETRY FACTOR IN APATITE FISSION-TRACK THERMOCHRONOLOGY" tem como objetivo discutir fazer comparações de idades através do Método das Populações, MP com o Método do Detector Externo, MDE, utilizando como dosimetria, vidros padrões calibrados com filmes finos de urânio natural. Como a datação pelo MDE necessita de uma correção na densidade induzida, novos valores do fator geométrico foram determinados, através de medidas das densidades induzidas na superfície interna da apatita e na mica detector-externo, ρ_{ED}/ρ_{IS} e medidas dos traços projetados. É também apresentada uma nova forma de se obter o fator geométrico, usando medidas de traços induzidos obtidas pelo MP e EDM. Dado que, para datação pelo MDE não é possível que se tenha o comprimento inicial dos traços induzidos, L_0, é comum a utilização de um valor arbitrário. É mostrado, portanto, uma variação significativa deste parâmetro bem como a influência e a necessidade de se ter um valor de L_0 da própria amostra quando histórias térmicas são geradas. O segundo capítulo intitulado "DETERMINATION OF FISSION-TRACK AND RETENTION AGE THROUGH *PLATEAU* AND SIZE CORRECTION METHOD" verificar a validade e a necessidade de uma correção de idade. Para isso, dois métodos foram aplicados: a curva de correção pelo tamanho dos traços e a correção pelo método de *plateau*. Para isso, amostras de apatitas de rochas alcalinas foram utilizadas. Neste caso, como as idades destes corpos são bem conhecidos através de outros métodos radiométricos, foi possível fazer uma análise mais crítica sobre os resultados. No caso da técnica de *plateau*, apenas datação onde se considera uma população de idades pode ser aplicada.

Considerando que para a datação pelo MP um pré-aquecimento é necessário, foram também realizadas datações utilizando o Método da Subtração, MS, no qual a amostra não passa por nenhum aquecimento prévio antes de ser irradiado por nêutrons térmicos. Desta forma foi possível verificar se esse pré-aquecimento influencia no comportamento dos traços frente a novos aquecimentos. Discussão a respeito da necessidade de correção e o significado de idade de traço de fissão e idade de retenção dos traços estão bem documentados. Por fim, o terceiro capítulo, intitulado: "RECALIBRATION OF DOSIMETER GLASS THROUGH NATURAL URANIUM THIN FILM" tem como objetivo a re-calibração da dosimetria de nêutrons usando filmes finos de urânio. Resultado de idades de rochas alcalinas e apatita de Durango foram comparados com métodos tradicionais de ativação metálica usando dois reatores nucleares com características diferentes frente à termalização de nêutrons. Ambas as metodologias apresentam resultados compatíveis entre si e com outros métodos de datação radiométrica.

Introdução

Em Julho de 1971 na conferência intitulada *"Calibration of Hominoid Evolution"* na Austrália, Fleischer e Hart, pioneiros do MTF, sugeriram uma serie de cautela na utilização da dosimetria absoluta (determinação direta da fluência de nêutrons), juntamente com a utilização da constante de fissão espontânea do ^{238}U, λ_f. Em geral, o Método das Populações, MP, foi a única técnica aceita para datação utilizando a dosimetria absoluta. Isso porque as eficiências de revelação e observação em ambas as alíquotas (traços espontâneos e induzidos) são, em princípio, iguais. Para o caso do Método do Detector Externo, MED, a restrição foi ainda maior. Isso porque um fator geométrico que engloba eficiências na detecção, revelação e observação na superfície interna da apatita e no detector externo, são intrinsecamente diferentes. Uma alternativa mais pragmática e plausível foi sugerida, que é a medida do fator zeta (Hurford e Green, 1982; 1983). Esta medida envolve a calibração de uma posição específica do reator nuclear juntamente com vidros padrões dopados com urânio e uma amostra que seja padrão de idade (datada com outro método de datação). Não houve restrições para o uso dessa metodologia. Com os

avanços nas medidas das constantes, e valores aceitos pela "*International Union of Pure and Applied Chemistry*", alguns pesquisadores vêm apontando a necessidade de uma dosimetria que torne o MTF independente.

É conhecido que amostras que sofreram algum grau de *annealing* possuem traços espontâneos menores do que traços induzidos, isentos desse efeito. No entanto, a necessidade de correção de idade quando amostras apresentam graus de *annealing* baixo (por exemplo, apatita de Durango) têm sido bastante discutida. Esta característica é também observada em amostras pertencentes a rochas alcalinas, cuja historia térmica é bastante simples. Em geral, duas técnicas que levam em consideração a perda de eficiência na contagem dos traços espontâneos são: correção pelo tamanho dos traços e correção pelo método de *plateau*. A importância em se definir a necessidade de correção de idades (e conseqüentemente a correção da densidade espontânea) considerando amostras com baixo grau de *annealing* está no fato de amostras como a apatita de Durango ser rotineiramente empregada quando se usa o parâmetro zeta. Desta forma, se há necessidade de correção de idade para essa amostra, deve-se considerar também para a obtenção da calibração zeta. Outro ponto importante é que, aquecimentos laboratoriais, no qual são feitos para entender a cinética de *annealing*, apontam uma diminuição da densidade em qualquer taxa de *annealing*. A relação entre a diminuição da densidade e comprimento é de 1:1 para graus de *annealing* de ≈30-35%. Isto significa que, se uma amostra com baixo grau de *annealing* não precisa necessariamente de correção, a diminuição dos traços em tempo geológico pode não se comportar exatamente igual a diminuição para aquecimentos laboratoriais. Este efeito implica em uma correção para obter a modelagem de história térmica.

Estes pontos apresentados acima compõem o assunto deste presente trabalho. Estão sendo apresentados três artigos no qual abordam estes aspectos.

Com o objetivo de re-investigar as medidas do fator geométrico para aplicação do MDE, foram realizadas medidas das densidades induzidas na superfície interna das apatitas e nas micas detector-externo. Também foi possível obter novas medidas do fator geométrico fazendo análises dos traços projetados, sendo possível obter o valor de GQR, onde G(=2π/4π = 1/2), $Q = [\eta q]_M/[\eta q]_A$ é a razão entre as eficiências de observação (η) e revelação (q) para os traços na mica detector-externo (sub-índice "M") e na superfície interna da apatita (sub-índice "A).

Um aspecto na aplicação do MDE é que o valor do comprimento inicial dos traços de fissão, L_0, não é rotineiramente analisado. Este valor é um parâmetro importante para quantificar com boa confiança o grau de *annealing* dos traços espontâneos em uma amostra não conhecida, sendo essencial para se obter uma história térmica acurada. O impacto usando um valor arbitrário de L_0 foi investigado e discutido. Este valor pode ser obtido irradiando uma porção extra pré-aquecida (*annealing* total). Como ganho secundário, a densidade induzida no detector externo e na apatita utilizado para obter o valor de L_0 pode dar um fator g para cada amostra a ser datada. No total, oito amostras de apatitas de embasamento cristalino foram usadas para determinar o fator g. Os valores de L_0 medidos nestas oito amostras variam entre 14.9 e 16.0μm. Implicações na calibração zeta são discutidas.

Como dito acima, com a utilização da dosimetria absoluta, o efeito do *annealing* aparece nas idades de traço de fissão. Desta forma, a idade obtida é uma idade aparente, uma idade menor que a idade "verdadeira" da amostra. Para considerar esse efeito, duas técnicas de correção podem ser aplicadas, a correção pelo tamanho dos traços, Storzer e Wagner (1969) e a correção pelo método de *plateau*, Storzer e Popeau (1973).

É apresentada a potencialidade das técnicas de correção supracitadas utilizando apatitas de rochas alcalinas pertencentes ao Arco do Alto Paranaíba e Arco de Ponta grossa e amostras de apatita de Durango. As medidas foram feitas considerando o MP.

Visto que para a datação pelo MP um pré-aquecimento é necessário, também foram realizadas datações utilizando o Método da Subtração, MS, no qual a amostra não passa por nenhum aquecimento prévio antes de ser irradiada por nêutrons térmicos. Esta comparação tem como objetivo verificar se este pré-aquecimento utilizado para datação pelo MP influência a correção pelo método de *plateau*. Esses dados foram também comparados com idades destas apatitas presentes na literatura, obtidas através de outras técnicas de datação radiométrica.

Por fim, é apresentada a re-calibração da dosimetria de nêutrons através de filmes finos e urânio natural. Esta calibração foi primeiramente apresentado por Iunes *et al.* (2002). Para isso, amostras de rochas alcalinas e Durango apatita foram datadas e os resultados foram comparados com a tradicional ativação metálica. Essa nova calibração se mostrou adequada e ambas as dosimetrias são concordantes entre si. Também foi utilizadas

amostras padrões de idade (Durango) para se obter idades de algumas de rochas alcalinas e novamente os resultados são concordantes. Finalmente, estes resultados estão estatisticamente compatíveis com dados gerados por outros métodos radiometricos, no qual possui uma temperatura de fechamento acima do traço de fissão. Isto indica que amostras com baixo grau de *annealing* (≈11%) as idades de traço de fissão não precisa ser corrigida, resultado este que corrobora com dados apresentados fazendo medidas de *plateau*. Histórias térmicas dos arcos do Alto Paranaíba e Ponta Grossa são apresentadas e discutidas.

Capítulo 1

FURTHER INVESTIGATION OF THE GEOMETRY FACTOR IN APATITE FISSION-TRACK THERMOCHRONOLOGY

Abstract. External Detector Method is a widely used technique in Fission Track Thermochronology, FTT, in which two different minerals are concomitantly employed: spontaneous tracks are observed in apatite and the induced ones in the external detector muscovite. They show intrinsic differences in detection and etching properties that should be taken into account. In this work, new geometry factor values, g, in apatite were obtained by ρ_{ED}/ρ_{IS} ratio and GQR values through the measurement of projected lengths. Five mounts, two of which were large area prismatic sections (Mn-1 and Mn-2, Durango apatite) and three samples composed of random orientation pieces (D-2 and D-3, Durango apatite and TF-42, from crystalline rock) have been used to determine the g-values. A side effect of applying EDM is that the value of the initial confined induced fission track, L_0, is not measured in routine analyses. The L_0-value is an important parameter to quantify with good confidence the degree of annealing of the fossil fission tracks in the unknown age sample, being essential for accurate thermal history modeling. The impact of using arbitrary L_0-values on the inference sample thermal history was investigated and discussed. The L_0-value can be measured for each sample as an extension of the ρ_{ED}/ρ_{IS} ratio method for measuring g, which consists in simultaneously irradiating pre-annealed apatite grains and the EDM mount of the same sample. The ratio between densities of induced tracks in the muscovite and in the extra apatite mount gives the value of g. L_0 is the mean length of the confined fission tracks measured in the extra apatite mount. Eight apatite samples from crystalline basement, with grains at random orientation, were used to determine the g-values. The resultant average found (0.56±0.02) is statistically in agreement with the values found for Durango samples cut in prismatic section (0.55±0.02) and for Durango samples crushed and mount at random orientation (0.58±0.01). There was no observable variation in efficiency regarding crystal orientation, showing that it is relatively safe using non prismatic grains, especially in samples with paucity of grains, as it is the case of most basin samples. Values of L_0 measured for these same eight samples varied between 14.9 and 16.0 μm. Implications for the ζ-calibration of fission-track ages are also discussed.

Keywords: Fission-track dating, Geometry factor, ϕ-method, ζ-calibration, Initial fission-track length.

1. Introduction

Apatite, $Ca_5(PO_4)(F,Cl,OH)$, is a mineral present in many types of rocks. It can be found in different types of geological environments as basement and detrital samples and contain a relatively high uranium concentration, between 1 to 100µg/g, being a very suitable mineral for Fission-Track Thermochronology (FTT) application (Wagner and Van den Haute, 1992).

Fission-track dating is based on the spontaneous fission decay of ^{238}U, which releases a great amount of energy that is, in part, stored as a damaged channel along the fission fragment trajectory. This damaged zone is called latent track. Spontaneous latent fission tracks are accumulated inside the crystal lattice at geological time scale. In this way, the fission-track age can be found using the spontaneous fission decay constant, λ_f, and determining the number of uranium atoms present in the mineral, N_U, the surface density of spontaneous fission-tracks crossing an internal surface and the efficiency factor, ε_{238}, that embraces etching, observation and geometry factors.

Normally, the number of uranium atoms is determined through counting of induced fission tracks generated after the interaction of ^{235}U nuclei with thermal neutrons, which can be observed in another aliquot of the same apatite sample (Population Method, PM) or in an external detector (usually muscovite) coupled with the mount containing the fossil tracks during neutron irradiation (External Detector Method, EDM). In this case, induced tracks in muscovite are located at the mirror image position of the grain from which the fission fragments were ejected.

EDM is the most used procedure because it allows for single-grain dating even in samples with non-uniform uranium content. This characteristic is important mainly for dating two or more age population samples, as the case of basin samples analyses. Thus, it is possible to distinguish age populations, which can lead to the identification of source areas for sediments in the basin (Dias et al., 2011).

An important parameter for EDM dating is the geometry factor, g, which accounts for the differences in observation efficiency between apatite and muscovite. Although the g-value is crucial for fission-track dating, there are only a limited number of studies on it (Gleadow and Lovering, 1977; Green and Durrani, 1978; Iwano and Danhara, 1998;

Jonckheere and Van den haute, 2002; Jonckheere, 2003; Enkelmann and Jonckheere, 2003).

It has been recommended that only clear, c-axis parallel (prismatic) grains of apatite should be selected for fission-track dating (e.g., Gleadow et al., 2002). However, under certain conditions, in which there is paucity of apatite grains, it is common practice to count tracks also in non-prismatic section grains, while the g-value is measured in closely ideal large area single crystals (referred to as extensive surfaces through the rest of this work). Grain surfaces at different orientation present different etching characteristics (Jonckheere and Van den haute, 1996), which may lead the observer to vary his/her efficiency for counting tracks, in such a way that the value of g measured in prismatic section crystals may not reflect the actual observer counting efficiency.

Thus, it is shown in this work a further investigation on the geometry factor used to calibrate EDM ages, including samples presenting non ideal conditions. The geometry factor has been determined using two different methods. First, the geometry factor was measured through projected length distributions, applying the method proposed by Jonckheere (2003) being possible to obtain GQR (= g) value. $G = (2\pi/4\pi) = 1/2$, $Q = (\eta q)_M/(\eta q)_A$ is the ratio between observation (η) and etching (q) efficiency factors for tracks in muscovite external detector (sub-index "M") and in apatite internal surface (sub-index "A"). R is range factor that take into account the range of the fission fragment within mineral bulk. (Jonckheere, 2003). The second method employed consists in the measurement of induced track densities in an apatite internal surface, $\rho_{IS,}$ and muscovite external-detector, $\rho_{ED.}$ The ρ_{ED}/ρ_{IS} ratio directly gives the value of the geometry factor. For these two determinations, Durango apatite crystals were cut to expose extensive prismatic surfaces (Mn-1 and Mn-2) and two mounts, composed by pieces of crushed Durango apatite at random orientations (D-2 and D-3), were assembled. In addition, one apatite sample from a crystalline rock from Ilha Bela, São Paulo State, Brazil (TF-42) was also used. These two methods have been developed and applied previously. (Iwano and Danhara, 1998; Jonckheere and Van den haute, 2002; Jonckheere, 2003; Enkelmann and Jonckheere, 2003). Thus, these methods were applied in grains at random orientations and compared with the usual practice of measuring g on prismatic or basal extensive sections.

As a side effect, the application of EDM precludes the determination of mean induced confined fission-track length in apatite, which is used as proxy for the mean length of the unannealed confined fission track and is normally referred to as initial confined fission-track length, L_0. Thus, L_0 is the reference to quantify the degree of annealing of a fission track, making it a key parameter for thermal history modeling.

In general, for modeling purposes, the fission-track researchers apply a standard L_0-value of 16.30 μm, the mean of all values for Durango apatite reported by Green et al. (1986) and close to that reported by Carlson et al. (1999) or, if the sample is chemically characterized, adopt the value of L_0 for the sample with the closest chemical composition among those tabulated by Carlson et al. (1999) or Barbarand et al. (2003). Even if the sample composition matches exactly the one of a tabulated sample this procedure is risky. Variation in experimental procedure, microscopy equipment and particular analysis criteria can contribute for the variation in the lengths of the confined tracks measured by different analysts even working in the same laboratory. Recent published work (Ketcham et al., 2009) shows significant variation in L_0-value measured for a grain mount with Durango apatite samples measured by a group of analysts during the the 2007 IGCP workshop on fission-track thermochronology in Pisa, Italy. Other works (Tello et al., 2006) also present this variation in L_0-value for Durango apatite. Barbarand et al. (2003) present a study about the reproducibility with well controlled laboratorial conditions, in which it was found significant variation within analysts of the same research group and for a single analyst over time. In conditions where the degree of annealing become higher (the fission tracks are more shortened), the reproducibility becomes significantly worst.

The discussion above as well as discussions in Barbarand et al., 2003; Ketcham, 2005; Ketcham et al., 2009, makes explicit the necessity of an adequate procedure for obtaining the thermal history modeling. The ideal situation is to obtain the L_0-value in the same laboratory and analysis criteria in which the spontaneous fission-tracks were measured. Thus, a laborious but reliable alternative is to obtain the L_0-value directly in the apatite sample to be dated. For this, it is proposed in this work that a pre-annealed aliquot of the same sample should be irradiated together with the EDM mount. Thus, the sample used to dating and thermal history modeling is also used to obtain the L_0-value. Thus, even L_0 depending on the apatite chemical composition and lattice structure (Carlson et al.,

1999; Barbarand et al., 2003), the direct measurement of this parameter for each sample makes thermal history modeling more accurate (Donelick et al., 2005; Ketcham et al., 2009).

To illustrate the proposed method, values of L_0 are measured in eight apatite samples from the crystalline rocks from Mantiqueira Mountain Range, Mar Mountain Range and adjacent areas of southeast, Brazil, (Tello et al., 2005). It is also shown the influence of the L_0-value in the thermal history modeling with a practical example. The same apatite sample has its thermal history modeled with values of L_0 in the range of values found in literature and in this work (14.9 to 16.7 μm).

As a secondary gain of preparing an extra apatite mount for measuring L_0, it is possible to obtain a value of the geometry factor measuring also de induced track density in this extra sample and using the ρ_{ED}/ρ_{IS} ratio method described above. Thus, it was possible to measure g and L_0 directly in the samples being dated.

The procedures we propose in this work make FTT routine more laborious. However, they contribute to increase the accuracy of data analysis, mainly for thermal history modeling. For dating, determination of g, these procedures are justifiable in cases where the observation efficiency is considerably different between the sample used to obtain g-value (e.g. standard Durango apatite) and the unknown age sample. For example, basin samples contain commonly different apatite populations that might merit a more rigorous investigation of their etching, annealing and geometry properties.

The g-value has been regarded as a necessity only for the application of the ϕ-method for the calibration of the neutron dosimetry, i.e., the dating method in which neutron fluency is independently measured by neutron monitors (Jonckheere, 2003). However, as it will be shown below, the ages obtained by ζ-calibration may also be affected because the standard and the unknown age samples do not necessarily have their tracks counted with the same efficiency. Particularly, the automatic counting of fission tracks that has been recently proposed (Gleadow et al., 2009) would benefit much by the measurement of g-value for the standard and for the unknown age sample. In this case, the surface characteristics of each sample have a more significant effect on counting efficiency than in the traditional counting.

2. Method

2.1. Fission-track age equation

Fission-track ages can be calculated, by the ϕ-method, using the following equation (Bigazzi et al., 1999; Iunes et al., 2002):

$$t = \frac{1}{\lambda}\ln\left[1 + g\frac{(\rho_S/\rho_I)}{(\varepsilon^{238}/\varepsilon_I)}\left(\frac{\lambda}{\lambda_f}\right)\left(\frac{R_U}{C_{238}}\right)\right] \quad (1)$$

In Eq. (1), λ is ^{238}U alpha decay constant; λ_f is the ^{238}U spontaneous fission decay constant; ρ_S (ρ_I) is the spontaneous (induced) density; C_{238} is the ^{238}U isotopic concentration; $\varepsilon^{238}(\varepsilon_I)$ is the ratio between the number of observed spontaneous (induced) fission track of ^{238}U (^{235}U) per unit surface and the spontaneous (induced) fission for unit volume in the mineral, R_U is the number of fissions by target nuclei which can be determined by measuring the muscovite attached to standard glass, calibrated against natural uranium thin film (Iunes et al., 2002) and g is the geometry factor.

Several experimental procedures have been developed along the years for the calibration of the FT age equation, (Fleischer et al., 1975; Naeser et al., 1979; Greadow and Duddy, 1981; Hurford and Green, 1982; Storzer and Wagner, 1982). These procedures can be distinguished from each other mainly in the way the induced fission tracks are analyzed. The two most applied procedures are: Population Method, PM, and External Detector Method, EDM

In PM, both spontaneous and induced fission tracks are analyzed in different aliquots of the same apatite separate, which are supposed to have uniform uranium concentration and chemical composition. It is also assumed that only one age population is present. Spontaneous and induced fission-track densities are determined as averaged values over the spontaneous fission track (non-irradiated aliquot) and induced fission track (irradiated aliquots) populations, respectively. In addition, spontaneous and induced track densities are determined in internal surfaces of the grains (under 4π geometry) and the

registration, etching and observation factors cancel out. The geometry factor, defined as the ratio between the measurement geometries of spontaneous and induced tracks equals 1. Thus, PM should only be applied to apatite samples belonging to embasement rocks. In addition, , for the application of PM, a large number of analyzed grains are needed; PM is a multi-grain methodology.

