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Kinetic model for the relationship between mean diameter shortening and age reduction in glass samples

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Abstract

Fission tracks in glass samples are shortened when they experience thermal treatment. Consequently, the observable dimensions associated with track length also diminish. The dimension normally used to characterize track size is the mean diameter of the etched track at the surface, D. Another consequence of length shortening is the reduction of surface density, ρ, implying a reduction of the fission-track age. In this work, a kinetic model is developed to describe the relationship between fission-track mean diameter shortening and surface fission-track density reduction in glass samples, based on geometrical and etching hypotheses. The result is a two-parameter equation given by

$$\frac{\rho}{\rho_0} = \left(\frac{D}{D_0}\right)^{2(1+1/n)} \left(\frac{4h^2 + D_0^2}{4h^2 + D_0^2(D/D_0)^2}\right)^2 \left(\frac{4h^2 - D_0^2}{4h^2 - D_0^2(D/D_0)^2}\right)^{1/n} \tag{1}$$

in which h is the thickness of the layer removed during etching and n is a parameter related to the latent track geometry and etching reaction rate. The equation was compared with experimental data on Australite glass found in the literature and with fresh data on Macusanite glass presented in this work. The model fits the experimental curves quite well, showing that it describes the etching effects correctly (within its limitations). In addition, the model equation derived in this work can be very useful in age correction of partially annealed glass samples.

Keywords: Glass; Etching; Fission tracks; Mean diameter; Surface density; Annealing

1. Introduction

Fission tracks are continuously generated in glass and mineral samples as a result of the passage of uranium fission fragments inside the bulk material. The fragments are released with high energy in the uranium fission, spreading electrons and even breaking chemical bonds, causing a trail of damage in the lattice structure. In the case of glasses, mainly natural volcanic glasses, there is no organized lattice, but pieces of microstructure are randomly oriented. As minerals and glasses are dielectric materials, the damaged regions are not immediately repaired and the fission tracks are relatively stable. Thus, the number of fission tracks is used to determine the time during which tracks have

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been generated in the sample. This measure of time is the so-called fission-track age (FT age) of the sample. For the purpose of dating, the sample is polished to attain a smooth internal surface (4π geometry) and etched with a suitable chemical etchant to enlarge the tracks.

Another important feature of the fission tracks is that they are shortened by the action of temperature: the higher the temperature the faster the tracks shorten. This process of track shortening is commonly referred to as “annealing”. The first immediate consequence of track annealing is the reduction of the FT age as the track lengths shorten. In minerals, the most used measure of track dimension is the length of the etched confined fission tracks parallel to the observation surface. As these tracks are fully contained in the bulk, they provide the best available information. For glasses, the surface-etched track diameter is commonly used as the main characteristic of fission tracks since confined tracks cannot be observed in this case.

Researchers have early recognized that the thermal annealing of fission tracks can be used to infer the thermal history the sample experienced since the time of its formation (Fleischer et al., 1975). For this purpose, however, it is necessary to know how tracks shortens with temperature and how etching determines the track dimensions, particularly, the track diameters in glasses. Physically, the track shortening should be described by a function of time, t, and temperature, T, $f(t,T)$ and the etching process by the etching response function, V. However, due to the lack of appropriate knowledge on track formation, annealing and etching, an empirical approach has been preferred. In this case, an empirical equation is used to fit laboratory data on etched fission track dimensions. For instance, Storzer and Wagner (1969) have used this empirical approach for glasses and Laslett et al. (1987), Crowley et al. (1991), and Ketcham et al. (1999), for apatite, respectively. The principal weakness of this method is that these empirical models have to be used in time scales much greater than the laboratory one. In case of minerals, the time interval to be considered can reach several million years for glasses and around 500 Ma for minerals while laboratory experiments are no longer than 10,000 h. For apatite, the use of the empirical models has been validated by geological evidence found in borehole sample data from the Otway Basin (Green et al., 1989). However, a recent work (Jonckheere, 2003) has shown that data from another borehole, the KTB, are not compatible with the empirical models, which casts doubt on the validity of this approach.

On the other hand, there are few elements to develop physical models. Just data on etched tracks are available. In this way, it is necessary to better understand the evolution of the effect of etching on the observable dimensions of the fission tracks as the annealing proceeds. A suitable laboratory for this study is the relationship between fission-track density reduction and the shortening of the observable dimensions of the fission tracks, since this relation does not depend on the particular thermal history the track experienced. It just depends on geometrical relations and etching effects. Guedes et al. (2004) developed a kinetic model which succeeded in explaining the above-mentioned relationship through etching effects for apatite, zircon and titanite data found in the literature. The present work is aimed at showing that this approach is also applicable for glasses. A two-parameter kinetic model relating fission-track surface density and mean track diameter reduction in glasses is developed and fitted to the literature and fresh data. Besides studying etching effects, an analytical expression can be very useful for correcting ages in glass samples that have experienced thermal annealing.

2. The model

Consider a glass sample polished to attain an internal surface and properly etched to enlarge the fission tracks. The number of tracks crossing the surface per unity area gives the surface track density, ρ. Suppose that just one population of tracks with mean etchable track length l is present. After etching, just tracks with incidence angle (measured from the observation surface) greater than the critical angle, θ_C, can be revealed. As glasses are isotropic materials, i.e. there is no preferential etching direction, the following relation holds without restriction (Wagner and Van den haute, 1992):

$$\rho = \varepsilon N_0 l \cos^2 \theta_C(l) \tag{2}$$

in which ε is the observation efficiency and N_0 is the number of fissioned nuclei per unity volume. In Eq. (2) it has been assumed that the critical angle is not constant, but dependent on the characteristic track length. Such an assumption is based on the fact that insofar as the annealing increases the amount of ionization of the tracks diminishes and the etching efficiency must change. In glasses, however, the measurable characteristic quantity is the mean diameter, D, of the fission tracks at the surface. Fortunately, it has already been shown (for instance, Somogyi and Szalay, 1973) that the track diameter is related to track length. In this way, the ratio between ρ and a reference density, ρ_0, associated to a reference diameter, D_0, is given by

$$\frac{\rho}{\rho_0} = \frac{l}{l_0} \frac{\cos^2 \theta_C(D)}{\cos^2 \theta_C(D_0)}. \tag{3}$$

In terms of the response function, V, defined as the ratio between the track etching velocity and the bulk etching velocity, the critical angle is (Somogyi and Szalay, 1973)

$$\sin \theta_C = V^{-1}. \tag{4}$$

Using Eq. (4), Eq. (3) becomes

$$\frac{\rho}{\rho_0} = \frac{l}{l_0} \frac{1 - [V(D)]^{-2}}{1 - [V(D_0)]^{-2}}. \tag{5}$$