Terrestrial Ages of Antarctic Meteorites Based on the Thermoluminescence Levels Induced in the Fusion Crust

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The fusion crust of eight Antarctic meteorite finds show natural thermoluminescence (TL) levels about 100 times higher than the levels in the fusion crust of freshly fallen meteorites, Dhajala, Jilin and Bansur. If it is assumed that this TL is due to cosmic ray received on the surface of Antarctica, the terrestrial residence times of the meteorites is calculated to lie between 10^4 - 10^5 years. Strictly, these periods represent lower limits of terrestrial ages of these meteorites, and are generally consistent with terrestrial ages calculated from cosmogenic radionuclides.

I. Introduction

The interior material of a chondrite typically has about 2000 Gy dose equivalent of natural thermoluminescence accumulated due to ambient cosmic ray irradiation in the interplanetary space. The cosmic ray dose rate near 1 A.U. is estimated to be about 100 mGy/a. This TL, particularly the low temperature (< 250°C) component fades after the fall of the meteorite on the earth by natural recombination processes at the ambient temperature in the absence of any radiation dose. This slow fading of natural TL, TL (NTL), has been used to estimate the terrestrial ages of chondrites[1−3] Since all the chondrites do not have identical NTL at the time of fall, because of its dependence on perihelion distance and extent of shock experienced by the meteorite, coupled to the variability of anomalous fading rates on the earth, this method leads to terrestrial age estimates which have been found not to be precise, and sometimes unreliable[2,3].

The meteorites undergo severe frictional heating of their surface during their passage through the earth's atmosphere, resulting in the formation of fusion crust. It is estimated that the temperature of the surface exceeds 1000°C, resulting in vapourisation of surface material and melting, and recrystallisation of material just below the surface, upto several millimetres[4] The fusion crust is usually sub millimeter and occasionally 2 millimeter thick, but the heat conducts down to several centimeters in favourable cases[5,6] where NTL is partially erased. The fusion crust and the material just below it are thus expected to have no natural thermoluminescence at the time of fall.

During the past decade, many meteorites have been recovered from the Antarctic ice shelf where they have been found lying on the surface of ice. Cosmic ray dose on the polar ice cap is appreciable and can induce TL in the fusion crust. Ninagawa et al.,[6] found measureable amount of natural TL levels in fusion crust of an Antarctic meteorite and related this to the terrestrial age of the meteorite. We have studied the TL characteristics of the fusion crust of several meteorites and describe here procedures for determining TL levels which, in tum, give their residence time on the surface of Antarctic ice sheet, based, on the TL build up in the fusion crust since their fall.

The terrestrial age of meteorite finds is an important parameter in many such studies such as evaluation of the frequency of fall of various meteorite types, its time variation and distribution on the earth. This information, in turn can be used to understand the meteorite injection mechanism in earth crossing orbits in the interplanetary space. Furthermore, the terrestrial ages of Antarctic meteorites are useful in understanding the ice dynamics and the mechanism of meteorite concentration on the ice shelf. It is for these reasons that lot of efforts have been made to determine the terrestrial ages of Antarctic finds[1−3,7−11]

Apart from fading of low temperature natural TL, a method commonly used to determine the terrestrial ages is the extent of decay of some cosmogenic radionuclides such as ^{14}C, ^{26}AI, ^{10}Be and ^{53}Mn [7−10], compared to their expected production in the interplanetary space. However since isotope production rates depend on size of the meteoroid, shielding depth of the sample and the degree of saturation of the radioisotope, these estimates are usually accurate within a factor of 2. Some special procedures for more accurate estimation of terrestrial ages using several isotope pairs have been suggested[11] but since the terrestrial ages of most Antarctic meteorites (1 to 100 Ka) is small compared to the mean life of many of these radionuclides used [T(^{26}Al) ~1 Ma, T(^{10}Be) ~2.5 Ma and T(^{53}Mn) ~5 Ma] the error in computation of the terrestrial ages is generally large.

The fusion crust method based on accumulated TL on the earth may provide a reliable alternative method of estimating terrestrial ages. In view of this possibility eight Antarctic meteorites have been studied in detail and merits and limitations of this technique are discussed here.

II. Principle and methodology of fusion crust dating

There are two basic assumptions in dating fusion crust by TL method:

(i) The interplanetray TL is totally removed from the fusion crust during transit through the earth's atmosphere. Although this is expected because of high temperatures attained in the atmosphere, experimental data on fresh falls supporting this assumption will be discussed later.