For EDM, the spontaneous fission-track densities are measured in grains of the host (apatite) mineral (under 4π geometry), while induced fission-track densities are measured in an external detector (under 2π geometry), attached to the mineral mount during irradiation. It is possible to identify each grain with its mirror image on the muscovite external-detector and the spontaneous and induced surface densities are determined by measuring tracks from the same grain. Using EDM allows for the individual grain dating . Thus, EDM can be applied, even for apatites belonging to sedimentary rocks. The IUGS (International Union of Geological Sciences) recommended that an absolute (ϕ-method) calibration with the accepted λ_f value could be used to date apatite together with PM analysis, whereas ζ-calibration is recommended in all fission track techniques (including PM). (Hurford, 1990; Iwano and Danhara, 1998). This restriction is because in EDM the differences in efficiency between apatite and muscovite must be accounted for through the measurement of a geometry factor if the ϕ-method is applied. (Naeser, 1967; Hurford, 1990; Iwano and Danhara, 1998).

Measurements of λ_f value, carried out by the fission-track method were based on neutron dosimetry and often through the comparison of resulting ages with radiometric ages. It turns out that the obtained values, 7.0×10^{-17} a^{-1}, were in disagreement with the values obtained by other methods, around 8.5×10^{-17} a^{-1} (see discussion in Hadler et al., 2003). Thus, instead of a fundamental constant, λ_f, was treated as an irradiation parameter along with neutron fluence. However, in the last two decades the research in neutron dosimetry has significantly evolved (De Corte et al., 1988; Suzuki and Tomura, 1988; Van den Haute et al., 1988; Tagami and Nishimura, 1989; Bigazzi et al., 1989, 1993, 1995; Falleiros et al., 1994; Iunes et al., 2002, 2004, 2005) and several fission-track measurements of λ_f have been carried out involving neutron dosimetry (Hadler et al., 1981; Guedes et al., 2000, 2003a; Yoshioka et al., 2005) and without neutron dosimetry (Guedes et al., 2003b). All these determinations resulted in values around 8.5×10^{-17} a^{-1}. This

extensive body of methodological work assures the reliability of the ϕ-method for the neutron dosimetry calibration and its utilization in the EDM technique as it has already been done (Enkelmann et al., 2005; Curvo et al., 2007; Franco-Magalhães et al., 2010; Hiruma et al., 2010; Dias et al., 2011). Unlike PM, in which both spontaneous and induced tracks are measured under the same conditions, analyses with EDM are performed in minerals with different detection thresholds, chemical etching and observation and geometric efficiencies. In this way, the efficiencies do not cancel out and the geometry factor that ideally equals 0.5 has to be determined from separate experiments. The values found in literature can reach 0.61 (Enkelmann and Jonckheere, 2003).

2.2. Measurement of the geometry factor through projected fission-track length distributions

As described above, when EDM is applied, spontaneous fission-track densities are measured in apatite grains (under 4π geometry), whereas induced ones are measured in muscovite external-detector (under 2π geometry). Thus, the geometry correction is G=(2π/4π)=1/2. However, for EDM ages, both spontaneous and induced fission-track densities are measured in different detectors, so differences in the efficiency of revelation by etching and observation under microscopy should be considered. This parameter is the ηq value (Jonckheere and Van den haute, 1999).

The ηq-value in both an apatite internal surface and a muscovite external detector is determined by measuring induced fission tracks. For this, prior to irradiation, spontaneous fission tracks are totally annealed in the apatite sample. The apatite grains are mounted in epoxy resin, polished and then attached to the muscovite external detector before irradiation. After irradiation, the assembly of apatite is polished again until the 4π geometry is attained, i.e., the layer removed is thicker than the fission-fragment range. Projected fission-track lengths, which are the distances, parallel to the observation surface, between the pits and the ends of the tracks, are measured. For the determination of ηq, every track that matches the observer criteria for counting should have its projected length measured. The ideal projected length distribution is (Jonckheere and Van den haute, 1999):

$$N(p)dp = N\left(1 - \frac{p}{L_I}\right)dp \qquad (2)$$

In Eq. (2), p is the projected length, which crosses the analyzed surface and L_I is the fully etched length of the combined range of the fission fragments, which are propelled in opposite directions. In Figure 1, examples of projected length distributions from apatite external and internal surface and muscovite external-detector obtained in this work are shown. Note that the shorter projected lengths are less abundant than expected by the ideal triangular distribution. For this method of determining ηq, it is assumed that all the loss in efficiency occurs for the shorter projected lengths. Longer tracks are then used to fit the theoretical distribution. The value of ηq is the ratio between the areas of the measured and ideal distributions. The same procedure is applied for internal and external surfaces of apatite and the external detector.

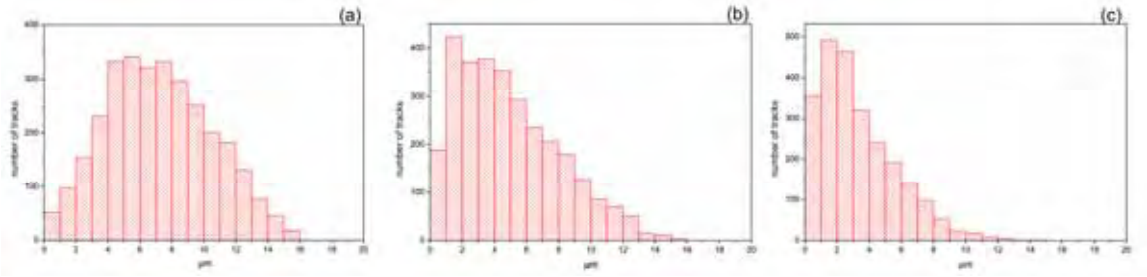

Figure 1. Projected length distributions for Durango apatite (a) an external and (b) internal surface and (c) external detector, respectively.

The etchable range of a track is not the same in apatite and in muscovite. A given track may be observed only in apatite or muscovite external-detector, in both or neither (Iwano et al., 1992, 1993; Jonckheere, 1995, 2003). Therefore, it is required the introduction of a range deficit factor value (R). The R-value is calculated by the relationship between latent (L_{lM}-muscovite, L_{lA}-apatite) and etched (L_{eM}-muscovite, L_{eA}-apatite) track lengths: $R=(L_{eM}/L_{eA})/(L_{lA}/L_{lM})^{-1}$ (see discussion in Jonckheere, 2003). The geometry factor is, therefore, given by GQR, the product of $G(=2\pi/4\pi = 1/2)$, $Q = (\eta q)_M/(\eta q)_A$ and the range deficit factor value, R.

2.3. Measurement of the geometry factor through induced fission-track densities

Another way to obtain the geometry factor is the direct measurement of induced fission-track densities in both apatite internal surfaces and muscovite external detector surfaces. The fission-track densities were determined counting all projected tracks which were measured to obtain the GQR value. The value of the geometry factor is the ratio between the induced fission track densities in the apatite internal surface and the muscovite external detector.

In this work, we propose an extension of this procedure, which consists of simultaneously dating unknown age samples by EDM and PM. The value of the geometry factor is found by imposing that both dating protocols give the same ages. Rewriting the Eq. (1) for PM and EDM dating, respectively then yield:

$$T_{PM} = \frac{1}{\lambda}\ln\left[1 + g_{4\pi}\frac{(\rho_S/\rho_I)_{PM}}{(\varepsilon^{238}/\varepsilon_I)_{PM}}\left(\frac{\lambda}{\lambda_F}\right)\left(\frac{R_U}{C_{238}}\right)\right] \quad (3)$$

$$T_{EDM} = \frac{1}{\lambda}\ln\left[1 + g_{2\pi}\frac{(\rho_S/\rho_I)_{EDM}}{(\varepsilon^{238}/\varepsilon_I)_{EDM}}\left(\frac{\lambda}{\lambda_F}\right)\left(\frac{R_U}{C_{238}}\right)\right] \quad (4)$$

For simultaneously dating by EDM and PM, besides having to prepare the apatite/external detector mount, an aliquot an aliquot or mount of pre-annealed apatite grains must be simultaneously irradiated with thermal neutrons. In this way, the constants (λ, λ_F and C_{238}) and irradiation parameters (R_U) are the same in Eqs. (3) and (4) and the ratio of these two equations results in

$$\frac{\exp^{T_{PM}} - 1}{\exp^{T_{EDM}} - 1} = \frac{g_{4\pi}}{g_{2\pi}}\left[\frac{(\rho_S/\rho_I)_{PM}}{(\rho_S/\rho_I)_{EDM}}\left(\frac{(\varepsilon^{238}/\varepsilon_I)_{EDM}}{(\varepsilon^{238}/\varepsilon_I)_{PM}}\right)\right] \quad (5)$$

Then, imposing the same fission-track age, it follows from Eq. (5) that:

$$\frac{g_{2\pi}}{g_{4\pi}}\frac{\left(\varepsilon^{238}/\varepsilon_I\right)_{PM}}{\left(\varepsilon^{238}/\varepsilon_I\right)_{EDM}} = \frac{\left(\rho_S/\rho_I\right)_{PM}}{\left(\rho_S/\rho_I\right)_{EDM}} \quad (6)$$

The reason why the geometry factor (ideally $g_{2\pi}/g_{4\pi}$) assumes values different from ½ is that the ratio of efficiency factors does not cancel out, since etching and counting efficiencies are different in PM (induced tracks in apatite) and EDM (induced tracks in muscovite). For practical applications, the geometry factor must incorporate this effect. Thus,

$$g = \frac{g_{2\pi}}{g_{4\pi}} \frac{\left(\varepsilon^{238}/\varepsilon_I\right)_{PM}}{\left(\varepsilon^{238}/\varepsilon_I\right)_{EDM}} \Rightarrow g = \frac{\left(\rho_S/\rho_I\right)_{PM}}{\left(\rho_S/\rho_I\right)_{EDM}} \quad (7)$$

The efficiency ratio is the product between $Q = (\eta q)_M/(\eta q)_A$ and the range deficit factor value, R.

Considering uniformity in the uranium/surface track distribution, there is no reason why spontaneous densities should be different. Actually, the same apatite mount can be used to measure the spontaneous fission density in both dating protocols. Thus,

$$g = \frac{\left(\rho_I\right)_{EDM}}{\left(\rho_I\right)_{PM}} = \frac{\rho_{ED}}{\rho_{IS}} \quad (8)$$

Eq. (8) is therefore, the same used when g-value is determined through induced fission track densities in both apatite internal surface and muscovite external detector. However, in this case, regular unknown age samples can be used to measure the g factor provided an extra aliquot or mount of apatite grains is irradiated along with the sample to be dated. Ideally, this procedure would make possible to measure one individual g-value for each sample.

3. Experimental procedures

Durango apatite crystals were cut to expose extensive prismatic surfaces (Mn-1 and Mn-2) and two mounts, composed of apatite grains at random orientations (D-2 and D-3), have been assembled for this experiment. Samples from crystalline basement (TF-5, TF-9, TF-10, TF-12, TF-17, TF-21, TF-30, TF-32 and TF-42), collected in the State of São Paulo, Brazil (Tello et al., 2005), were also used. Samples used to obtain the geometry factor were heated at 450°C for 24 hours. This heating is sufficient to erase the spontaneous fission

tracks (total annealing). Samples were grinded and polished, attached to muscovite (external detector) and irradiated with thermal neutrons to induce ^{235}U fission. The ^{235}U induced-fission fragments that escape the mineral through the surface can be detected by the external detector, forming a mirror image of the grain contained in the mount. To obtain an internal surface, the irradiated apatite samples were grinded and polished again. This was necessary because the surface attached to muscovite collected tracks only from below (2π geometry). The surface obtained after irradiation and polishing collected tracks from below and from above (4π geometry). These samples were used for the measurement of the geometry factor through the ratio between densities, ρ_{ED}/ρ_{IS}, and GQR measurement through projected length distribution.

Samples from crystalline rocks were divided in three aliquots: two for PM and one for EDM dating. For PM, an aliquot was used for the determination of the spontaneous-track density and the other was pre-annealed in the same way as described above and irradiated with thermal neutrons to induce ^{235}U fission. Once spontaneous and induced track densities are determined, both in apatite, PM fission-track ages were calculated.

For EDM, samples were mounted, spontaneous fission-tracks were revealed by etching, and the mounts were attached to muscovite plates (external detector) for thermal neutron irradiation. Once spontaneous and induced densities in apatite and muscovite external-detector, respectively, are determined, EDM fission-track ages are calculated as functions of the geometry factor. Combining the induced densities obtained in both dating procedures, geometry factor was determined (Eq. 8).

Samples were irradiated at the IEA-R1 reactor (IPEN/CNEN, São Paulo, Brazil) using standard glasses (CN5), calibrated through natural uranium thin films, as dosimeters (Bigazzi et al., 2000; Iunes et al., 2002, 2004, 2005). The neutron fluence was measured using the calibration by natural uranium thin film yield the value of $\phi=2{,}2\text{x}10^{15}$neutrons/cm^2. One irradiation was done using a wrapper of cadmium and no significant fission track content was observed, indicating that no significant influence of epithermal/fast neutrons was present.

All apatite samples analyzed in this work were mounted in epoxy resin and had the same sanding (with sandpaper of #1200, 2400 and 4000), polishing (with diamond paste of

1 and ¼ μm) and etching. For apatite, the etching was carried out in a 5% NHO_3 solution, at 20 ºC, for 55 seconds (Tello et al., 2003, 2005, 2006). Muscovites were etched in a 48% HF solution, at 15 ºC, for 90 minutes. All etching procedures were carried out in an ultrathermostatic bath SP Labour model SP-152/30. Variation in temperature was ≈0.1°C.

Density and projected length measurements were carried out using a Zeiss Axioplan 2 Imaging microscope, with nominal magnification of 1000×, dry.

4. Results and Discussion

4.1. Geometry factor

Geometry factor data using projected length distributions were measured and the values of ηq and GQR were determined (Table 1).

The average of GQR values (last column) for samples containing grains at random orientation, D-2 and D-3, is 0.59±0.01. For extensive surface samples, Mn-1 and Mn-2, the average is 0.60±0.01. For crystalline embasement sample, TF-42, the geometry factor value is 0.56±0.03. Considering a greater error for sample TF-42, GQR values are statistically compatible.

For these same samples, density measurements are also shown in Table 1. The values of g obtained by the ratio between induced fission tracks in muscovite (external detector) and in apatite (internal surface) are shown in the sixth column. The average value for g between random-oriented grain samples D-2 and D-3 is 0.58±0.01. For extensive surface samples, Mn-1 and Mn-2, the average is 0.55±0.02. For crystalline embasement sample, TF-42, the value is 0.53±0.02, in agreement with the average of Mn-1 and Mn-2. Also, measurements in Durango apatite samples are statistically in agreement.

Table 1. Values of fission track densities and measured projected track.

N_{ES} (f)	N_{IS} (f)	$N_{ED(ES)}$ (f)	$N_{ED(IS)}$ (f)	ρ_{ED}/ρ_{ES} (±σ)	ρ_{ED}/ρ_{IS} (±σ)	ρ_{IS}/ρ_{ES} (±σ)	$(\eta q)_M$ (±σ)	$(\eta q)_A$ (±σ)	GQR (±σ)
					D-2				
2673 (42)	3199 (30)	2668 (42)	1897 (30)	1.00 (±0.03)	0.59 (±0.02)	1.68 (±0.04)	0.92 (±0.02)	0.94 (±0.01)	0.59 (±0.02)
					D-3				
2275 (38)	2685 (26)	2320 (38)	1517 (26)	1.02 (±0.03)	0.57 (±0.02)	1.73 (±0.05)	0.93 (±0.02)	0.96 (±0.03)	0.59 (±0.02)
					Mn-1				
2446 (45)	3116 (30)	2497 (45)	1703 (30)	1.02 (±0.03)	0.55 (±0.02)	1.91 (±0.05)	0.93 (±0.02)	0.95 (±0.01)	0.59 (±0.02)
					Mn-2				
2353 (38)	2944 (25)	1589 (38)	1588 (25)	0.68 (±0.02)	0.54 (±0.02)	1.90 (±0.05)	0.92 (±0.02)	0.92 (±0.01)	0.61 (±0.02)
					TF-42				
1925 (45)	1979 (22)	1602 (45)	1055 (22)	0.83 (±0.03)	0.53 (±0.02)	2.10 (±0.07)	0.89 (±0.01)	1.00 (±0.05)	0.56 (±0.03)

N is the number of counted tracks in external surface (ES), internal surface (IS), external detector coupled to external surface (ED(ES)), external detector coupled to internal surface (ED(IS)), (f) is the number of counted fields, ρ are densities (using the same sub-index), ETA-q (ηq) values for muscovite (M) and apatite (A). GQR is the product between G(=2π/4π = 1/2), Q = $(\eta q)_M/(\eta q)_A$ and the range deficit factor value, R. For Durango apatite, the R-value is 1.210±0.03 (as in Jonckheere, 2003), with L_0=16.28±0.21. For TF-42 R-value is 1.250±0.04 with L_0=15.71±0.28.

Geometry factor values obtained through ρ_{ED}/ρ_{IS} are systematically lower than GQR values, which may be an artifact of considering the projected length distribution as generated by tracks with only one average length, which results in the triangular distribution of Eq. (2). In fact, the two fragments propelled in the fission process have different masses and ranges and, therefore there should be two mean track lengths.

Note that, for the two methods, the values of g found for TF-42 are lower than the values found for the Durango apatite mounts. However, although this difference is larger for GQR values, error bars do not allow the conclusion that efficiency has changed. For this reason, it is appropriate to analyze the geometry factors obtained for other embasement rock samples.

Geometry factors calculated for eight embasement samples, according to Eq. (8), are shown in Table 2. The weighted average value is also presented. The weighted average of these geometry factors is 0.56±0.02 (χ^2_ν ~1.34; $P_\nu(\chi^2_\nu)$~0.23). Comparing this value with the average values for the prismatic sections of Durango (Mn-1 and Mn-2) and for the Durango

samples containing grains at random orientation (D-2 and D-3), no statistically significant difference is observed. While this should not be taken as general rule, the counting efficiency did not show an important difference between analyzing grains at prismatic or random orientation.

As stated before, the method presented in this work allows the measurement of a value of g for each dated sample. For instance, the ages of the eight embasement samples used to determine g were calculated using Eq. (1). Ages presented are EDM, which, using the geometry factors shown in Table 2, are (by definition) identical to PM ages e.g. Grimmer et al. 2002; De Grave and Van den haute, 2002; Glorie et al., 2010. The last column of Table 2 shows the results for the χ^2 test for the individual grain ages of each sample. The P-values are around 100% showing that it is safe to apply the PM protocol to these samples. Values of the constants are: $C_{238} = 0.99275$ (Lederer and Shirley, 1978), $\lambda_f = 8.45 \times 10^{-17}$ a^{-1} (Holden and Hoffman, 2000). This value is compatible with values measured using the fission-track method (Hadler et al., 1981; Guedes et al., 2000; Guedes et al., 2003a, 2003b; Yoshioka et al., 2005), $\lambda = 1.55125 \times 10^{-10}$ a^{-1} (Lederer and Shirley, 1978), $\varepsilon_I / \varepsilon^{238} = 1$ (Bigazzi et al., 2000).

The R_U value is given as:

$$R_U = \frac{\rho_U^V}{N_U^V \varepsilon^V} \qquad (9)$$

In Eq. (9), ρ^V_U is the induced density in muscovite coupled to standard glasses and $N^V_U \varepsilon^V$ is the calibration factor of dosimeter glass obtained using uranium thin film (Iunes et al. 2002, 2004). For the samples dated in this work, $R_U = (9.02 \pm 0.4) \times 10^{-9}$, which represent the number of fission events by target nucleus.Using the presented method, it is possible to build a database of geometry factor values for different apatite species, just as Donelick et al. (2005) have suggested for the length of the unannealed fission tracks (discussion below). This database would allow for using a, at least closely, adequate geometry factor, even in situations, in which it is not possible to measure a value of g for each sample, i.e., when the number of apatite grains is not sufficient to an additional mount to be irradiated together with the EDM mount.

Note also that the values of g presented in this work are in the range of values presented in literature: 0.51±0.02 (Gleadow and Lovering, 1977); 0.55±0.02 (Iwano and Danhara, 1998); 0.61±0.01 (Enkelmann and Jonckheere, 2003).

TABLE 2. Values of spontaneous and induced fission track densities using PM and EDM and ages obtained through EDM.

Sample	$(N_S)_{(EDM)}$	$\rho_S x10^6$ (±1σ)	$N_{I(EDM)}$	$\rho_{I(EDM)}$ $x10^6$ (±1σ)	$N_{I(PM)}$	$\rho_{I(PM)}x10^6$ (±1σ)	L_0(±1σ)	T_{EDM} (±1σ)	g= $\rho_{I(EDM)}/\rho_{I(PM)}$ (±1σ)
TF-5(1)	107	0.14(±0.01)	139	0.17(±0.01)	202	0.33(±0.02)	15.5(±0.2)	46.1(±6.3)	0.52(±0.06)
TF-9(1)	312	0.33(±0.02)	301	0.26(±0.01)	495	0.42(±0.02)	15.8(±0.2)	70.9(±6.7)	0.62(±0.05)
TF-10(1)	335	0.70(±0.04)	147	0.51(±0.04)	625	0.93(±0.04)	15.8(±0.1)	76.6(±8.5)	0.55(±0.05)
TF-12(1)	329	0.33(±0.02)	370	0.44(±0.02)	638	0.78(±0.03)	16.0(±0.2)	42.0(±3.8)	0.56(±0.04)
TF-17(2)	177	0.34(±0.03)	54	0.21(±0.03)	347	0.43(±0.02)	15.4(±0.3)	90.3(±14.7)	0.49(±0.07)
TF-21(3)	130	0.58(±0.05)	232	0.78(±0.05)	307	1.55(±0.08)	15.4(±0.2)	41.6(±5.0)	0.50(±0.04)
TF-30(2)	394	0.41(±0.11)	253	0.56(±0.04)	586	0.91(±0.04)	---	41.0(±3.9)	0.62(±0.05)
TF-32(2)	509	0.26(±0.01)	127	0.28(±0.02)	404	0.45(±0.02)	14.9(±0.4)	51.9(±5.7)	0.62(±0.06)
						average			**0.56±0.02**

$N_S(N_I)$ are the numbers of counted spontaneous (induced) fission track for External Detector Method (EDM) and Population Method (PM); ρ_I are the induced densities for EDM and PM, the ratio $\rho_{I(EDM)}/\rho_{I(PM)}$ is the geometry factor for each sample, L_0 are the initial length of horizontal confined fission tracks and χ^2_ν is the reduced chi-square for the individual grain ages. These samples were presented previously in Tello et al., 2005.

(1)Mantiqueira Mountain Range

(2)Mar Mountain Range

(3)Adjacent Areas

4.2. Initial length of horizontal confined fission tracks

The lengths of the horizontal confined induced fission tracks have been measured. The values found for this relatively homogeneous group of samples can vary up to ~6%,

from 14.9 to 16.0 μm and values up to 16.7 μm can be found in literature (Carlson et al., 1999). As indicated in the introduction, an advantage of irradiating pre-annealed apatite samples together with the EDM assembly is the possibility of measuring the initial fission-track length (lengths of the unannealed fission-tracks), L_0, which is a necessary input for modeling thermal histories and is a natural output in PM dating (Tello et al., 2003, 2005, 2006; Ribeiro et al., 2005). It turns out that as EDM is currently the most applied dating method, the practice of measuring L_0 was abandoned because, for EDM, induced tracks are registered in muscovite. For modeling, literature values are often adopted, which may yield inaccurate thermal histories, since values of L_0 may vary significantly from one sample to another (Table 2) and among analysts (Ketcham et al., 2009). Donelick et al. (2005) suggested a database composed of unannealed lengths of apatite samples. They suggest that 50 or more apatite species with different chemical compositions would give significant additional information.

The method suggested in this paper would allow the measurement of a L_0 for each apatite and, at the same time, contribute to the formation of a database to be used in cases in which the measurement of L_0 is not possible. One such example of this is the determination of the fission-track age via LA-ICP-MS. In this case, uranium content is determined via mass spectrometry and neutron irradiation is not necessary (Hasebe et al., 2004; Hadler et al., 2009, Ito and Hasebe, 2011).

To illustrate the impact of L_0 in thermal history modeling, one of the samples analyzed for measurements of g and L_0, TF-10, was modeled with different values of the initial fission track length. This sample was chosen because of the good number of horizontal confined for both spontaneous and induced fission track that could be measured, respectively 75 and 56. Thermal histories were modeled with the computer program HeFTy (Ketcham, 2005). The Linear Arrhenius Model (Laslett et al., 1987) was assigned. Two comprehensive Monte Carlo boxes were initially chosen, in order to search for general trends in thermal history model. Firstly, to the segment parameter value of zero for Halve and Episodic for Randomizer stile was attributed. In this way, only segments with their end points inside Monte Carlo boxes are randomly chosen and any breakup out the box was forbidden. The merit value for accepted fit was 0.0001. Thus, the weight for average curve is significantly major for a good fit. We chose the values of L_0 to vary in the range of

values obtained in this work and presented in the literature (Carlson et al., 1999), 14.9 to 16.7 µm. The thermal histories modeled with the different values of L_0 are shown in Figure 2 (a to f).

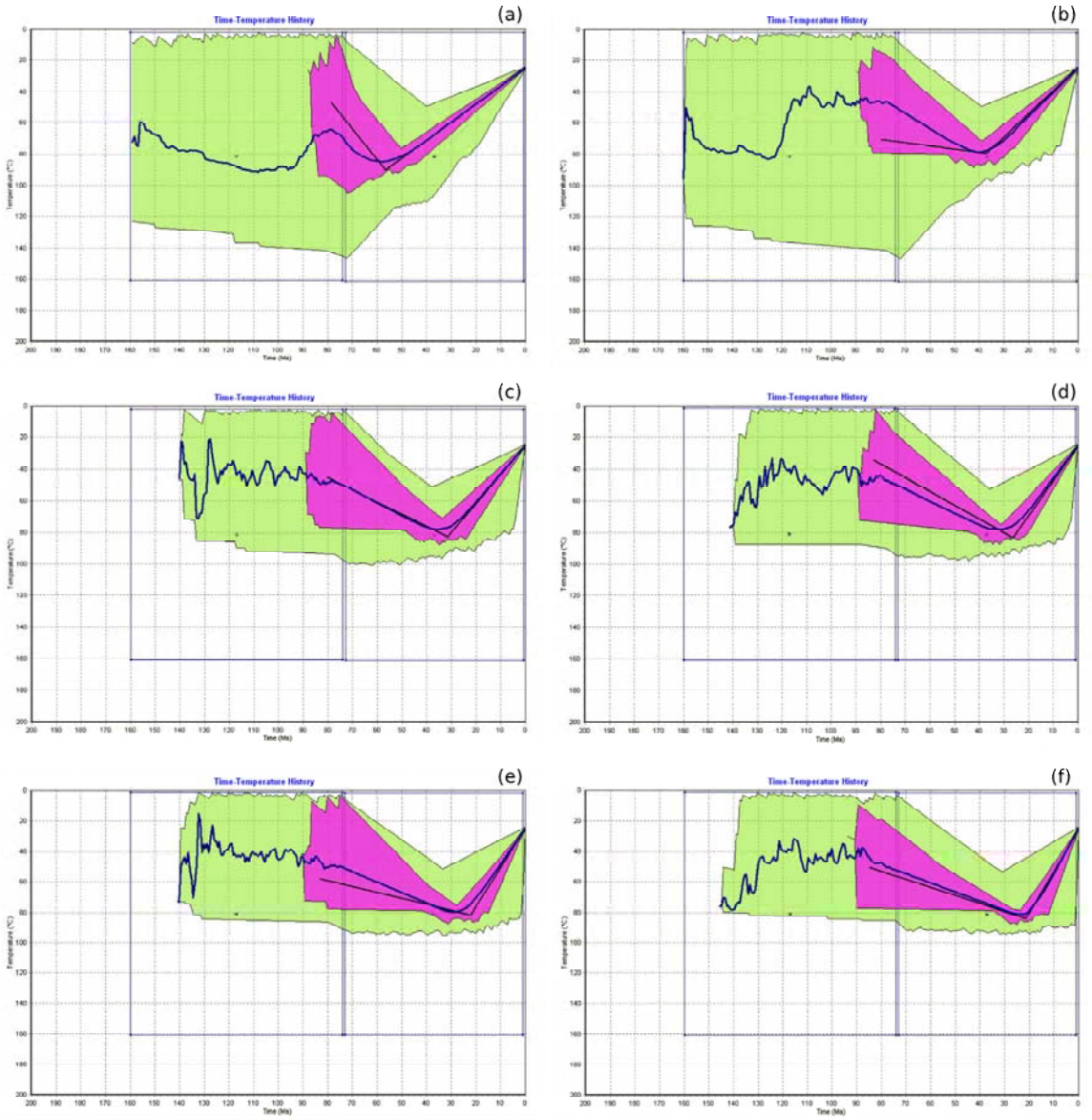

Figure 2. Thermal history for different values of L_0: (a) 14.9µm, (b) 15.5µm, (c) 15.8µm, (d) 16.0µm, (e) 16.3µm and (f) 16.7µm. The attribution of segment parameter value was zero for Halve and Episodic for Randomizer stile.

The most pronounced variation is in the last cooling episode, which becomes faster as the value of L_0 increases. This trend is more clearly observed in Figure 3, which shows the relationship between angular coefficient (ratio temperature/time) and the L_0 value. The greater is L_0-value, higher is the reduction in length (higher degree of annealing), which must be modeled with a longer residence inside the partial annealing zone.

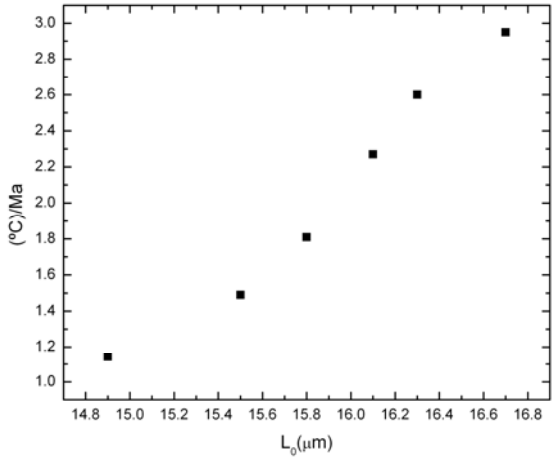

Figure 3, Relationship between angular coefficient (ratio temperature/time) of the last cooling of each thermal history in Fig. 2 and the L_0-value.

Finally, thermal histories were modeled remodeled, but this time setting Halve with one segment parameter and keeping the Episodic for Randomizer stile. Again, it is observed that the residence inside the partial annealing zone is longer for greater values of L_0 (Figure 4).

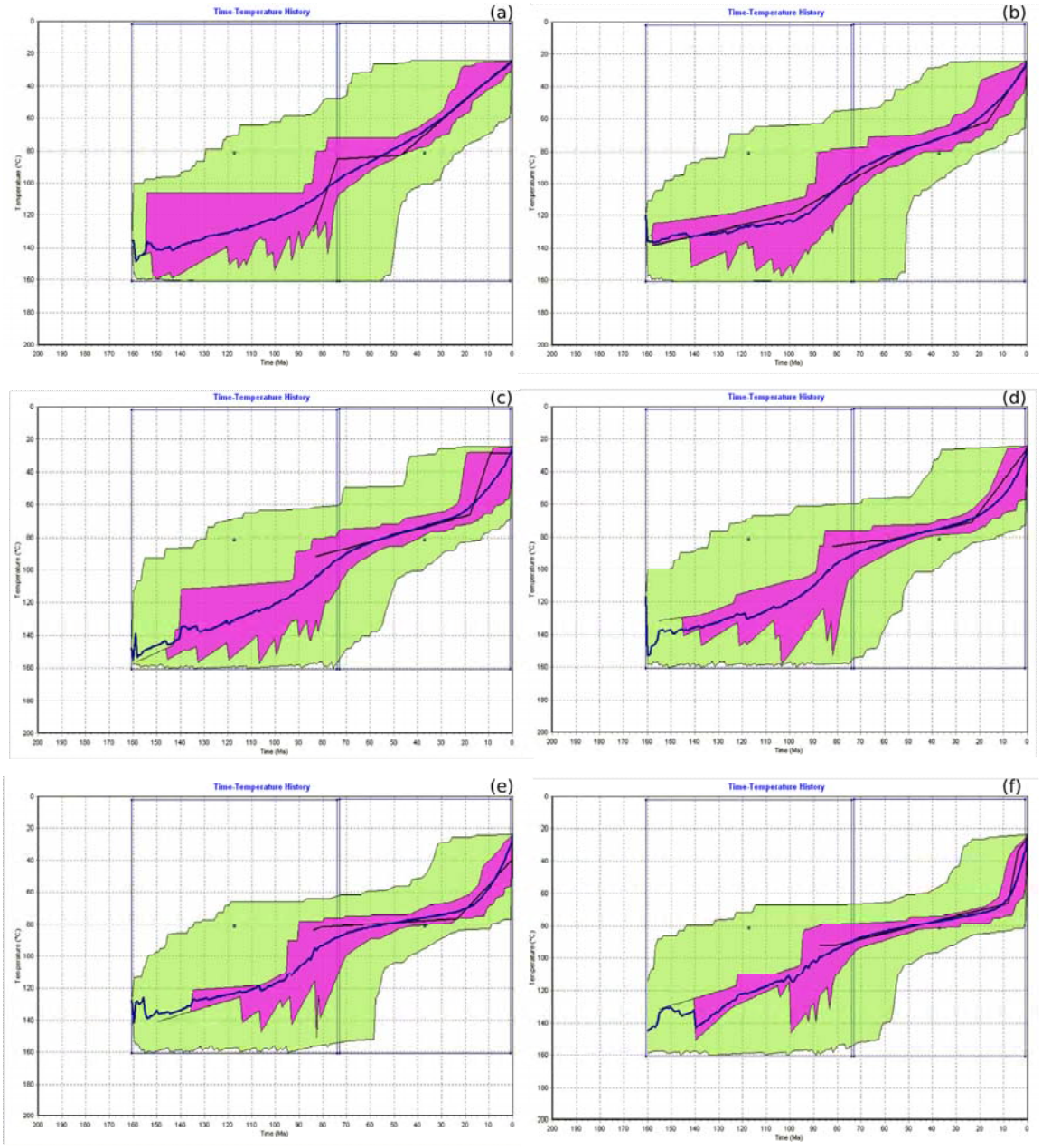

Figure 4. Thermal history for different values of L_0: (a) 14.9µm, (b) 15.5µm, (c) 15.8µm, (d) 16.0µm, (e) 16.3µm and (f) 16.7µm. The attribution of segment parameter value was one for Halve and Episodic for Randomizer stile and keeping the Episodic for Randomizer stile.

4.3. Implications for the ζ-calibration

All values for the geometry factor presented above are independent from the neutron dosimetry, as they result either from projected length measurements or from ratios of induced densities in mounts of the same apatite samples, irradiated together. In this way, the values of g and L_0 could equally have been determined by and serve to ζ practitioners. The g factor appears in the ζ-age equation (for instance, Donelick et al., 2005):

$$t = \frac{1}{\lambda} \ln\left[1 + \lambda \zeta g \rho_d \frac{\rho_S}{\rho_I}\right] \quad (10)$$

In Eq. (10), ρ_d is the induced fission-track density for a uranium dosimeter glass used to monitor neutron fluence during neutron irradiation, ζ is the calibration factor, which depends on the analyst efficiency, on the dosimeter glass and on the age of an apatite standard, usually Durango. The other symbols are the same as for Eq. (1). The ζ-calibration factor is found inverting Eq. (10) for the standard sample:

$$\zeta = \frac{\exp(\lambda t_{STD} - 1)}{g_{STD} \lambda \rho_d (\rho_S / \rho_I)} \quad (11)$$

where the sub-index STD refers to the standard sample. Note that the geometry factor appears also inside ζ. It is normally assumed that the geometry factor is the same in both standard and unknown age sample. However, this may not be the case. A more accurate approach would be to determine g_{STD}, instead of assuming g_{STD}=0.5, and the value of g for that sample. In this way, the eventual differences in efficiency are accounted for.

5. Conclusion

New values for geometry factors, obtained through projected length distribution and the ratio between induced fission-track densities in muscovite external detector and apatite, were presented. For this, five apatite samples have been used, two of which were extensive surface (Mn-1 and Mn-2, Durango apatite) in prismatic section and three random orientation grain mounts (D-2 and D-3, Durango apatite and TF-42, from crystalline rock). No significant differences were observed among the obtained values.

The average value of the geometry factors found for basement samples (measured in random orientation grains) is statistically in agreement with the values measured for Durango samples, both in prismatic sections and random orientation grain samples. This result indicates the possibility of dating non prismatic section grains in cases in which there is paucity of grains as it is the case, for instance, of most basin samples.

With an example, it was illustrated how the value of L_0 determines the modeled residence of the sample inside the partial annealing zone, showing the important role of the L_0-value for thermal history modeling. In this way, the possibility of measuring this parameter in the same sample and at the same analysis condition the fossil tracks are measured, including etching, microscopy equipment and analyst observation criteria, should be consider. It was also shown an extension of the usual method of measuring g-value that makes possible to obtain an individual value of L_0 for each sample to be dated.

In addition, the possibility of obtaining g-values when the ζ-calibration is applied is advisable because the geometry factor measured for the standard may be different from the geometry factor measured for the unknown age sample.

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Capítulo 2

RECALIBRATION OF DOSIMETER GLASS THROUGH NATURAL URANIUM THIN FILM

Abstract. This work presents the recalibration of U-doped glass as neutron dosimeter calibrated against natural uranium thin film previously presented by (Iunes *et al.,* 2002, *Chem. Geol.,* **187**, 201-211). For this, apatite samples from two alkaline formations (Alto Paranaíba and Ponta Grossa arches) and standard Durango apatite were used considering well known their ages obtained by other radiometric dating methods. The ages of these samples carried out employing two nuclear reactors with different characteristics were compared. For the samples from Alto Paranaíba arch age results agree with those determined by other radiometric dating methods which present a higher closure temperature indicating that for a low degree of annealing, fission-track age should not be corrected. For the samples from Ponta Grossa arch there is no agreement between fission track and radiometric ages; the reasons for this are discussed. Thermal histories of Alto Paranaíba and Ponta Grossa arches are presented and discussed.

Keywords: Fission-track thermochronology, alkaline intrusions, dosimeter glass, uranium thin film, ϕ-method, heavy ion irradaition.

1. Introduction

Age equation for Fission-Track Thermochronology, FTT, can be written according to Iunes et al. (2002):

$$t = \frac{1}{\lambda}\ln\left[g\frac{(\rho_S/\rho_I)}{(L/L_0)}\frac{\lambda R_U}{C_{238}\lambda_f} + 1 \right] \quad (1)$$

Where $\rho_{S(I)}$ is the spontaneous (induced) fission-track density; g is the factor which embraces geometric, etching and observation efficiencies; λ_f is the ^{238}U spontaneous fission decay constant; $L_{(0)}$ is the spontaneous (induced) fission-track average length; C_{238} is the ^{238}U isotopic concentration in the natural uranium and R_U=Σ R_i (R_i is the event by target nuclei of the ^{i}U, i=235.238). R_U-value is given by:

$$R_U = \sum_i C_i \int_0^\infty \sigma_i(E)\phi(E)dE \quad (2)$$

In the Eq. (2), C_i is the isotopic concentration of the ^{i}U in the natural uranium; $\sigma_i(E)$ is the cross section for the nuclear reaction ^{i}U(n,f), in function of neutron energy, E; $\phi(E)$ is the neutron fluence with energy E per unit energy.

Certainly one of the most discussed problem within FTT is the difficulty to determine the R_U-value in a neutron facility when the traditional dosimetry is applied (e.g., Green and Hurford, 1984; Crowley, 1986; Tagami and Nishimura, 1989). In this case, the neutron fluence is determined through metal monitor as Au and Co (Van den haute *et al.,* 1988; De Corte *et al.,* 1991, 1998; Jonckeehere, 2003). For thermal energies (neutron energy less than 0.5eV) their cross section curves are represented by 1/v relationship, where v is the neutron velocity. In this energy interval cross sections for the metal monitors ^{197}Au(n,γ)^{198}Au and ^{59}Co(n,γ)^{60}Co are approximately parallel to the one of ^{235}U(n,f) reaction. However, for the energy range of the epithermal neutrons a resonance is observed for both metal monitors, i.e., in a nuclear reactor whose Cd to Au ratio is less than 3, the equations above cannot be applied because there is a significant difference in the response between the reactions ^{235}U(n,f) inside the mineral and (n,γ) measured in the metal monitors (Green and Hurford, 1984). In addition, epithermal neutrons can also induce the ^{235}U

fission with different response (compared to thermal region) and ^{238}U fission which can lead to error in the age. For fast neutron, ^{235}U, ^{238}U, ^{232}Th are fissionable whereas the (n,γ) cross sections for ^{59}Co and ^{197}Au are negligible.

Th/U ratio presents the average value of ≈4 in the earth's crust (Dickin, 2005). However, this ratio in the Durango apatite samples, present a value around 25 (Hurley and Fairbairn, 1957). Thus, the ^{232}Th fission can reach 15% of the total fissions in a considered well thermalized neutron facility (Iunes, 1999). If U-doped glasses are calibrated against metal monitors this situation is softened but still the ^{238}U fission cannot be neglected.

Another problem widely discussed in the FTT was the accepted ^{238}U spontaneous fission decay constant, λ_f, whose values using fission-track procedures are $\approx 7.00\times10^{-17}y^{-1}$ (Fleischer and Price, 1964), as exception of Spaggiari, 1980; Hadler *et al.,* 1981; Vartanian, 1984. However, when other techniques are taken into account, λ_f presents two central values $\approx 7.00\times10^{-17}y^{-1}$ (Fleischer and Price, 1964; Roberts *et al.,* 1968; Hurford and Gleadow, 1977) and $\approx 8.5\times10^{-17}y^{-1}$ (Segré, 1952; Wagner *et al.*, 1975; Hadler *et al.*, 1981; Ivanov, 1985). This discrepancy is well documented in (Thiel and Herr, 1976; Bigazzi, 1981; Wagner and Van den Haute, 1992). More recently works (*e.g.,* Guedes *et al.,* 2003a; 2003b; Hadler *et al.,* 2003; Yoshioka *et al.,* 2005) present values which have been acceptable by *International Union of Pure and Applied Chemistry*, with value of $\approx 8.5\pm0.10\times10^{-17}y^{-1}$ (Holden and Hoffman, 2000).

Due to these two problems, the *Subcommission on Geochronology of the International Union of Geological Sciences (IUGS)* (Hurford, 1990) has recommended the procedure which links the FTT with other dating methods. This procedure is so-called ζ-calibration (Hurford and Green, 1983) and is based in the calibration of neutron facility by using a standard sample, e.g., Durango apatite (dated with other radiometric dating), and a glass dosimeter. This procedure was important for the standardization of the fission-track procedures as well as it makes possible results comparison among different laboratories. However, recent re-evaluations of the *IUGS* recommendation in the employment of the ϕ-method has been suggested (Hurford, 1998; Van den haute et al., 1998; Jonckheere et al., 2000). Some articles have reported that the utilization of natural uranium and thorium thin

films can be applied even in a low thermalized neutron facility (Bigazzi et al., 1999; Iunes et al., 2002; 2004; 2005). The use of uranium thin film brings a great advantage: the rate of uranium induced fission reaction that takes place in the dosimeter and the mineral are exactly the same, independently the neutron distribution and any variation or fluctuation during irradiation (Hadler et al., 1981) Concerning fast neutrons, the Th/U ratio of the mineral has to be considered and if necessary thorium thin films must be employed to take into account the thorium fissions by fast neutrons inside the mineral. When uranium thin films are irradiated against muscovite external-detector, induced fission-track density is proportional to R_U-value (Iunes et al., 2002):

$$\rho_F = \varepsilon^F N_U^F R_U \qquad (3)$$

In the Eq. (3), $N_U{}^F$ is the number of uranium atoms per unit area of the thin film (Bigazzi et al., 1999; Iunes et al., 1999; 2002; 2004) and ε^F is the efficiency detection for muscovite attached to it. Bigazzi et al. (1995) and Hadler et al. (1996) have shown through comparison with nuclear emulsion that the efficiency detection for muscovite external-detector is 1.00±0.025.

When geological samples are irradiated, neutron fluence should be around $\approx 10^{14}$ to 10^{15}cm^{-1} and thin films cannot be applied. This restriction is due the high uranium content deposited when thin films are manufactured. Thus, with high uranium content together with neutron fluence described above, there is superposition of fission tracks in the muscovite external-detector attached to thin films, makes impossible to count under ordinary optical microscopy. Problems in the manufacturing thin films with sufficiently low uranium content are reported by Bigazzi et al. (1999). Fortunately U-doped glass dosimeters with suitable uranium content can be calibrated against thin uranium films and then employed to carry out neutron dosimeter inside fission track dating. Density results, ρ_d, from glass dosimeters can be related with R_U through the following the equation:

$$\rho_d = \varepsilon^V N_U^V R_U \qquad (4)$$

In the Eq. (4), $N_U{}^V$ is the number of uranium per unit volume in the glass dosimeter; ε^V is the efficiency detection of the muscovite external-detector attached to it (Iunes et al.,

2002; Bigazzi et al., 1991). The calibration of a U-doped glass against a thin U film can be done if they are irradiated together and juxtaposed. With the determination of R_U-value through Eq. (3) by using uranium thin films and measuring the fission-track density, ρ_d, the $\varepsilon^V N_U{}^V$ can be determined through Eq. (4). Thus, R_U-value can be determined of subsequent irradiations for dating by using U-doped glass.

The aim of this work is the recalibration of the U-doped glasses through uranium thin film, previously presented by Iunes et al. (2002). The main difference presented in this work is the calibration of thin film through alpha-particle electronic spectroscopy, making the U-doped glass calibration more direct and not depending on the ingenious laboratorial procedure as presented in Bigazzi et al. (1999) and Iunes et al. (2002; 2004). This method allows the determination of the R_U-value. Then, the fission-track age can be determined through the ϕ-method.

Also, apatite samples from Brazilian alkaline-carbonatites (Ponta Grossa and Alto Paranaíba arches) and Durango were dated. The results obtained with fission-tracks are compared with reported values obtained through other radiometric dating methods. Samples with low superficial tracks content were irradiated with heavy ions, allowing improvement of statistics of the spontaneous confined fission-track length measurements. These measures were used for obtain thermal history modeling of these arches.

2. Methods

2.1. Preparation and calibration of the uranium thin film

Uranium thin films have been prepared through procedure described by Yagoda, (1949), where it is prescribed a solution of uranyl nitrate plus parlodium dissolved in alcohol and ether. This solution is deposited in an appropriated muscovite plate. After the evaporation of volatile materials (drying stage), a thin pellicle containing a convenient quantity of uranium is obtained. The muscovite plate is then heated at 400ºC and the final result is the uranium oxide film adherent to this basis (Bigazzi et al., 1995; Iunes et al.,

2004). The thickness of the thin film is sufficient to neglect the self-absorption of fission-fragment.

Iunes et al. (2002; 2004) have calibrated uranium thin film through alpha activity by using solid state nuclear track detectors (CR-39) calibrated against nuclear emulsion (Bigazzi et al., 1995). Therefore, this procedure involves the efficiency of detection, revelation and observation of these detectors, making multistep and ingenious procedure. A better solution for uranium thin film calibration is through alpha particle electronic spectroscopy. For this, a detector with diameter of 51mm and with distance sample/detector of 16.7mm is used. Vacuum was made in the sample/detector chamber to eliminate the alpha particle loss of energy in the air. The detection efficiency for alpha particles in the probed energy range is close to 100%. The geometric efficiency for each measured uranium thin film was calculated through Monte Carlo simulations. The detector channel/energy calibration was carried out by ^{241}Am (5500.8keV) and ^{239}Pu (5182.0keV) sources from the *Institute for Research, Production and Application of Radio-isotopes* – former Czechoslovakia, whose activity in December 1980 was of 0.669 kBq (± 3%) for the americium and of 0.816 kBq (± 3%) for the plutonium.

2.2. Nuclear reactor irradiation

Neutron dosimeter calibration was carried out in the IEA-R1 nuclear reactor, São Paulo state, Brazil. Glass dosimeter and uranium thin films were attached with muscovite external-detector plates. To avoid systematical error associated to neutron gradient, irradiations were carried out over sandwiches made up by juxtaposed surfaces of glass dosimeter/mica and mica/uranium thin film so that no more than 1mm of mica separate both dosimeters. The nominal neutron flux was $\approx 1\times10^{8}$cm^{-2}s^{-1}. An irradiation using such low neutron flux was necessary to control and obtain the suitable neutron fluence, considering high uranium content of the thin film. Thus, with irradiation of 60 hours it was possible to obtain a good amount of tracks in muscovite external-detectors attached in both,

thin film and U-doped glass. Through the Eq. (2) considering ^{235}U fission, the neutron fluence was $\approx 2.5 \pm 0.08 \times 10^{11}$ cm^{-2}. The muscovite external-detectors attached to dosimeter glass and uranium thin films, were etched with HF 48%, for 90 minutes at 15°C. Their track densities, ρ_d, for the former and, ρ_F, for the latter were measured under ordinary optical microscope Dialux 20 EB, with nominal magnification of 500×, dry.

Alkaline apatite samples were collected in the alkaline-carbonatites complex associated with uplift area on the boundary of Paraná basin. Araxá, Catalão and Tapira complex have radiometric ages between 78 and 85Ma and are inserted in Alto Paranaíba arch. Barra de Itapirapuã, Ipanema and Jacupiranga complexes, with radiometric ages of ≈130Ma, are inserted in Ponta Grossa arch (Gomes et al., 1990). Radiometric ages are shown in table 1.

The grains of each sample were divided in two aliquots: one of them was used to measure the length of the spontaneous horizontal confined fission-tracks. Another aliquot was used to determine the fission-track age. Grains were submitted to the following treatment: i) dumped in a rubber form (Struers Flexform, 25mm), ii) covered with epoxy resin (Struers Epofix) and dried in a stove at 45ºC for ≈24 hours, iii) robbed with #1200 sand paper so that an internal surface was reached and iv) polished with liquid suspensions of diamond in granulometries of 6, 3, 1 e 0.04 μm. The Spontaneous fission-tracks in apatite grains were revealed with HNO_3 25%, for 15s at 25ºC, and in order to obtain the induced tracks first a muscovite external-detector was coupled to each sample and then irradiated with neutrons. In the present case, irradiation was carried out in the FRM-II (Garching, Germany) research nuclear reactor at Technische Universität München, under a thermal to epithermal neutron ratio of ≈600×, thus a very well thermalized facility.

In this way, the Induced fission-tracks crossing the polished apatite grain surfaces were recorded in the external detector, forming a mirror image of (the grains) each sample. Glass dosimeters already calibrated were used for determination of neutron fluence. Taken into account that a very well thermalized neutron facility was employed thorium induced fission by fast neutron could be neglected.

After irradiations muscovite external-detectors were etched at the same conditions for the glass dosimeter calibration, described above. To perform track counting, the repositioning technique firstly proposed by Jonckheere et al. (2003) was used. In this technique the muscovite is re-placed over the mount containing all the apatite grains in the same position they were irradiated, so that spontaneous, ρ_S, and induced, ρ_I, fission track densities can be measured just changing the focus of the microscope.

As this work presents an evaluation of the calibration of dosimeter glass, samples were also irradiated in the IEA-R1 nuclear reactor, São Paulo, Brazil. This nuclear reactor does not present a well thermalized neutron spectrum as is the case of FRM-II research nuclear reactor. Apatite samples from Catalão, Jacupiranga and Tapira alkaline besides the standard Durango apatite were dated. Due the low uranium content in the alkaline samples, especially in the case of Jacupiranga, a low amount of induced tracks is observed in the muscovite external-detector, making difficult the repositioning of the detectors. Then these samples were dated through the population method. It is important to mention that the alkaline samples dated with external detector method, irradiated in FRM-II research nuclear reactor are not the same of samples dated with population method, irradiated in IEA-R1 nuclear reactor. However, they were collected from the same alkaline complex and mustn't give different fission-track age. The track density measurements were carried out with an optical microscopy Zeiss Axioplan II by using the nominal magnification of 1000×, dry.

Table 1. Radiometric ages of alkaline samples used in the present work.

Sample	**Radiometric age(Ma)**	**Reference**
Araxá	78,4±0,7	Gomes et al., 1990
Catalão	84±4.0	Gomes et al., 1990
Tapira	87.2±1.2	Biondi, 2005
Barra de Itapirapuã	128±19	Ruberti et al., 1998
Ipanema	133±4.0	Gomes et al., 1990
Jacupiranga	131±3.0	Gomes *et. al.,* 1993

2.3. Heavy ion irradiation

Horizontal confined fission-tracks have been used to quantify the annealing degree suffered by geological (apatite) samples. As these tracks are totally inside volume of the mineral, they are etched when cross superficial tracks or cleavages. However, can be difficult to found an adequate statistical when samples present a low fission-track density. A proper way to improve the amount of horizontal confined fission-tracks is to create channels through the mineral surface. For this, the minerals can be irradiated with ^{252}Cf fission fragments or with heavy ions produced in a particle accelerator. In the present work, ^{78}Kr ions accelerated to 11.1 MeV/amu on the linear accelerator of Gesellschaft für Schwerionenforschung (GSI), Darmistadt, Germany, were employed. Ions are acquired by scattering or evaporation and delivered by an electric field to the linear accelerator UNILAC, where they are accelerated at a distance of 120m reaching 20% of the light velocity. Total energy of the ^{78}Kr ions is 866MeV. This energy results in a range of 95.4μm in apatite, calculated by SRIM2003 program (Ziegler and Biersack, 2003). However, it is interesting that ions reach the mineral surface with energy dE/dx (specific energy loss) maximized. For this, samples were covered with aluminum foils (total thickness of 64.3μm) reducing the incident particle energy to ≈250MeV.

After ion irradiation, samples are etched as described above. The etching opens a channel (micropores) in the region damaged by the ion. Confined fission-tracks that cross these channels are also etched. Horizontal confined fission-tracks measurements were carried out in an optical microscopy Zeiss Axioplan II, with aid of a digitalized table and the CCD camera, using a nominal magnification of 1500×, dry. Fig. 1a shows an etched confined fission-track that crossed a heavy ion channel. Etched channels are also shown in Fig. 1b.

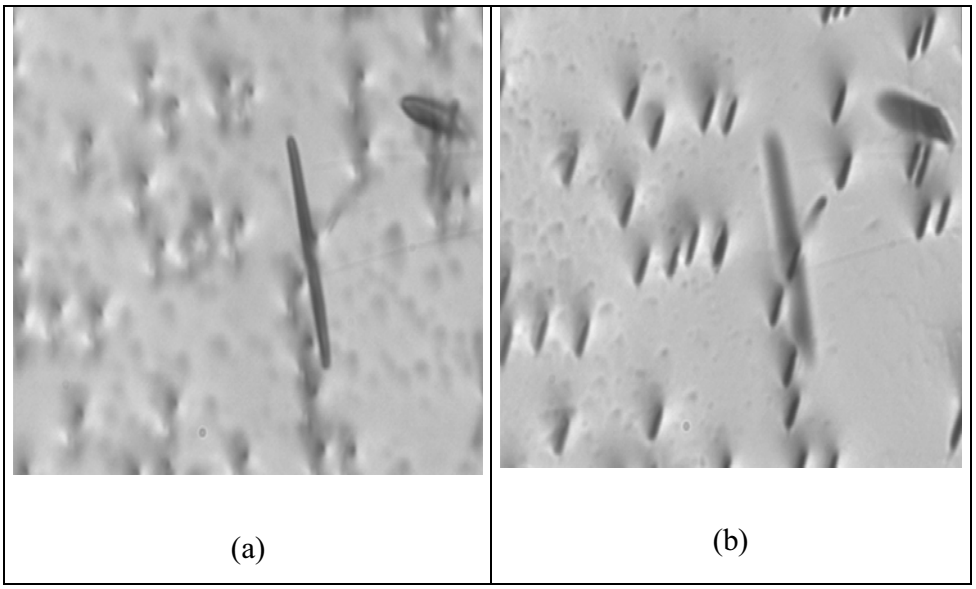

Figure. 1. A confined fission-track etched through a ^{78}Kr (250MeV) channel. (a) Focus on the confined fission-track; (b) Focus on ^{78}Kr channels.

2.4. Determination of geometric factor, g.

The geometric factor, *g*, in the Eq. (1) takes into account track efficiency (track counting per fission) due to differences in geometry, etching and observation under optical microscopy between apatite internal surface and the muscovite external-detector coupled to it during neutron irradiation. The main difference is that fission-tracks in apatite samples are recorded under 4π–geometry whereas in the external-detector 2π–geometry is involved.

To determine the *g*-value, two standard Durango apatite sections were heated for 48 hours at 450°C to erase spontaneous fission-tracks (total annealing). After this, samples were mounted and polished. Muscovite external-detector plates were attached and these assemblies were irradiated at the Thetis nuclear reactor of Institute for Nuclear Sciences, Ghent University (Belgium). After irradiation, apatite samples were again polished so that a 4π–geometry was reached. Both apatite internal surface and muscovite external-detector were etched through procedures described above. The experimental procedures used for

determination of the geometric factor are exactly the same used in the external detector method. In practice, g is determined by track density ratio of the muscovite-external detector and apatite internal surface.

3. Results

3.1. Dosimeter glass calibration

Table 1 shows recalibration of dosimeter glass through R_U-value determined by Eq. (3) and applied in the Eq. (4). The final $N_U{}^V\varepsilon^V$ values of the calibration are the average of the values determined through two irradiation: CN-1: $N_U{}^V\varepsilon^V=(1.43\pm0.05)\times10^{14}cm^{-2}$; CN-2: $N_U{}^V\varepsilon^V=(1.26\pm0.05)\times10^{14}cm^{-2}$; CN-5: $N_U{}^V\varepsilon^V=(0.40\pm0.03)\times10^{14}cm^{-2}$; IRMM540: $N_U{}^V\varepsilon^V=(0.52\pm0.02)\times10^{14}cm^{-2}$. The values for each dosimeter glass for irradiations I-1 and I-2 are statistically compatible, however, systematically lesser than the corresponding $N_U{}^V\varepsilon^V$ values previously presented by Iunes *et al.* (2002) between 2.4% for CN-1 to 6.7% for CN-2.

Table 2. Calibration of uranium standard doped glass.

Irradiation	Thin Film	N_{IF}	$\rho_F \pm 1\sigma$ ($\times 10^5 cm^{-2}$)	$R_U \pm 1\sigma$ ($\times 10^{-10}$)	Glass	N_{IG}	$\rho_D \pm 1\sigma$ ($\times 10^4 cm^{-2}$)	$N_U{}^V\varepsilon^V \pm 1\sigma$ ($\times 10^{14} cm^{-2}$)
I-1	XXVIII-2	9545	3.05±0.03	1.34±0.04	CN-1	2841	1.97±0.04	1.47±0.06
						1248	1.92±0.05	1.43±0.06
					CN-5	572	0.56±0.02	0.39±0.02
	XVIII-3	2377	2.98±0.06	1.39±0.05	CN-2	2849	1.75±0.03	1.26±0.05
						1829	1.79±0.04	1.28±0.06
					IRMM540	1067	0.74±0.02	0.53±0.03
						1332	0.76±0.02	0.55±0.03
I-2	XVIII-3	6153	3.28±0.04	1.53±0.05	CN-5	569	0.60±0.03	0.41±0.02
					IRMM540	949	0.73±0.02	0.48±0.02
	XXVIII-2	5980	3.21±0.04	1.41±0.05	CN-1	1315	1.96±0.05	1.39±0.06
					CN-2	1253	1.75±0.02	1.24±0.05
						917	1.68±0.06	1.19±0.06

I-1 and I-2 are different irradiations; $N_{IF(IG)}$ is the amount of counted fission-tracks in muscovite external-detector attached to the thin film (glass dosimeter); $\rho_{F(D)}$ is the superficial fission-track density in muscovite external-detector attached to the thin film (glass dosimeter); $N_U{}^V\varepsilon^V$ is the calibrator factor for uranium glass dosimeter.

3.2. Geometric factor determination

As the geometric factor depends on the observation efficiency it is important that each observer should carry out his own g measurements. Table 3 shows the result of the geometry factor, *g*, for two samples.

Table 3. Measurements of geometry factor

Sample	N_{ap}	$\rho_{ap}\pm1\sigma(10^5 cm^{-2})$	N_{ed}	$\rho_{ed}\pm1\sigma\ 10^5 cm^{-2})$	$g\pm1\sigma$ (N_{ap}/N_{ed})
1	3801	4.07±0.07	2262	2.42±0.05	0.595±0.017
2	3822	4.09±0.07	2275	2.43±0.05	0.595±0.020

$N_{ap(ed)}$ is the amount of counted fission-tracks in apatite (muscovite external-detector); $\rho_{ap(ed)}$ is the superficial fission-track densiy in apatite (muscovite external-detector).

From Table 3 the average of g-value is: g=(0.595±0.013). This value was used to obtain fission-track age which will be discussed below.

3.3. Ages determination through U-doped glass calibrated with uranium thin film

Table 5 shows the alkaline and Durango apatite ages, obtained through irradiations carried out in the FRM-II (Garching, Germany) research nuclear reactor at Technische Universität München. The fission-track ages were calculated according to Eq. (1). The used constants were: C_{238}=0.99275 (Lederer and Shirley, 1978), λ_f=8.5±0.1×10^{-17} a^{-1} (Holden and Hoffman, 2000), λ=1.55125×10^{-10} a^{-1} (Lederer and Shirley, 1978). To obtain ϕ-value (cm^{-2}), cross section for ^{235}U(n,f) reaction was adopted as σ=584.33b (Carlson, 2011). The ϕ and R_U-value can be determined through Eq. (2) and (4), respectively. For Catalão and Tapira samples from Alto Paranaíba Arch (Araxá, Catalão and Tapira) the ages agree with the values of literature dated with other radiometric methods (shown in Table1). Durango apatite measured here (table 5) present an acceptable value comparing with other radiometric methods e.g., McDowell et al. (2005) by using (U-Th)/He (31.0±1Ma). Also Green (1985) have reported age of K-Ar in feldspar (31.4±0.5Ma) and employing fission-track technique with ζ-calibration (30.4±1.7Ma). These values also agree with Van den Haute *et al.,* (1988) and Jonckheere, (2003) applying fission-track (ϕ-method) by using Au and Co monitors. However, for samples from Ponta Grossa arch (Ipanema, Barra de

Itapurapuã and Jacupiranga) the ages are considerably below the reference ages shown in Table 1.

Table 4. Fission-track age determined with U-doped glass dosimeter

Sample	Irradiation	$N_S \pm 1\sigma$	$N_I \pm 1\sigma$	$\phi(10^{15}\ cm^{-2})$	T (Ma)	L (μm)(N)
DUR2	B	1974±2.3	4605±1.5	2.58±0.08	33.1±2.0	14.6±0.1(36)
DUR3	A	9870±1.0	1343±2.7	0.127±0.005	28.0±1.7	14.6±0.1(36)
Araxá	B	---	---	---	---	13.2±0.2(29)
Catalão	B	1460±2.6	1360±2.7	2.58±0.08	82.6±5.3	14.3±0.1(139)
Tapira	B	623±4.0	539±4.3	2.58±0.08	88.9±7.4	14.2±0.2(64)
Barra de Itapurapuã	B	152±8.1	119±9.2	2.58±0.08	98.2±12.7	13.3±0.2(105)
Ipanema	C	348±5.4	519±4.4	3.44±0.08	69.1±6.5	13.7±0.2(112)
Jacupiranga	C	495±4.5	536±4.3	3.44±0.08	94.9±8.0	14.1±0.1(182)

1. $N_{S(I)}$ is the amount of counted fission-track in apatite (muscovite external-detector); ϕ is the neutron fluence determined by the glass dosimeter, L is the average of spontaneous fission-track length . t is the fission-track age in Ma.

2. Araxá sample cannot be dated due to its low spontaneous fission-track density. However, applying the heavy ion irradiation technique, it was possible to obtain the distribution of spontaneous confined fission-track length.

3. Standard U-doped glass used was CN-5 with the calibration showed in Table 1.

Tapira, Catalão and Jacupiranga alkaline complex and Durango apatite samples were also dated using a nuclear reactor presenting a neutron spectrum with a relatively low thermaliztion, the IEA-R1, São Paulo state, Brazil. These data are shown in Table 5. As discussed above, for the samples from Alto Paranaíba Arch, the ages agree with value of literature whereas sample from Ponta Grossa arch, the age is considerably below that of the reference age. Also, two Durango apatite samples present very similar values which are in agreement with data presented in table 4. Thus, the discussion above can be applied here.

Table 5. Fission-track age determined with U-doped glass dosimeter

Sample	N_S	$\rho_S \pm 1\sigma (x10^5)$	N_I	$\rho_I \pm 1\sigma (x10^5)$	$L \pm 1\sigma$	$L_0 \pm 1\sigma$	$t \pm 1\sigma$(Ma)
DUR-4	2087	1.88±0.04	3165	8.17±0.14	14.8±0.3	16.3±0.12	31.7±2.0
DUR-5	2530	1.93±0.04	3135	9.31±0.17	14.8±0.3	16.3±0.12	28.6±1.8
Tapira	602	2.93±0.11	490	2.72±0.12	14.9±0.3	16.6±0.79	86.9±7.0
Catalão	761	1.31±0.06	793	1.35±0.05	13.9±1.0	15.9±0.95	78.3±4.2
Jacupiranga	378	0.35±0.02	355	0.33±0.02	14.3±0.3	16.5±0.13	85.5±7.7

1. $N_{S(I)}$ is the amount of spontaneous (induced) fission-track counts; $\rho_{S(I)}$ is the spontaneous(induced) track density. $L_{(0)}$ is the average of spontaneous (induced) fission-track length. It is important to mention that L values presented in this work are in agreement with showed in table 5. t is the fission-track age in Ma.
2. Standard U-doped glass used was IRMM540 with the calibration showed in Table 1.

3.4. Age determination by using Durango standard age

In general, fission-track dating is obtained through so-called ζ-calibration. In order to apply this method, a significant number of irradiations should be carried out with purpose of obtaining a good calibration of the ζ-parameter. This calibration basically depends of the uranium content in dosimeter glass, age standard sample, nuclear reactor facility and the observer efficiency (Hurford, 1998). Articles as e.g., Shing and Nishimura (1991) and Hurford and Green (1983) present ζ-calibration measures for CN's and NIST glasses, where a variation in individual ζ-calibration values is observed. This can occur because the nuclear reactor configuration was changed along the time so the same conditions weren't kept during calibration and dating irradiations.

Another way to determine fission-track dating with aid of standard samples is making irradiation of the standard sample together with unknown samples. This procedure is the so-called *Z*-method. In this case, all constants and parameters related to irradiation are the same for both samples, and the age can be determined through following the equation:

$$t = t_{STD} \frac{\left(\rho_S / \rho_I\right)_{UKN}}{\left(\rho_S / \rho_I\right)_{STD}} \tag{5}$$

Where t_{SDT} is the age of the standard sample, dated independently by another radiometric technique, $(\rho_S/\rho_I)_{STD}$ is the ratio of spontaneous and induced fission-track density for the standard sample, $(\rho_S/\rho_I)_{UNK}$ is the ratio of spontaneous and induced fission-track density for the unknown age samples.

Eq. (5) was used to calculate fission-track ages for alkaline samples using the data presented in Table 5. The irradiation "B" was used in this procedure. Barra do Itapirapuã sample presented an age of ≈93.6±1.2Ma, Catalão ≈78.6±3.8 and Tapira ≈84.7±5.5Ma. These values present the same situation when ϕ-method is used, i.e., the ages are statistically compatible with fission-track ages using ϕ-method and the ages for Catalão and Tapira are in agreement with accepted values in the literature, however, Barra do Itapirapuã sample is below that of the reference age.

3.5. Spontaneous fission-track length distribution

Confined spontaneous fission-track length distributions obtained for some Brazilian alkaline samples are shown in Fig. 2. There are two distributions for Jacupiranga and Ipanema samples because they refer to two different samples from these two alkaline formations. All confined fission-track distributions shown in this figure can be related to fast cooling: average length lesser but not much different from L_0, relatively sharp and non significant tail on the left side of the distribution. This hypothesis can be evaluated using a program which allows the reconstruction of the thermal history of the geological samples (*e.g.*, Gallagher, 1995; Ketcham et al., 2000; Hadler et al., 2001). This kind of program is based on models (most of them are empirical) developed using data from annealing experiments carried out in the laboratory plus a geological calibration. For this it was considered that tracks are generated and accumulated along of geological time scale. Each accumulated fission-track records its thermal evolution considering the time-temperature path. Thus, the distribution of spontaneous fission-track gives information about the thermal evolution of the sample. The better approach is the spontaneous confined fission-track length. However, to determine properly the annealing degree, the L_0-value, i.e., average of unannealed fission-track length has to be measured under the same experimental conditions the annealed tracks were submitted to. For instance, Barbarand (2003) points out

that different observer in the same lab presented different measurement efficiencies, and, also variation in efficiency for a single observer along the time was observed. Ketcham et al. (2009) have reported a significant difference in L_0-values for different observer even using the ideal Durango apatite sample. Soares et al. (Submitted) have also reported a significant variation of L_0 for different samples with a consequent influence in the obtained thermal history. Thus, the thermal histories presented below were carried out by using L_0-values presented in table 5, considering that these samples are collected in the same geological setting. The software used was the Thermal History Analysis, *THA*, presented by Hadler et al. (2001) and group analysis was adopted, i.e., sample from the same geological settings should give the same thermal history (Hadler et al., 2008).

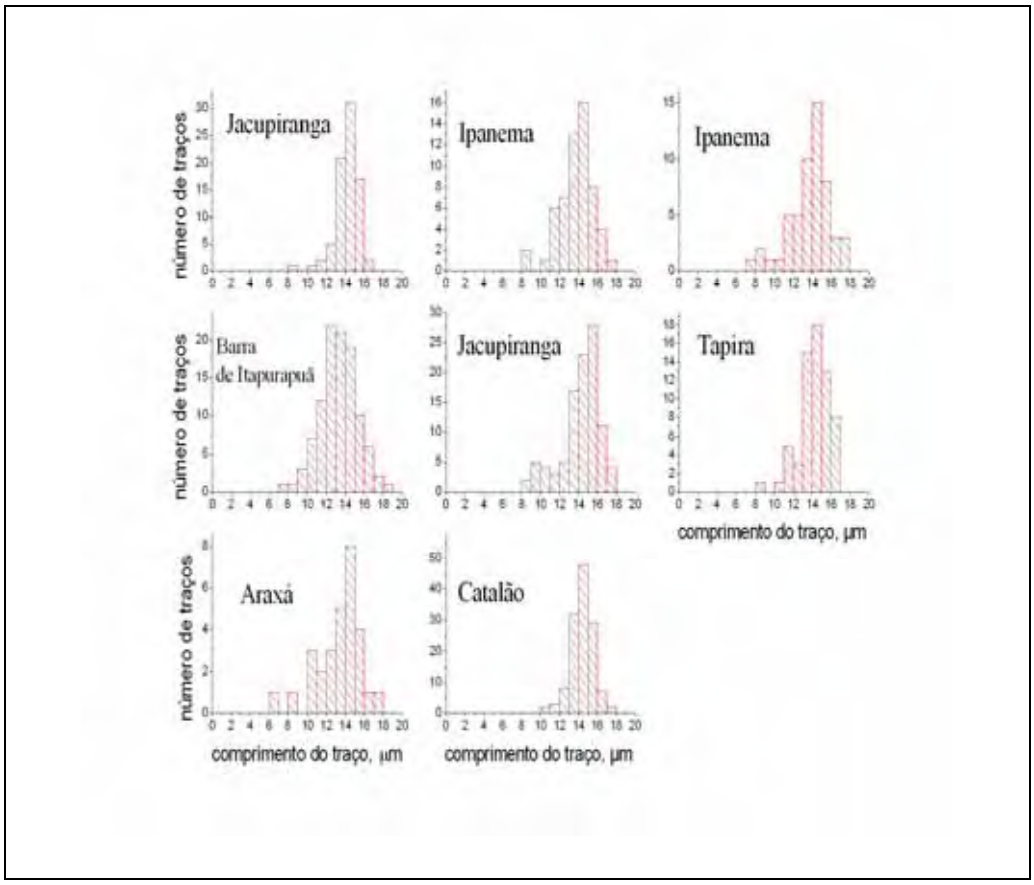

Figure 2. horizontal confined spontaneous fission-track distribution of the samples from the alkaline rocks studied here.

According to location, samples were divided in two groups: 1) samples associated with Ponta Grossa arch (Barra de Itapirapuã, Ipanema and Jacupiranga); and 2) Arco do

Alto Parnaíba arch (Araxá, Tapira and Catalão). It was assumed that formation of Barra de Itapirapuã has the same age of Ipanema and Jacupiranga samples. Fission-track age for Araxá samples cannot be determined but was supposed be equivalent to Tapira and Catalão ages. The results are showed in Fig 3. The dotted lines limit the set of all accepted thermal history associated with Alto Parnaíba arch whereas the full lines limit the set of all accepted thermal history associated with Ponta Grossa arch. The confidence interval is 90%.

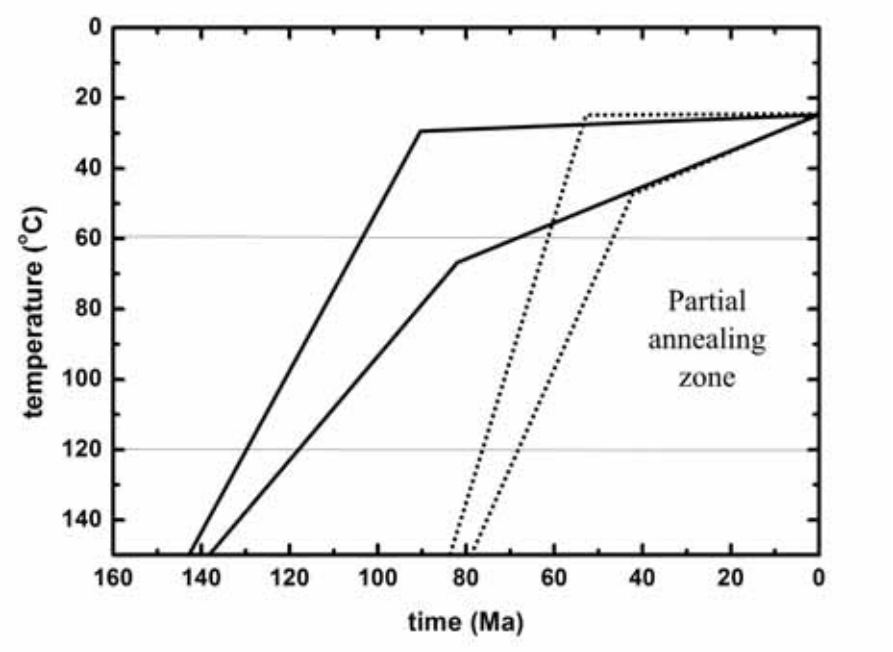

Figure 3. Thermal history associated with Ponta Grossa arch (Full line) and Alto Paranaíba Arch (dotted line).

Geological and geomorphologic evidences indicate that the evolution of the Ponta Grossa arch can be associated to post/sin-rupture of the Gondwana South-Western (Estrella, 1972; Asmus and Ferrari, 1978; Asmus and Porto, 1980; Asmus, 1981; Chang et al., 1992). The process of separation of South America from Africa and the formation of the South Atlantic Ocean took place along the divergent plate boundary. It began around ≈130Ma ago (Renne et al., 1992; Turner *et al.*, 1994; Renne *et al.*, 1996; Stewart *et al.*, 1996; Ernesto *et al.*; 1999), as is corroborated by, e.g., Rb/Sr isochron for mica and carbonatite (Roden et al., 1985) and K/Ar data (Amaral, 1978). This stage is characterized by numerous

dikes, shear zones, faults and fractures that seem to be reactivated at shallow crust levels, especially between Upper Cretaceous and Paleogene/Neogene, which caused block tilting and normal faulting. Gallagher and Brown (1999) have reported the supply of the clastic materials to the Santos basin around ≈100 Ma ago. However, the main period of deposition is ≈90Ma, which can be related to a fast denudation due the uplift of Ponta Grossa arch and the formation of Mar mountain range in the Albian-Aptian. This association was also reported by Pereira and Feijó (1994); Zalán and Oliveira (2005). These events can be interpreted as uplift at ≈90Ma and consequently the reactivation of shear zone. Through structural analysis of the São Bento group (Paraná basin), in the Ponta Grossa arch, Strugale et al. (2003) identified a brittle deformation event with direction of NW, and E-W, at period of Neocretace and Tertiary, as a reactivation of previous shear zone, probably formed in the Eocretace. Almeida and Carneiro (1998) have reported that Ponta Grossa arch was submitted to a major rise in the period of Upper Cretaceous, between 90 and 65Ma. According to Lima (2000), this episode of uplift in the Upper Cretaceous was associated by compressive effort intraplate as consequence of Andean collision causing a rising and erosion of the boundary of the Paraná basins in the South American Platform.

The fission-track ages determined for the Ponta Grossa arch samples may have been influenced by the uplift of in the Upper Cretaceous (≈90Ma) which was sufficient to erase fission-track in apatite whereas other radiometric dating presenting higher closure temperature were not significantly influenced. After this, no significant geological event was observed, as can be seen by the fast cooling shown in Fig (2) and (3). This evidence is supported by Vignol-Lelarge et al., (1994) which applied fission-track thermochronology in samples of basement rocks from Mar mountain range in areas of Ponta Grossa arch. They concluded that ≈86Ma ago this region suffered an uplift followed by erosion of 2.5Km, through a fast passage in the partial annealing zone. This uplift is associated to the formation of Mar mountain range, also evidenced by Tello et al. (2003; 2005).

According to Hackspacher et al. (2007), alkaline intrusions located in the boundary of Paraná basin and some alkaline bodies as Ipanema (Ribeiro et al., 2001), Poços de Caldas, Catalão and Salitre (Amaral et al., 1997) suffered with intense heating. The age of ≈60Ma can be related to thermal and tectonic inversion with reactivation of fault and uplift associated to denudation and tectonic process. Different ages between two stages of the

Mantiqueira mountain range are evidenced by normal fault which can be associated with two blocks: high and low Mantiqueira mountain range: the former with age in the Lower Cretaceous and the latter with age of Upper Cretaceous (Tello et al., 2003, 2005; Ribeiro et al., 2004).

For the case of a simples thermal history, i.e., where sample transit through partial annealing zone is relatively fast fission-track age can be associated to mineral age formation, also given by other radiometric methods. For samples from Alto Paranaíba arch, the results had shown in tables 5 and 6 and Fig 2 and 3 supports this hypothesis. The Alto Paranaíba arch is located between Paraná and São Francisco basin. During the early Cretaceous there was intense alkaline volcanism that preceded the latter Cretaceous by intrusions of dikes and possible basaltic effusion (Almeida, 1983). The geological event associated to this arch took place around ≈ 90Ma; especially concerned with the samples treated here this dating is around 90 Ma, with K/Ar (Amaral et. al., 1967), This is in agreement with fission-track dating and the thermal history presented in this work.

4. Conclusion

Durango standard and Alto do Paranaíba arch samples (Catalão and Araxá) dated in this work present ages in agreement with those obtained by other radiometric methods.

In the case of the samples of Ponta Grossa arch (Jacupiranga and Barra do Itapurapuã and Ipanema), fission track ages are significantly lesser than those obtained by other radiometric ages (see table 1). This can be associated with an expressive geological event that took place during the Upper Cretaceous, as commented above, which provoked a temperature rising sufficient to annul the fission track age. Thus, in this case this method gives the age of this event instead of that of the rock formation.

Ages of Durango samples obtained using the ϕ-method (using Au and Co dosimeters) in a nuclear reactor presenting very well neutron thermalization (Van den Haute *et al.,* 1988; Jonckheere, 2003) are in agreement the ones obtained in this work where U-doped glass dosimeters (calibrated against thin uranium films) were employed. In this case the neutron spectrum of the IPEN–CNEN nuclear reactor of São Paulo where the

irradiations took place was not well thermalized (Cd ratio for Au around 6). A consequence of this agreement is that the latter reactor did not induce a significant number of thorium fissions in the Durango apatite. Concerning induced fission in uranium, it should be noted that the employment of the neutron dosimetry by thin uranium films implies that the same nuclear reaction, U(n,f), produces fission tracks inside mineral to be dated and in the dosimeters. For this reason this methodology can used in the neutron facilities not well thermalized.

Finally, the age agreement between fission track and radiometric method for Durango and Alto do Paranaíba arch samples (Catalão and Tapira) also indicates that samples with low degree of annealing doesn't need any correction.

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Capítulo 3

DETERMINATION OF FISSION-TRACK AND RETENTION AGE THROUGH PLATEAU AND SIZE CORRECTION METHOD

Abstract. In order to study the effect of annealing on the ages determined by the Fission-Track (FT) Dating, FT ages (apparent ages) and retention (corrected) ages have been determined for apatites from Durango, Bamble, Brazilian alkaline rocks (Catalão, Tapira and Jacupiranga) and samples from Mantiqueira mountain range (MMR-1 and MMR-2). Retention ages have been obtained by the Plateau and Size Correction methods. Population method (PM) and subtraction method (SM) were applied because it is not possible to determine Plateau ages through the external detector method (EDM). It was found that for Durango, Catalão and Tapira, the FT and Plateau ages are in agreement with each other and with their reference ages, while their Size correction ages overestimate these reference ages. These samples present a reduced mean horizontal confined fission track (HCFT) length around 0.90. Therefore, the FT age is about 10% lower than the Size correction one. Bamble apatite, which presents mean HCFT length around 0.90, presents the same behavior. For the MMH samples, Plateau and Size correction ages overestimate FT ages and are in agreement with the age of the South Atlantic opening, event used as reference for these samples, which have reduced mean HCFT lengths around 0.75. The most probable hypothesis to explain the found results is that there is no age reduction for samples in which the reduced lengths is greater to or approximately 0.90 (lower degree of annealing), while samples that experienced heavier annealing (reduced lengths significantly less than 0.90) have their ages reduced according to the size correction hypothesis (proportional to the length reduction).

Keyword: Apatite, plateau method, size correction method, apparent age, retention age.

1. Introduction

Fission track Thermochronology, FTT, is based on accumulation of latent fission-tracks generated by passage of high-energy fragments from spontaneous fission decay of ^{238}U, present as impurities in minerals and volcanic glasses. Tracks that crossed a polished mineral surface are revealed by chemical etching and can be counted under ordinary optical microscopy.

Latent fission tracks are sensitivity to thermal treatment and are shortened by the combined action of temperature and time. This phenomenon is called annealing. Fission tracks are continuously generated over geological periods in the host mineral. Each track starts its shortening history at a different point on the temperature-time (T-t) path the host mineral experienced. After all, the net result of the mineral thermal history is a distribution of fission-track lengths. The measurable quantity that best approximates the fission-track length is the Horizontal Confined Fission-Track length, HCFT, parallel to observation surface (Wagner, 1987; Green *et al.,* 1989; Guedes *et al.,* 2004).

When these tracks are partially shortened (partial annealing), HCFT distribution provides information about the evolution of the host mineral in geological environments through thermal history reconstructions (Gallagher *et al.,* 1998, Tello *et al.,* 2003, Tello *et al.,* 2005) from the time-temperature point tracks start to be retained in the mineral. The time coordinate of this point is the retention age and temperature coordinate is the total annealing temperature. Total annealing temperature, T_A, is more precisely defined for monotonic cooling histories as the temperature at which the oldest track, still present in the sample, was generated (Issler, 1996; Ketcham et al., 1999). Thus, the retention age is the age of this track. The retention age is, in most cases, inaccessible though direct dating because the shortened tracks have a lesser probability of crossing the polished surface (Laslett *et al.,* 1984; Green, 1988; Guedes *et al.,* 2004), in which the age is determined by counting tracks. The age that comes out from the density measurements is a diminished apparent age, referred to as Fission-Track (FT) age. This effect was first observed by Bigazzi (1967) through analysis of muscovite samples, in which the ages were younger than the expected. Dodson (1973) defined the closure temperature, T_C, for monotonic

cooling histories, as the temperature at the apparent age of the mineral. The FT age is directly measured and T_C can be obtained through the cooling history modeling. There are two methods to estimate the retention age, in order to compensate for thermal annealing. These methods are referred to as age correction methods: size correction method, Storzer and Wagner (1969) and the plateau method, Storzer and Popeau (1973). In the first method, the mean fission-track length reduction is used as proxy for the degree of annealing the sample experienced and the retention age is obtained increasing the FT age by the same amount. Because of this, the retention age is sometimes called corrected age (Tello et al., 2003; 2005). The size correction method is a standard procedure in the inverse modelling of thermal histories, in which theoretical length distributions and FT ages are obtained from an arbitrary T-t path and compared with the measured data. Length distributions are calculated using annealing equations, whose parameters are found through laboratory data fitting to geological time scale (Laslett et al., 1987; Crowley et al., 1991; Ketcham et al., 1999; Ketchan et al., 2007; Guedes et al., 2006; Guedes et al., 2007; Guedes et al., 2008) and the FT age is calculated relating length shortening and age reduction through a size correction curve (Green, 1988; Guedes et al., 2004; Tello et al., 2006). In this way, retention (corrected) age and T_A are natural outcomes from thermal history modeling and are implicit in every FT study in which modeling softwares (Gallagher, 1995; Ketcham, 2005; Hadler et al., 2001) are applied for data interpretation.

The plateau method is based on the assumption that track length shortening and density reduction occur at faster rates in less annealed samples. Therefore, if the sample is divided in two aliquots, one with spontaneous tracks and other with induced tracks (after pre-annealing and neutron irradiation), it is possible to find a time-temperature condition of heating, in which tracks in both samples reach the same degree of annealing. Thus, in the ratio between the track densities in the two aliquots, the effects of annealing are canceled out and the retention age is obtained.

Another useful concept for the interpretation of fission-track data is the Partial Annealing Zone, PAZ, which is the time-temperature range limited by isoannealing lines representing 10% and 90% of fission track density reduction. Isoannealing lines are, for constant heating, formed by points whose coordinates are duration and temperature of heating, giving the same length or density reduction. Estimates of T_A, T_C and PAZ are

usually obtained by extrapolation of annealing equations (Ketcham et al., 1999; Ketcham 2007; Guedes et al., 2005, 2008). The three thermal indexes are related as follows. In a time-temperature diagram, the point FT age-T_C is inside PAZ, closer to the high temperature boundary. The point retention age-T_A is outside PAZ and T_A is higher than the high temperature boundary.

It has been shown that apatite from undisturbed volcanic rocks have FT ages similar to those obtained by other isotopic techniques, which have higher closure temperatures. These results oppose the idea of age correction, because their spontaneous fission-track lengths are shorter than the induced (taken as proxy for the unannealed length) ones by approximately 10%. In direct contradiction with these results, studies carried out on induced fission tracks show that there is a linear, 1:1, relation between length shortening and density reduction up to 30% of annealing (Green *et al.,* 1988; Tello *et al.,* 2006) and indicate that even for samples from volcanic rocks the proportion is valid.

The aim of this work is to verify the validity of age correction in samples with different degrees of annealing. For this, size correction and plateau methods were applied using Population Method, PM, in apatite samples belonging to alkaline rocks from Alto Paranaíba Arch, with intrusions of Catalão (Midwest) and Tapira (Southeast), and from Ponta Grossa Arch, with intrusion of Jacupiranga (Southeast), Brazil (Gomes *et al.,* 1990). These methods were also applied to Durango apatite, belonging Cerro del Mercado, north of Durango city, Mexico, apatite samples belonging to basement rocks from Mantiqueira Mountain Range, MMR, Southwest, Brazil, in which track lengths are shortened by more than 10% (higher degrees of annealing), and to Bamble chlorapatite sample from south of Norway. Additionally, Durango apatite was also dated using the Subtraction Method, SM. In this procedure, sample does not undergo heating before thermal neutron irradiation. The goal of this experiment is to verify if this pre-annealing used in PM (to erase spontaneous tracks in the aliquot to be irradiated) has some influence in the correction by plateau method.

For apatite samples from MMR, the degree of annealing is higher than the alkaline, Durango and Bamble ones. This is because these samples have a more complex thermal history. Thus, there is no another dating technique that can provide a reference age. However, the geological evolution in the samples location is well known, given a reference

age of ≈130Ma (South Atlantic opening; Tello *et al.*, 2003; 2005; Larson and Ladd, 1973). This age will be used as reference for comparison with the FT ages obtained in this work.

Data presented by Green (1988) was carried out by using External Detector Method, EDM. According to this author, this methodology was chosen to prevent the influence of non-homogeneous uranium content. As plateau method can be only applied to samples that can be divided in two aliquots (in this case, PM and SM), the non-homogeneity in uranium content can be an important source of error. Thus, to control the amount error introduced by this source, alkaline rock samples were also dated using EDM.

Figure 1. Location of alkaline intrusions in the Ponta Grossa and Alto Paranaíba arches.

2. Method

2.1. Age Correction

Fission-track ages can be calculated, by the ϕ-method, using the following equation (Bigazzi *et al*., 1999; Iunes *et al*., 2002):

$$t = \frac{1}{\lambda}\ln\left[1 + G\frac{(\rho_S/\rho_I)}{(\varepsilon_{238}/\varepsilon_I)}\left(\frac{\lambda}{\lambda_f}\right)\left(\frac{R_U}{C_{238}}\right)\right] \quad (1)$$

In Eq. (1), λ is the ^{238}U alpha decay constant; λ_f is ^{238}U spontaneous fission decay constant; ρ_S (ρ_I) is the spontaneous (induced) fission-track surface density; G is the geometry efficiency (1 for PM and ½ for EDM); C_{238} is ^{238}U isotopic concentration; ε_{238}(ε_I) is the mineral detection efficiency, *i.e.,* the ratio between the number of observed spontaneous (induced) fission track of ^{238}U (^{235}U) per unit surface and the spontaneous (induced) fission for unit volume in the mineral; and R_U is the number of fissions by target nuclei.

For the population method, ε_{238} is usually lower ε_I because the spontaneous tracks are partially annealed during the sample geological thermal history, while the induced tracks were generated during neutron irradiation and are therefore supposed to be unannealed. The FT age (apparent age) is obtained disregarding any thermal effects, $\varepsilon_{238} = \varepsilon_I$ (Bigazzi et al., 1991). Therefore, following the usual assumption that tracks shortening is directly correlated (1:1) with density reduction (Green, 1988), FT ages are, in principle, underestimated.

Retention ages (corrected ages) are obtained by estimating the value of ($\varepsilon_{238}/\varepsilon_I$) through a curve correlating length shortening and density reduction (size correction method, Storzer and Wagner, 1969) or taking spontaneous and induced tracks to the same degree of annealing through laboratory heating (plateau method, Storzer and Poupeau, 1973). Both methods are discussed in detail below.

2.1.1. Size correction method

The size correction method was proposed by Storzer and Wagner, (1969). Aliquots of an apatite sample containing only induced fission tracks are heated for different time-temperature conditions, leading to different degrees of annealing. Mean HCFT lengths, L, and surface track densities, ρ, are measured and a curve of ρ/ρ_0 as a function of L/L_0 is built up (L_0 and ρ_0 are the mean HCFT length and surface density of tracks in the unannealed aliquot). Assuming that variation in density is caused by the variation in efficiency, $(\varepsilon_{238}/\varepsilon_I)=\rho/\rho_0$ and that the ratio L_S/L_I of the sample, where $L_S(L_I)$ is the mean spontaneous (induced) HCFT length, can be represented by L/L_0 of the correction curve, the retention age can be calculated dividing ρ_S by ρ/ρ_0 of the curve correction.

2.1.2. Plateau method

In the *plateau* method (Storzer and Poupeau, 1973), it is assumed that fission tracks, which already have some degree of annealing, for instance spontaneous fission tracks, are more resistant to additional heating than freshly induced fission tracks. Also, it is considered that does not matter how the fission tracks have been shortened but how much the current degree of annealing is. Thus, if spontaneous tracks in a piece of a given sample that experienced geological time scale annealing has a mean HCFT length of ≈14μm and induced tracks in another piece of the same sample is annealed in laboratorial treatment until presenting same mean HCFT length, both should respond in the same way if exposed to the same thermal treatment.

Thus, the plateau age is obtained making simultaneous step-heating in different aliquots containing spontaneous and induced fission tracks. This thermal treatment can be carried out keeping the temperature constant and ranging the heating time (isothermal plateau), or keeping the time constant and ranging the temperature (isochronous plateau). When the degree of annealing due to artificial annealing for the induced fission tracks is the same of natural plus artificial annealing for the spontaneous ones, the *plateau* fission-track age is reached. I.e., when the L_S/L_I ratio is ≈1 (equivalent to $\varepsilon_{238} = \varepsilon_I$), the ρ_S/ρ_I ratio tends to

a constant value. The plateau age is the age that would have been obtained if the spontaneous track population were thermally unaffected (Berter and Märk, 1983).

As the plateau method is basically the comparison between different response of spontaneous and induced fission tracks to thermal annealing, this methodology is applicable only when it is possible to separate two aliquots of the same sample. Thus, in this work, samples were dated by the Population Method (PM) and Subtraction Method (SM).

2.2. Dating methods

Hurford and Green (1982) presented five different procedures for obtaining the fission-track age (Fleischer *et al.,* 1975; Naeser *et al.,* 1979., Gleadow, 1981; Hurford and Green, 1982; Storzer and Wagner, 1982), whose principal difference is the way to count induced fission tracks. Two of these procedures are: Population Method, PM, and Subtraction Method, SM, which described below.

2.2.1. Population Method, PM

To apply PM, both spontaneous and induced fission tracks are analyzed in different aliquots of the same mineral. For this, one aliquot is firstly pre-annealed to erase spontaneous fission tracks and then irradiated with thermal neutrons. It is assumed that only one age population is present, that uranium is homogeneously distributed and there is no significant chemical composition variation in the sample to be dated. Spontaneous (ρ_S) and induced (ρ_I) mean fission-track densities are determined in the aliquots containing spontaneous and induced fission track populations, respectively. The ρ_S/ρ_I ratio gives the PM FT age.

PM should only be applied to apatite samples belonging to basement rocks. Furthermore, both spontaneous and induced track densities are measured in internal surfaces of the mineral (4π-geometry). In this case, the geometry efficiency in the eq. (1) is

1 (*e.g.,* Soares *et al., submitted-a*). In addition, for the application of PM, a large number of grains must be analyzed, *i.e.,* PM is a multi-grain methodology.

A major gain in using PM is that the initial mean HCFT length, L_0, can be directly measured. This parameter is essential for accurate thermal history modeling because it is the reference to quantify the degree of annealing of spontaneous fission tracks. The most applied dating method, external detector method (EDM), is normally preferred because single-grain ages are obtained. However, the information about the initial fission track length is lost since induced tracks are measured in a different mineral (muscovite). The value of L_0 varies significantly among different samples (Carlson *et al.,* 1999; Barbarand *et al.,* 2003; Tello *et al.,* 2006; Ketcham *et al.,* 2009) and it is generally not safe to use a standard L_0. Soares *et al.* (*submitted-a*) proposed a procedure to measure L_0 for each sample to be dated, which is basically to irradiate an extra apatite mount along with the EDM assembly.

2.2.2. Subtraction Method, SM

Subtraction Method, SM, is a variation of PM, in which the irradiated aliquot does not have the spontaneous tracks previously erased. Thus, in one aliquot only spontaneous fission-track density (ρ_S) is measured and, in the other, spontaneous plus induced ($\rho_{(S+I)}$) fission-track density is measured. The (ρ_S)/($\rho_{(S+I)}$-ρ_S) ratio gives the SM fission-track age.

An important difference between SM and PM is that, for SM, induced fission-tracks are analyzed in the non-restored structure, containing radiation (alpha recoil) and natural defects accumulated in the geological history of the mineral. For PM, the pre-annealing of the aliquot to be irradiated may restore the apatite structure, decreasing/eliminating natural and radiation defects.

3. Experimental procedure

Samples to be dated via PM were divided in two aliquots; one of them was used to analyze spontaneous fission tracks. The other aliquot was pre-annealed (450ºC during 24 hours) to erase spontaneous fission tracks and then irradiated with thermal neutrons to

induce ^{235}U fission. For the samples to be dated via SM, a non-annealed aliquot of the sample was irradiated. Thus, spontaneous plus induced fission tracks could be observed.

For Durango and Bamble apatite and samples from alkaline rocks, the plateau method was carried out using a non-annealed aliquot and three other heated respectively at 220, 290 e 340°C during 1 hour. For the basement rocks, samples from Mantiqueira mountain range only 290 and 340ºC heating experiments were carried out due to the paucity of apatite grains.

For size correction method, an isochronous curve was built up using the sample K-1, from the São Francisco Craton, Rio do Peixe, Bahia, Brazil. Temperatures for the annealing experiments ranged from 140 to 280°C. Duration of heating was 1000 hours in all experiments.

The annealing experiments presented here were performed in tubular furnaces (cylindrical) with 80 cm of length and 40 cm of diameter. The sample holders of stainless steel have a usable volume of 27 cm^3. The temperature control was done inside (internal control) and outside (external control) the sample holder through calibrated thermocouples.

The plateau ages were obtained using the ϕ-method, *i.e.,* direct measurement of neutron fluence through uranium standard glass calibrated by natural uranium thin films (Bigazzi *et al.,* 2000; Iunes *et al.,* 2004; Iunes *et al.,* 2005; Soares *et al., submitted*-b). These samples were irradiated at IPEN/CNEN nuclear reactor, SP, Brazil. To obtain the alkaline and standard apatite sample ages using EDM, the ϕ-method were carried irradiating in Thetis nuclear reactor of the Institute for Nuclear Science, Gent University (Belgium). The ratio between thermal and epithermal neutron fluences is ≈150 in the utilized position.

All apatite samples analyzed in this work were mounted in epoxy resin and had the same sanding (with sandpaper of #1200, 2400 and 4000), polishing (with diamond paste of 1 and ¼ μm) and etching. The etching was carried out in a 5% NHO_3 solution, at 20 ºC, for 55 seconds (Tello *et al.,* 2003, 2005, 2006). Muscovite plates, coupled with uranium standard glasses and apatites dated through EDM, were etched in 48% HF solution, at 15 ºC, for 90 minutes. The measurements of densities and HCFT lengths were carried out using Zeiss Axioplan 2 Imaging microscope, with nominal magnification of 1000×, dry.

The Brazilian alkaline apatite samples contain relatively low uranium concentration, particularly the samples from Jacupiranga. This low uranium content leads to low fission-track densities and, as consequence, the absence of HCFT. For this reason, an extra mount of each of these samples was prepared and irradiated with swift heavy ions to create additional channels, which increases the probability of etching confined tracks (Jonckheere et al., 2007).

4. Results and Discussion

4.1. Comparison of ages

Results for PM Plateau experiments on alkaline apatite samples are shown in table 1. Results for PM and SM Plateau experiments on Durango and PM Plateau experiments on Bamble are shown in table 2. Results for PM Plateau experiments on the basement samples from Mantiqueira mountain range are shown in table 3. Annealing experiments with MMR-1 and MMR-2 samples, at 220ºC have not been carried out due the paucity of apatite grains in these samples.

Table 1. Results for PM Plateau experiments on Brazilian alkaline samples.

Heating	N_S	$\rho_S \pm 1\sigma (x10^5)$	N_I	$\rho_I \pm 1\sigma (x10^5)$	$L \pm 1\sigma$(n)	$L_0 \pm 1\sigma$(n)	L/L_0 $l_I \pm 1\sigma$	t±1σ(Ma)
				Tapira				
Natural	602	2.93±0.12	490	2.72±0.12	14.88±0.8(22)	16.64±0.79(36)	0.89±0.06	86.9±7.0
220°C	551	2.59±0.11	357	2.67±0.14	14.64±1.2(21)	15.09±1.52(17)	0.97±0.12	78.3±6.7
290°C	250	2.07±0.13	529	1.91±0.08	14.54±1.8(17)	14.34±0.83(20)	1.01±0.14	87.4±8.1
340°C	331	1.92±0.11	280	1.73±0.10	12.94±0.4(15)	12.12±0.4(14)	1.06±0.21	89.5±8.6
				Catalão				
Natural	214	2.25±0.15	542	1.94±0.08	13.90±1.0(18)	15.9±0.95(07)	0.87±0.08	93.5±9.0
220°C	140	2.16±0.18	347	1.88±0.10	12.92±0.1(15)	15.0±1.21(09)	0.86±0.07	92.6±10.5
290°C	281	2.85±0.17	428	1.97±0.10	13.42±1.4(15)	14.0±1.43(18)	0.96±0.14	116±10.8
340°C	257	1.80±0.11	412	1.60±0.09	13.00±0.5(10)	13.5±1.40(18)	0.96±0.15	90.7±8.6
				Jacupiranga				
Natural	125	0.49±0.02	102	0.44±0.02	14.34±0.28(38)	16.5±0.13(6)	0.87±0.02	85.5±7.7
220°C	89	0.35±0.02	72	0.29±0.02	14.22±0.36(17)	14.25±0.58(11)	0.99±0.05	91.7±10.4
290°C	98	0.29±0.01	147	0.28±0.01	13.08±0.33(23)	14.14±0.25(30)	0.93±0.03	80.7±8.2
340°C	74	0.17±0.01	63	0.16±0.01	11.84±0.35(20)	12.00±0.34(23)	0.99±0.04	83.5±9.3

N_S(N_I) are the number of spontaneous (induced) counted tracks, ρ_S(ρ_I) are the spontaneous (induced) densities, L(L_0) are the mean HCFT lengths of spontaneous (induced) fission-tracks, (n) is the number of HCFT measured and t is the age in Ma.

Table 2. Results for PM and SM Plateau experiments on Durango and Bamble apatite samples.

Heating	N_S	$\rho_S \pm 1\sigma (x10^5)$	N_I	$\rho_I \pm 1\sigma (x10^5)$	$L \pm 1\sigma$(n)	$L_0 \pm 1\sigma$(n)	$L/L_0 \pm 1\sigma$	t±1σ(Ma)
Durango PM								
Natural	4617	1.91±0.03	6297	8.74±0.11	14.75±0.29(33)	16.30±0.12(81)	0.90±0.02	30.1±1.8
220°C	4403	1.89±0.03	5741	8.23±0.10	14.64±0.20(33)	14.85±0.15(90)	0.99±0.02	31.4±1.8
290°C	3932	1.69±0.03	3815	7.78±0.13	12.96±0.23(24)	13.38±0.10(104)	0.97±0.02	29.9±1.7
340°C	2938	1.27±0.02	4437	5.32±0.08	10.39±0.49(12)	10.41±0.11(52)	1.00±0.05	32.9±1.9
Durango SM								
Heating	N_S	$\rho_S \pm 1\sigma (x10^5)$	N_I	$\rho_I^* \pm 1\sigma (x10^5)$	$L \pm 1\sigma$(n)	$L_0 \pm 1\sigma$(n)	$L/L_0 \pm 1\sigma$	t±1σ(Ma)
Natural	4617	1.91±0.03	6747	9.35±0.11	14.75±0.29(33)	16.30±0.12(184)	0.90±0.02	28.1±1.7
220°C	4403	1.89±0.03	7048	8.92±0.10	14.64±0.20(33)	14.85±0.15(137)	0.99±0.02	29.2±1.8
290°C	3932	1.69±0.03	2951	7.84±0.14	12.96±0.23(24)	13.38±0.10(20)	0.97±0.02	29.7±1.8
340°C	2938	1.27±0.02	4663	5.54±0.08	10.39±0.49(12)	10.41±0.11(195)	1.00±0.05	31.6±1.9
Bamble- PM								
Natural	992	5.44±0.17	484	4.61±0.21	15.27±0.08(229)	16.58±0.12(87)	0.92±0.02	149.2±12.0
185°C	1742	5.29±0.13	1353	4.67±0.13	15.33±0.12(104)	16.34±0.15(101)	0.94±0.02	143.4±9.80
220°C	1525	5.54±0.14	1069	4.32±0.13	15.38±0.12(100)	15.84±0.13(99)	0.97±0.02	162.1±13.3
260°C	1346	5.50±0.15	1388	4.21±0.11	14.93±0.10(100)	15.51±0.17(62)	0.96±0.02	165.1±11.4
300°C	1188	4.75±0.14	1327	4.02±0.11	15.00±0.20(55)	15.14±0.16(60)	0.99±0.03	149.5±10.5
340°C	1145	4.13±0.12	1089	3.48±0.11	13.15±0.11(91)	13.50±0.13(56)	0.97±0.03	150.1±10.7
Heating	N_S	$\rho_S \pm 1\sigma (x10^5)$	N_I	$\rho_I^* \pm 1\sigma (x10^5)$	$l_{S/I} \pm 1\sigma$	$l_I \pm 1\sigma$	$l_S/l_I \pm 1\sigma$	t±1σ(Ma)

$N_S(N_I)$ are the number of spontaneous (induced) counted tracks, $\rho_S(\rho_I)$ are the spontaneous (induced) densities, ρ_I^* are the induced densities obtained by relationship $(\rho_{(S+I)}-\rho_S)$; $L(L_0)$ are the HCFT averages of spontaneous (induced) fission-tracks, L/L_0 are the HCFT mean lengths of the spontaneous plus induced fission-tracks in the induced SM aliquot, (n) is the number of HCFT measured and t is the age in Ma.

Table 3. Results for PM Plateau experiments on basement rocks from Mantiqueira mountain range.

Heating	N_S	$\rho_S \pm 1\sigma (x10^5)$	N_I	$\rho_S \pm 1\sigma (x10^5)$	$L \pm 1\sigma$(n)	$L_0 \pm 1\sigma$(n)	$L/L_0 \pm 1\sigma$	t±1σ(Ma)
MMR-1								
Natural	711	0.56±0.02	837	0.65±0.02	11.92±0.52(19)	16.41±0.22(45)	0.73±0.04	95.3±6.9
290°C	1223	0.56±0.06	1089	0.66±0.02	11.49±0.27(15)	13.38±0.26(18)	0.86±0.03	94±6.2
340°C	306	0.28±0.02	324	0.27±0.05	11.18±0.41(06)	10.82±0.36(6)	1.03±0.05	114.5±10.8
MMR-2								
Natural	1068	1.83±0.06	1102	1.66±0.05	11.22±0.13(185)	14.66±0.13(129)	0.77±0.01	121.6±8.1
290°C	1483	1.81±0.05	1276	1.50±0.04	12.05±0.15(73)	12.83±0.17(55)	0.94±0.02	133.0±8.4
340°C	914	1.33±0.04	982	0.94±0.03	9.44±0.30(12)	9.63±0.19(40)	0.98±0.03	155.7±11.7

$N_S(N_I)$ are the number of spontaneous (induced) counted tracks, $\rho_S(\rho_I)$ are the spontaneous (induced) densities, $L(L_0)$ are the mean HCFT lengths of spontaneous (induced) fission-tracks, (n)is the number of HCFT measured and t is the age in Ma.

Instead of age in the Plateau curves (Figure 2) were built up with the surface density ratio (ρ_S/ρ_I) plotted as function of the reduced fission-track length (L_S/L_I). ρ_S/ρ_I is the only part of the age equation that varies. The other quantities are physical constants and irradiation parameters, common to all aliquots, and introduce systematic deviations that can mask the variation we want to observe.

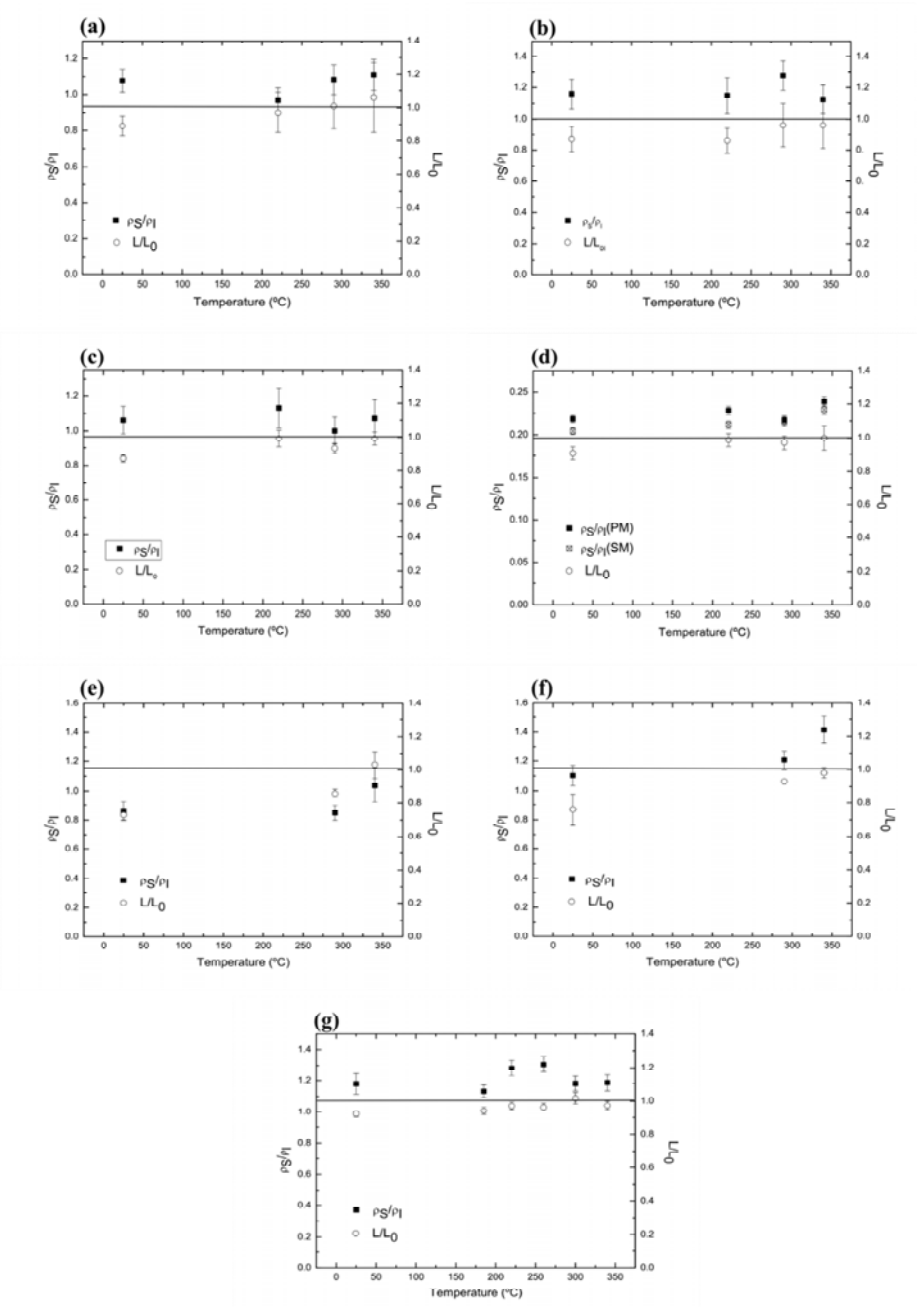

Figure 2. *Plateau* curve in apatite from alkaline and basement rocks and standard Durango apatite. (a) Tapira, (b) Catalão, (c) Jacupiranga, (d) Durango, (e) MMR-1, (f) MMR-2 and (g) Bamble samples.

The values of ρ_S/ρ_I composing Durango apatite Plateau curves, for both PM and SM, do not vary significantly (within one standard error). Therefore, for this sample, the

Plateau age (retention or corrected age) cannot be distinguished from FT age (apparent age), which is, in turn, undistinguishable from the reference Ar-Ar age of (31.4±0.2) Ma (McDowell *et al.*,2005). The Ar-Ar closure temperature is significantly higher than the fission-track one. Put together, these results support previous studies indicating that there is no age reduction in samples that experienced small amounts of annealing (Naeser and Fleisher, 1975; Green et al., 1988; Gleadow et al., 1981Jonckheere, 2003). The reduced length in Durango apatite is around 0.90 (Table 2 and Laslett et al., 1984; Green et al., 1986; Carlson et al., 1999; Tello et al., 2006). Following the same trend, Plateau curves for the apatite samples from Brazilian alkaline rocks (Figure 2) show no variation. The reduced lengths of these samples are also around 0.90 (Table 1) and, for two of them, Catalão and Tapira, the FT ages are in agreement with Ar-Ar ages (Table 5). However, the FT age for Jacupiranga apatite is significantly lower than the published Ar-Ar age. Geological evidences indicate that the evolution of the Ponta Grossa arch can be associated to post/sin-rupture of the Gondwana South-Western (Estrella, 1972; Asmus and Ferrari, 1978; Asmus and Porto, 1980; Asmus, 1981; Chang *et al*., 1992) which is characterized by the South Atlantic Ocean occurred probably at ≈130Ma ago (e.g., Renne *et al*.; 1992; Renne *et al*.; 1996; Stewart *et al*.; 1996; Ernesto *et al*., 1999). This event is characterized by numerous dikes, shear zones, faults and fractures that seem to be reactivated at shallow crust levels, especially between Upper Cretaceous and Paleogene/Neogene, which caused block tilting and normal faulting. In the Santos basin, the evidence of exhumation of the coastal mountains source area presents supply of the clastic materials. Thermochronological data present by Gallagher and Brown (1999), Tello et al., (2003), Hackspacher et al. (2004) and others reported this important event in the Brazilian southeastern passive margin in ≈100Ma.

However, the main period of deposition is ≈90Ma, which can be related to fast denudation with uplift of Ponta Grossa arch and the formation of Serra do Mar mountain range in the Albian-Aptian. (Pereira and Feijó, 1994); Zalán and Oliveira, 2005).These events can be interpreted as uplift with ≈90Ma and consequently the reactivation of main shears zones. Structural analysis of the Ponta Grossa arch, in the São Bento group (Paraná basin), identified a brittle deformation event with direction of NW, and E-W, in the period of Neocretaceous and Paleogene, as a reactivation of previous shear zone, probably formed

in the Eocretaceous Strugale et al. (2003). Almeida and Carneiro (1998) described the geologic events in the Ponta Grossa arch, in which the major rise was in the period of Upper Cretaceous, between 90 and 65Ma.

The fission-track ages determined for the Ponta Grossa arch samples may have been influenced by this stage of 90Ma, which was sufficient to erase the fission tracks in apatite, whereas other radiometric ages, that present higher closure temperature, were not affected. After this, no significant geological event was detected in the Jacupiranga apatite thermal history (Soares et al, submitted-b).This evidence is supported by Vignol-Lelarge, (1994) also applying fission-track thermochronology in samples from basement rocks from Mar mountain range in areas of Ponta Grossa arch, who concluded that at ≈86Ma the region suffered an uplift followed by erosion of 2.5 km, through a fast passage through the partial annealing zone. This uplift is associated to Mar mountain range and was also evidenced by Tello et al. (2003; 2005).

For simplest possible thermal history samples, i.e., samples formed in high temperature that were fast cooled until surface temperature fission-track ages can be associated with mineral age formation, also given by other radiometric methods. Samples from Alto Paranaiba arch give the results which support this hypothesis. On the early Cretaceous, intense alkaline volcanism, preceded the Late Cretaceous by intrusions of dikes and possible basaltic effusion (Almeida, 1983). The geological event associated to this arch, ≈70 to 90Ma (K/Ar, Amaral et. al., 1978), is in agreement with fission-track ages presented in this work.

It is worth noting that, for Durango and the Brazilian alkaline rock apatites, the reduced lengths of the unannealed samples were between ≈10 and 13%. In the first annealing experiment (220ºC), the values of L_S/L_I tend to ≈1, with the exception of sample Ca-1 (in which the value of L_S/L_I is ≈1 after the annealing experiment at 290ºC). Following the Plateau hypothesis, when L_S/L_I ≈1, the induced and fossil tracks are in the same degree of annealing. Therefore, the Plateau ages do have been obtained in these Plateau experiments.

Kolmogorov-Smirnov test was used to compare the distribution of spontaneous and induced HCFT, applied in each thermal treatment for Bamble sample dated through PM and Durango apatite samples dated through PM and SM. The results showed the same

mean lengths observed in table 2, i.e., at 220ºC during one hour of heating, the test shows that the hypothesis that spontaneous and induced fission-tracks belong to the same distribution cannot be discarded. Further details are shown in appendix 1.

Plateau ages for both MMR-1 and MMR-2 were reached only for the heating at 340ºC, where the ratios L_S/L_I are ≈1 (Table3 and Figure 2). However, differently from the samples discussed above, these ages are significantly greater than their respective FT ages. Also the reduced length of fossil tracks are around 25%, showing that these samples have experienced a heavier annealing, when compared to Durango, the Brazilian alkaline samples and Bamble apatite.

For comparison with the Plateau ages, retention ages where calculated also by the size correction method. The correction curve was plotted using K-1sample from São Francisco Craton, Bahia state, Brazil (Moreira, 2008). This curve was built up using induced fission tracks. Aliquots of K-1 apatite were irradiated at the IEA-R-1 nuclear reactor with nominal neutron fluence of 5×10^{15} cm^{-1}. Each annealing treatment was carried out by heating the samples for 1000 hours at different temperatures. Fig. 3 is the plot of ρ/ρ_0 versus L/L_0, using data presented in table 4.

Table 4. Results of the annealing treatment by plotting curve correction ρ/ρ_0 vs l/l_0 using K-1 sample from São Francisco craton, Bahia state, Brazil.

Temperature (°C)	l µm±1σ	N(l)	ρ x10^{-6} ±1σ	N(ρ)	$l/l_0$1±σ	$\rho/\rho_0$1±σ
-	16.10±0.10	50	21.43±0.67	1018	1,00±0,01	1,00±0,04
140	15.10±0.10	54	20.15±0.62	1058	0,94±0,01	0,94±0,04
220	12.75±0.12	48	17.86±0.57	982	0,79±0,01	0,83±0,04
240	10.40±0.15	49	15.41±0.49	1007	0,65±0,02	0,72±0,04
260	8.94±0.36	28	10.88±0.35	879	0,55±0,04	0,51±0,04
280	8.08±0.38	21	10.15±0.35	847	0,50±0,05	0,47±0,04

l is the mean of fission-track length, N(l) is the number of the confined fission-track measured, ρ is the fission-track density, N(ρ) is the number of counted tracks, l/l_0 is the ratio of confined fission-track annealed (l) and non-annealed (l_0), and ρ/ρ_0 is the ratio of density annealed (ρ) and non-annealed (ρ_0).

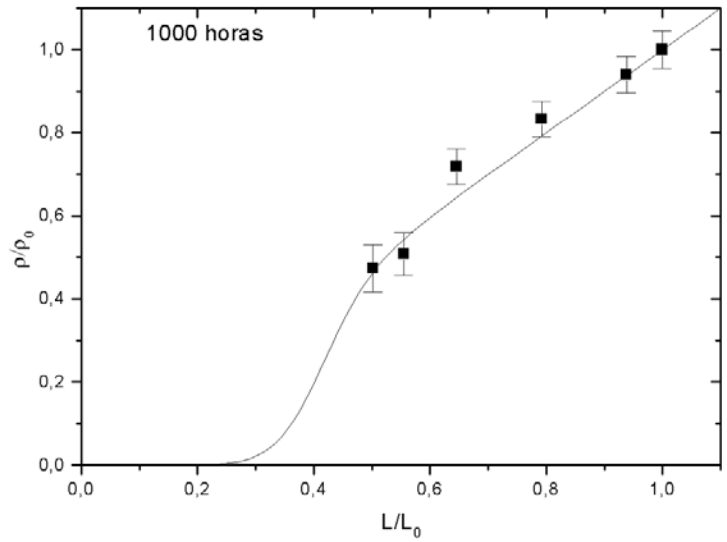

Figure 3. Size correction curve obtained through step-heating of 1000 hours with temperature at 140 to 280ºC.

The Size correction curve was fitted using the following equation (Guedes *et al.*, 2004): $(\rho/\rho_0)=(L/L_0)\{1-[1+(kL_0(L/L_0))^n]^{-2}\}/\{1-[1+(kL_0)^n]^{-2}\}$, where k=0.14 and n=8.3 (χ^2 = 1.12) are fitting parameters and L_0=(16.1±0.1)μm is the is initial mean fission track length measured for this sample. Fig. 3 shows a linear 1:1 relationship down to $L/L_0 \approx 0.60$. For more intense annealing this relationship becomes more complex and the correction is not advisable.

Table 5. Retention age obtained through FTT in undisturbed volcanic rocks and Durango apatite and published age obtained through other radiometric methods.

Amostra	T_{App} ±1σ	$T_{Corr(SizeCorr)}$±1σ	$T_{RetAge(Plateau)}$ ±1σ	t_{lit}±1σ	Reference
TA	86.9±7.0	97.5±9.1	89.5±7.0	87.2±1.2	Biondi, 2005
CA	93.5±9.0	107.3±12.5	90.7±8.6	84.0±4.0	Gomes *et al.,* 1990
Jac-1	85.5±7.7	98.2±8.0	83.5±9.3	131 ±2.0	Roden *et. al.,* 1993
Dur PM	30.1±1.8	33.4±1.8	32.9±1.9	31.4±0.2	McDowell *et al.,* 2005
Dur SM	28.1±1.7	31.3.5±1.9	31.6±1.9	31.4±0.2	McDowell *et al.,*2005
MMR-1	95.3±6.9	130.1±10.1	115±10.8	125-130	Larson and Ladd, 1973
MMR-2	121.6±8.1	157.5±13.4	156±11.7	125-130	Larson and Ladd, 1973

T_{FT} is the fission-track age, t_{lit} is the published age measured with another radiometric methods, t_{App} is the retention age obtained through size correction curve.

In Table 5, FT, Plateau, Size Correction and reference ages are compared. As discussed above, the size correction hypothesis seems not to hold for Durango, Catalão and Tapira samples, since Size Correction ages overestimate the reference ages. For MMR-1 and MMR-2, the Size Correction age agrees (within 1 deviation) with reference ages which can be associated with the age of the South Atlantic opening (Tello *et al.,* 2005; 2003; Larson and Ladd, 1973). To reconcile these results, one must accept that the annealing kinetics is not the same for both alkaline and MMR samples. It is however unclear if this difference is due to differences in chemical composition or if the samples are in different stages of a more complex annealing/etching trend. In the latter case, we would need to postulate that surface tracks are etched with constant efficiency for values of reduced length up to around 0.85-0.90. For heavier annealed samples, ages are reduced following the linear 1:1 relationship with the reduced HCFT lengths. The age correction curve sketching this model (Gleadow et al., 1981) is shown in Figure 4.

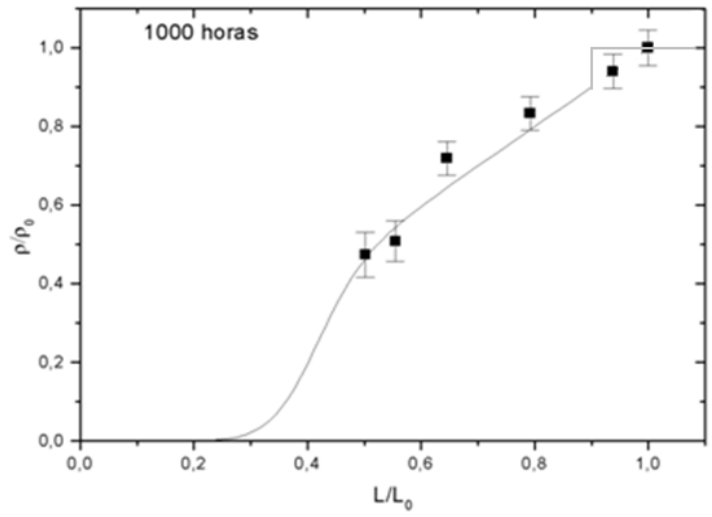

Figure 4. Size correction curve with constant density for reduced length up to around 0.85-0.90.

Table 6 shows the results of fission-track ages in Durango (Dur) and apatite from undisturbed volcanic rocks (alkaline rocks), dated using External Detector Method, EDM.

Table 6. Fission-track age determined with natural uranium thin film

Samples	**Irradiation**	N_S **(error, %)**	N_I **(error, %)**	***t* (Ma)**	***L* (µm) (*N*)**
Dur2	B	1974 (2.3)	4605 (1.5)	31.3±1.8	14.8±0,1
Dur3	A	9870 (1.0)	1343 (2.7)	26.5±1.6	14.8±0,1
Catalão	B	1460 (2.6)	1360 (2.7)	78±5	14.3±0,1 (139)
Jacupiranga	B	495 (4.5)	536 (4.3)	89±7	14.1±0,1 (182)
Tapira	B	623 (4.0)	539 (4.3)	84±7	14.2±2 (64)

1. $N_{S(I)}$ = number of counted tracks in apatite (muscovite). The error is the higher between standard and Poisson error. *t* = fission-track age calculated with fluency determined with metallic monitor (thin film).

Determined values using EDM agree statistically with determined values in the *Plateau* method using PM. This suggests that no significant error source due to non-homogeneous uranium distribution was observed. Furthermore, in fig. 5, present the HCFT distributions for Catalão, Jacupiranga e Tapira samples whose average of fission-track lengths is greater than 14 µm, consistent with fast cooling, following by exposition in a long time to ambient temperature (Green *et al.,* 1989). Catalão and Tapira samples present ages compatible with reference values. The compatibility for Jacupiranga sample is not reached even applying the size correction method.

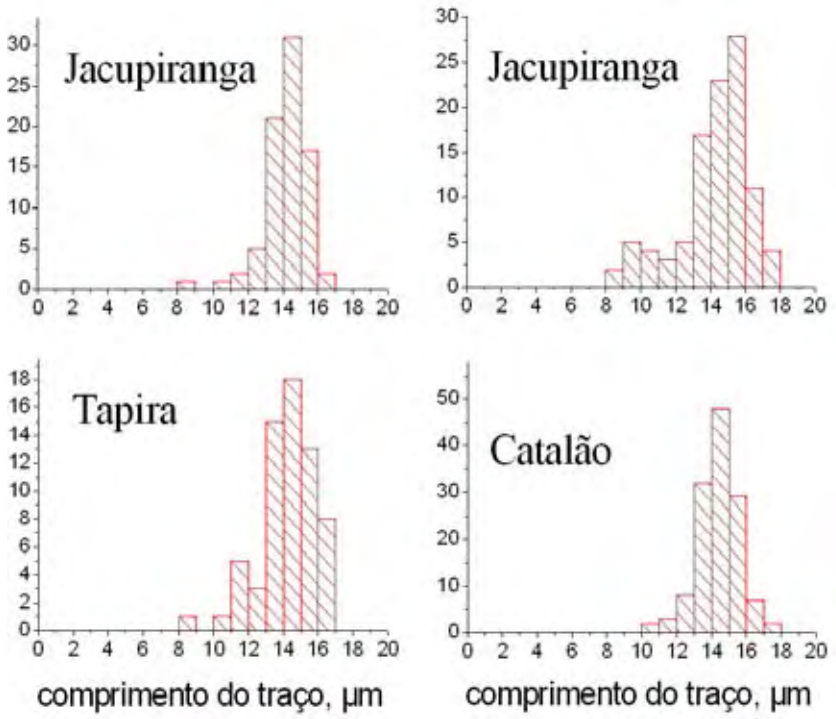

Figure 5. HCFT distributions of the alkaline samples.

4.2. Implications for the interpretation of FT data

Dodson (1973) defined the closure temperature (T_C) of a geochronological system as the temperature correspondent to the sample apparent age in a monotonic cooling history. Fission- track data is composed of the FT age (apparent age) and a HCFT

distribution. Thus, through the modeled thermal history, it is possible to obtain a temperature (T_C) associated to the FT-age. The only condition is that this history is a monotonic cooling.

Fig. 6 (a) is shows the accumulated track density plotted as a function of elapsed time for a linear cooling history of 100°C/Ma. In a first stage, all tracks are erased and there is no accumulation. In a second stage, the sample is inside PAZ and tracks start to accumulate, but the populations of tracks generated are still reduced. In the third stage, track densities in the generated populations are no longer diminished and the accumulated density is incremented of same size populations, resulting in a linear accumulation. The apparent age is the one obtained from the extrapolation of this linear behavior to zero accumulated density, i.e., it is the age disregarding the thermal effects that affected the density accumulation in stages 1 and 2. This graphic was obtained by simulation directly from the definition given by Dodson (1973). To simulate the effect of a particular thermal history, a track population has its length reduction calculated since its generation to the present day using an annealing model. If the thermal history is variable, the calculation is performed in steps using the principle of equivalent time. Then, the length reduction is related to density reduction using a curve as the one in Figure 3. Fig. 6 (b) shows the equivalent length histogram resulting from the cooling history generating the accumulated density curve.

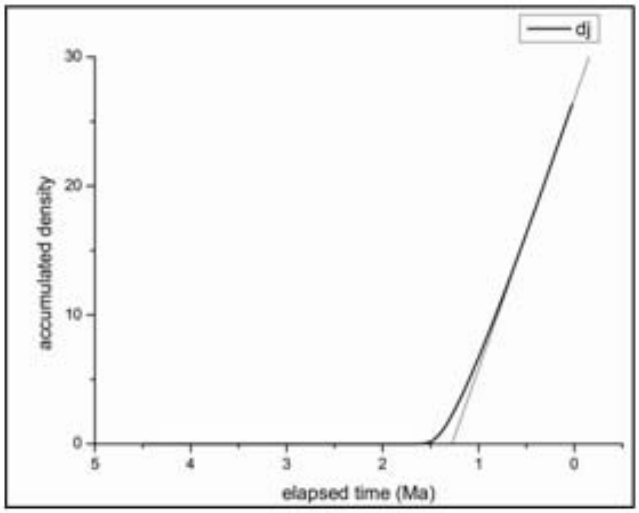

Figure 6. The graphic was simulated considerate the linear cooling (100°C/Ma).

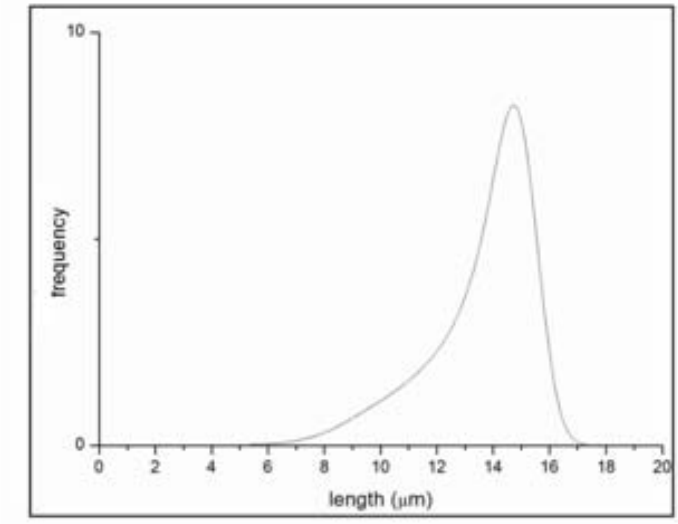

Figura 7. It is the simulated histograms which are considered a linear cooling (100°C/Ma).

In this work, we proposed a modification in the density-length reduction relationship (Figure 4). In this new version, track density is not reduced for $L/L_0 > 0.90$. Closure temperature should be affected by this change because track populations generated at lower temperatures will not be reduced, changing the FT age.

The annealing temperature, T_A definition, according to Issler (1996) is that whose fission-track population is totally erased considering a constant heating thermal history.

Ketcham *et al.* (1999) interprets T_A as the temperature at the time where remaining the oldest fission-track during the linear cooling, since the annealing models are bidirectional at the time.

Table 7 shows the FT ages, retention ages along with fission-track retention ages and its closure and retention temperatures of the simulated data. Fig. 8 shows the graphic, for the data of the table 7, of the closure temperature versus fission-track age in logarithm scale considering three different cooling degrees (0.1, 1, 10 e 100 °C/Ma). The T_C found for the sample dated in this work, considering its fission-track ages is ≈96°C for Tapira, ≈96°C for Catalão, ≈97°C for Jacupiranga and ≈104°C for Durango samples. A slight decrease is observed if the modifications introduced in this work. The new values are 92ºC for Itapira, 93ºC for Catalão, 93ºC for Jacupiranga and 100ºC for Durango samples.

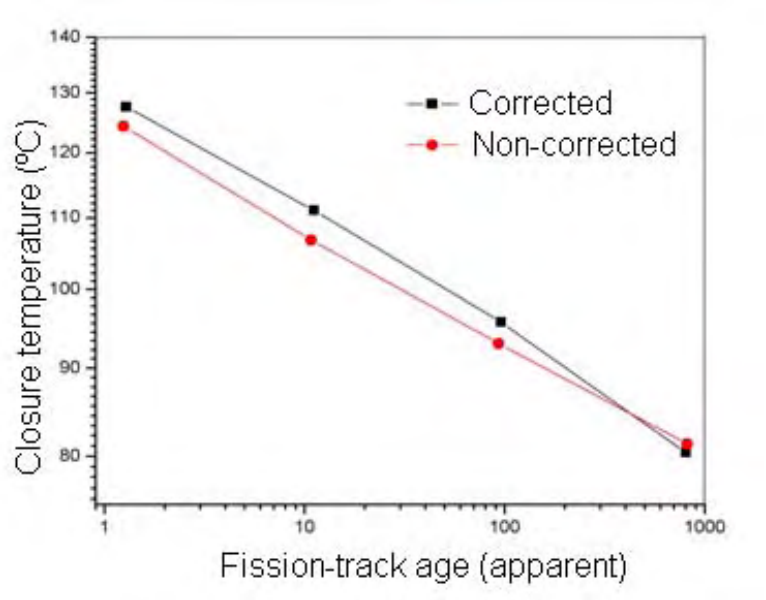

Figure 8. T_C versus fission-track age, considering cooling degree (°C/Ma) of 100, 10, 1 and 0.1, respectively.

Table 7. Presents the closure and retention temperature for the simulated data.

Resfriamento °C/Ma	t_{FT}(Ma)	T_C (°C)	t_{FT}(Ma)	T_C (°C)	t_{RA}(Ma)	T_{RA}(°C)
100	1.28	127.6	1.24	124.3	2.00	200.3
10	11.11	111.1	10.7	106.8	18.22	182.3
1	95.7	95.7	93	93	159.8	159.8
0.1	804.2	80.4	813	81.3	146.3	146.3

T_{FT} is the fission-track age, t_{RA} is the retention age, T_{RA} is the retention temperature, T_C is the closure temperature. The presented values are data generated through parameters following the Guedes et al. (submitted) model, adjusted with experimental annealing data for Durango apatite (Carlson et al., 1999).

The simulated data presented in table 7 show that, the faster cooling, the fission-track and retention ages are almost the same, as observed in data presented in this work for Durango and alkaline samples. The discrepancy becomes as accentuated as the slower the cooling rate, as the case of MMR samples.

5. Conclusion

The *plateau* and size correction methods were applied in samples from alkaline rocks, basement rocks as well as Durango and Bamble apatite. The plateau curves for low annealing degree, up to ≈10-13% present oscillations which can be associated with statistical fluctuation considering the general behavior of the curve. The fission-track age for Tapira and Catalão samples, are approximately equal to reference age. This result is also found to Durango samples for both PM and SM. For the case of low annealing degree, the behavior can be considered rigorously the same, suggesting that no age correction should be applied. For the case of MMR samples, whose annealing degree is greater than alkaline, Durango and Bamble samples, FT ages are lower than Plateau and Size Correction ones. The latter two agree with each other within the experimental deviation. Thus, two conclusions can be presented: 1) the necessity in age correction for annealing degree above 13%; and 2) both correction methods give the same results for the case where the correction should be applied. For the case of Jacupiranga sample, the presented ages are below the reference age. However, as well as low other low annealed samples, no variation in plateau curve was observed. Despite this, it is reasonable this behavior if consider the HCFT distribution which clearly is related to fast cooling.

Finally, it is discussed the meaning of fission-track ages, which can basically be defined as the age within PAZ correspondent to T_C. This value depends on the thermal history of the sample. The meaning of retention age was also presented, which can be defined as the age of the oldest observed fission-track, corresponding to T_A.

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Appendix 1:

Figures A1 and A2 show the normalized fission-track length distributions for Durango and Bamble apatite, respectively. These plots are the superposition of spontaneous (S) and induced (I) fission-track length distributions. For Durango apatite, it was also plotted spontaneous plus induced fission-track lengths (S+I) measured in induced SM aliquot. The Kolmogorov-Smirnov statistical test (Apendix A) was applied for analysis. This test can be used to investigate the relationship between two population distributions. Basically, if we have two populations, n_1 and n_2, their respective accumulative distributions S_{n1} and S_{n2} can be compared. This comparison is made through maximum distance, D_{max}, observed through experimental data, with tabulated critical value $D_{critical}$, (*e.g.,* Massey, 1951). It is also consider the degree of freedom and the adequate level of significance (α). If $D_{max} > D_{critical}$, the null hypothesis that two distribution belong the same population is rejected. On the other hand, $D_m < D_{critical}$, the hypothesis that two distribution belong the same population should be considered. If is adopted α=0.01, so, 100(1- α) =99% sure that the distributions belong to the same population distribution (Massey, 1951). Finally, it has been reported that the reasonably accuracy is reached if $(n_1 \times n_2)/(n_1 + n_2) \geq 4$. In this work, the data presented are within of this criterion. For Durango and Bamble samples, the Kolmogorov-Smirnov test was applied comparing S and I distributions. This test was also applied in Durango apatite by comparing S and I distributions with S+I. For convention, it is assumed α=0.01 as the level of significance (99% confidence that two distributions belong to the same population).

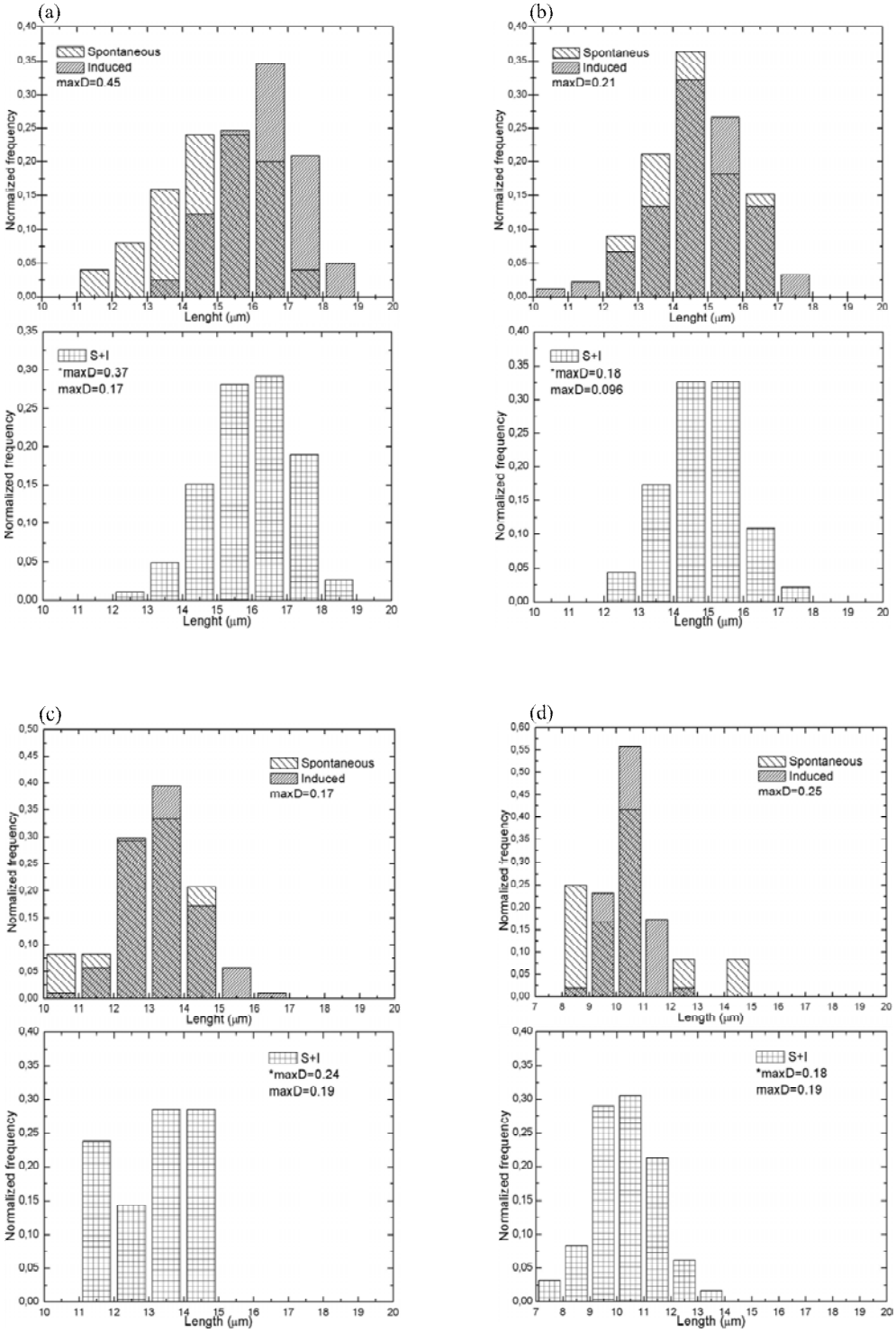

Figure A1. Graphics of the normalized HCFT distribution for Durango apatite sample for spontaneous (S) and induced (I) fission-tracks length and the distribution of spontaneous plus induced fission-track length (S+I). D_{max} is the maximum distance between normalized graphics.

For the unannealed aliquot (25ºC), it is observed a small displacement in the main peak between S and I distributions (which is about 11% of difference according to the average of fission-track length). For these distributions, $D_{19,\ 0.01}(0.45)>D_{critical}(0.36)$ and they do not come from the same population. S+I is closer to I distribution, $D_{56,\ 0.01}(0.17)<D_{critical}$ (0.22) than to S distribution, $D_{22,\ 0.01}(0.37)>D_{critical}(0.36)$. This means that the hypothesis that (S+I) and I distributions come from the same population cannot be rejected, whereas the hypothesis that (S+I) and S cannot come from the same population also cannot be rejected. This trend of S+I distribution towards I distribution is observed because there are 5 times as many induced as spontaneous tracks in S+I distribution. Therefore, for Plateau analysis, S+I distribution can be treated approximately as a I distribution. For annealing treatment of 220ºC, the main peak is exactly in the same place for S and I which is in agreement with $D_{24,\ 0.01}(0.21)<D_{critical}(0.32)$. Thus, the hypothesis that both distributions come from the same population cannot be rejected. The same assumption can be carried out by comparing S+I with S distribution, $D_{27,0.01}(0.18)<D_{critical}(0.32)$ and S+I with I distribution, $D_{54,0.01}(0.09)<D_{critical}(0.22)$. As discussed above, this point is where the fission-track correction should be reached by *plateau* method for both PM and SM.

Annealing treatment of 290 and 340ºC present the agreement in all of the analyzed cases as expected. In both cases, S and I distributions have the main peak exactly in the same place, as observed in the annealing treatment of 220ºC and $D_{20,0.01}(0.18)<D_{critical}(0.36)$ for 290ºC and $D_{10,0.01}(0.25)<D_{critical}(0.51)$ for 340ºC. Comparison between S+I with S distribution, $D_{10,\ 0.01}(0.24)<D_{critical}(0.49)$, for 290ºC and $D_{11,0.01}(0.18)<D_{critical}(0.47)$ for 340ºC, the hypothesis that these distributions come from the same population. Finally, the same hypothesis can be considered by comparing S+I with I distribution, $D_{16,0.01}(0.19)<D_{critical}(0.39)$, for 290ºC and $D_{41,0.01}(0.19)<D_{critical}(0.26)$ for 340ºC. This result supports the idea of the spontaneous and induced fission-tracks present the same behavior in annealing treatment when they present the same annealing degree. Also, there was no observed influence of the pre-annealing for MP.

The *plateau* method was applied in Bamble sample (B-2), however, in this case, a bit more annealing treatment has been carried out (table 2 and fig. 2). Unannealed and

annealing treatment of 185ºC presents basically the same ρ_S/ρ_I ratio, following by increase in 220 and 260ºC and finally a decrease in 300 and 360ºC which are approximately equals to observed in 25ºC.

Analyzing the L/L_0 ratio for B-2 apatite, it is observed the initial annealing degree up to ≈8%. This value tends to ≈1 with annealing treatment of 220ºC, and can be consider stability until 340ºC. In this point, the ρ_S/ρ_I ratio presents an increase but it is followed by decrease in the last two annealing treatments which is approximately equal to unannealed density ratio. Thus, it can be also considers a statistical fluctuation and the ratio of fission-track densities to remain constant. Thus, it is possible to consider that fission-track age cannot be corrected.

For unannealed aliquot (fig. A2), the $D_{63,0.01}(0.43)>D_{critical}(0.21)$ and can be consider that these distribution cannot come from to the same population. This context is the same for aliquots annealed at 185ºC $D_{51,0.01}(0.37)>D_{critical}(0.22)$. However, for annealed at 220ºC, $D_{50,0.01}(0.19)<D_{critical}(0.23)$ and in this case it can be considered to be the same population distribution. The statistical test for thermal treatments of 260, and 300ºC, also indicate the same population with the comparison of D_{max} and $D_{critical}$ values *i.e.*, $D_{36,0.01}(0.23)<D_{critical}(0.27)$ and $D_{28,0.01}(0.21)<D_{critical}(0.29)$, respectively. Finally, for the case of annealing treatment of 340ºC the D_{max} is the same of $D_{critica}$, *i.e.*, $D_{0.01,34}(27)=D_{critical}(27)$ and also cannot be rejected the hypothesis that these distributions come from the same population. This is assumed because in the case of B-2 sample a number of measured fission-tracks are significantly higher than Durango apatite sample and the Kolmogorov-Smirnov statistic test becomes more rigorous. Thus, these distributions indicate that correction by *plateau* method should be reached in annealing treatment of 220ºC and the fission-track densities should be remain constant.

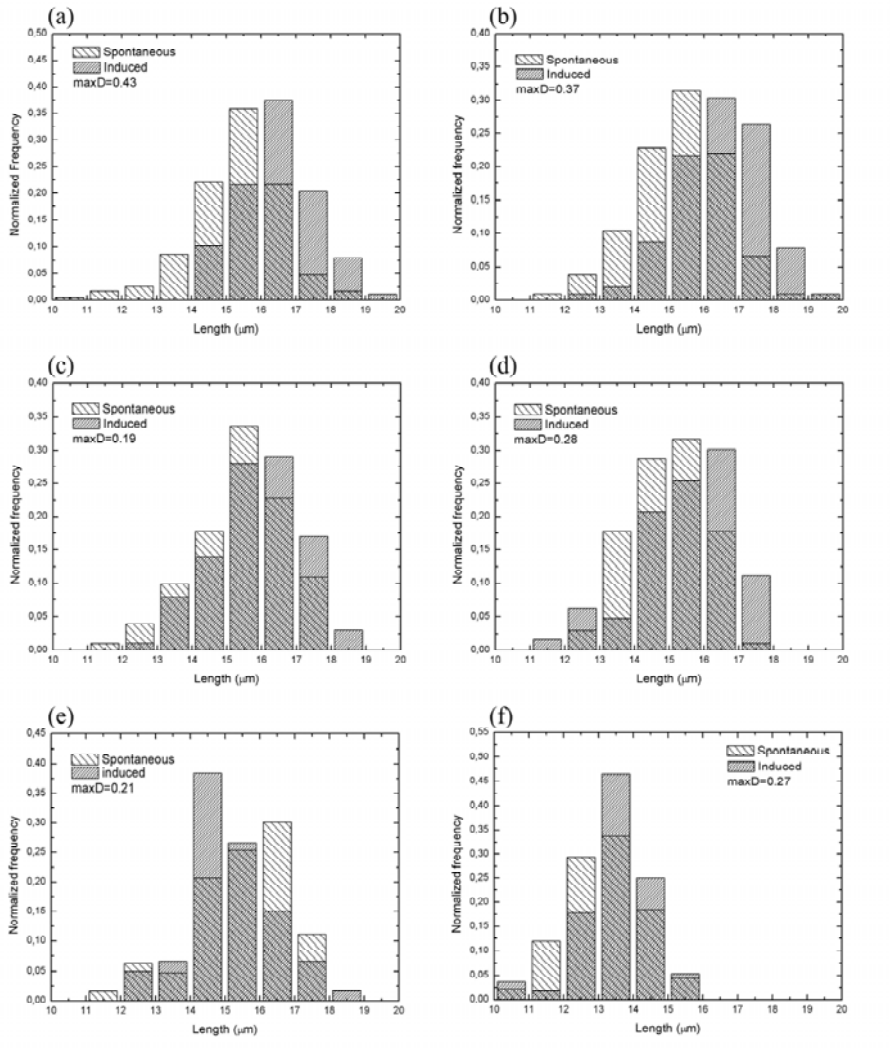

Figure A2. Graphics of the normalized HCFT distribution for Bamble apatite sample for spontaneous (S) and induced (I) fission-tracks length. D_{max} is the maximum distance between normalized graphics.

Considerações Finais.

Foram apresentados neste trabalho, três artigos que envolvem a metodologia por traço de fissão. Uma das discussões foi a respeito do fator geométrico utilizado quando o MDE é aplicado. Novos valores do fator geométrico foram determinados, através de medidas das densidades induzidas na superfície interna da apatita e na mica detector-externo, ρ_{ED}/ρ_{IS} e medidas dos traços projetados. Estas medidas estão estatisticamente compatíveis com valores documentados na literatura. Dado que, para datação pelo MDE não é possível que se tenha o comprimento inicial dos traços induzidos, L_0, é comum a utilização de um valor arbitrário. É mostrado, portanto, uma variação significativa deste parâmetro bem como a influência e a necessidade de se ter um valor de L_0 da própria amostra quando histórias térmicas são geradas.

Também foi investigado a necessidade de uma correção de idade, considerando amostras com diferentes graus de *annealing*. Para isso, dois métodos foram aplicados: a curva de correção pelo tamanho dos traços e a correção pelo método de *plateau*. Para isso, amostras de apatitas de rochas alcalinas foram utilizadas. Neste caso, como as idades destes corpos são bem conhecidos através de outros métodos radiométricos, foi possível fazer uma análise mais crítica sobre os resultados. Discussão a respeito da necessidade de correção e o significado de idade de traço de fissão e idade de retenção dos traços estão bem documentados.

Finalmente, foi apresentada a re-calibração da dosimetria de nêutrons usando filmes finos de urânio. Resultado de idades de rochas alcalinas e apatita de Durango foram comparados com métodos tradicionais de ativação metálica usando dois reatores nucleares com características diferentes frente à termalização de nêutrons. Ambas as metodologias apresentam resultados compatíveis entre si e com outros métodos de datação radiométrica.