(ii) A regrowth of TL in the fusion crust, occurs subsequent to fall due to irradiation from the ambient radiation environment i.e. cosmic rays and natural radioactivity from the surrounding media (ice in case of Antarctic meteorites and soil in case of other finds).

The terrestrial age equation may be written as

$$T_E = \frac{\text{TL acquired after the fall}}{\text{Annual rate of TL acquisition}}$$

or

$$= \frac{ED}{aD_\alpha + (D_{\beta\ \text{int}} + D_{\beta\ \text{ext}}) + D_\gamma + D_c}$$

where ED represents total accumulated "equivalent dose" and D is the dose rate. Subscripts α, β_{int}, β_{ext}, γ and c denote dose rates due to alpha particles, inherent beta radiation due to meteoritic material, beta radiation from the external medium, gamma radiation and cosmic rays respectively. Thus the estimation of the terrestrial age of the meteorite requires two inputs (i) TL acquired since the fall of the meteorite and (ii) the annual dose rate.

II.1. Annual dose in Antarctica

The radioactivity of the chondritic material is extremely low (Table 1) and therefore the internal dose is negligible. The Antarctic ice and snow also have low radioactivity (K= 0.4 ppm[12,13]), thus the dose due to self and ambient radioactivity can be neglected for the present purpose. The main source of radiation is the cosmic ray secondaries and because of high altitude and low ridigity cut-off near the poles, it is quite significant. From the data given by Herbst[14] and Prescott and Stephan[15] we estimate that, at the latitude of 70°S and altitude of 1500-2000m where most of the meteorites studied here have been found, the annual dose is in the range of 0.6 to 0.8 mGy/a. The relevant data for each meteorite studied is given in Table 1, which indicate that the dominant radiation component on Antarctica is due to cosmic rays and all other sources are negligible. In view of these data, the age equation (1) simplifies to

$$T_E = ED/D_c$$

Only in the case of meteorites kept in buildings, the dose due to ambient radioactivity is significant. In such cases i.e. Bansur, Dhajala and Jilin, we use the total dose rate.

The annual dose rate values given in Table 1 refer to the surface of the ice cap. Meteorites, however, do not spend all their time on the surface but, soon after their fall, get buried under gradually increasing over burden of ice and move with the flow of ice from the region of fall to the region of recovery. Therefore the terrestrial residence time T_E has two components, the surface residence time T_S and the residence time while they are deeply buried in ice T_D so as to have been shielded by cosmic rays.

$$T_E = T_S + T_D$$

Once the meteorites arrive in the ablation zone, they remain on the surface for ever. Ice velocities measured in Allan Hills area yield horizontal velocities of ~1 m/a[16], The maximum concentration of meteorites has been found where ice becomes static. In this region the catchment area has a radius of ~ 5 km and thus the meteorites may be spending about 5,000 years buried within ice. In comparison, the exposure ages have values up to 700,000 years. The dynamic considerations thus suggest that $T_D < T_S$ and therefore we may here assume $T_E \sim T_S$.

III.1. Sample description

Surface chips containing fusion crust from eight Antarctic meteorites collected from Allan Hills, Meteorite Hills and Mount Baldr were analysed. For the sake of comparison recently fallen meteorites with known terrestrial ages namely Dhajala, Jilin and Bansur were also studied. Eight of these are chondrites and the remaining three belong to diogenite, ureilite and shergotite groups. Except for ALHA 77257 and META 76001 where the crust was slightly weathered all the other meteorites had well developed and preserved fusion crust[17]. Small samples containing a few square millimeters of fusion crust were chipped off with the help of a water cooled diamond blade. The interior (meteorite) material was removed by grinding on an emery paper (grit size = 400) till a uniform black chip (typically ~2-3 mm wide and a few hundred microns thick) was obtained. The chip was polished in subdued red light on all sides to remove material exposed to natural light and subsequently washed in an ultrasonic bath and crushed. The powder was treated with lN HCl to remove any weathered material. The mineral grains in the size range of 45-110 microns were deposited on stainless discs with silicone spray.

III.2. Experimental details

The thermoluminescence signal was read using a photon counting system and the detection optics consisted of a Corning 5-58 filter and a Chance Pilkington HA3 filter coupled to an EMI 9635 QA photomultiplier tube. TL glow curves were recorded in an ambient atmosphere of ultra-pure nitrogen. The heating rate was maintained at $5^oC\ sec^{-1}$. The system and the procedure are described in detail by Singhvi et al[18].

The acquired Equivalent Dose (ED) was estimated using the second glow normalisation method. This method is useful when the amount of sample is limited and TL sensitivity of the material is low as in the present case. In this method the ED corresponding to the TL intensity of a virgin sample is estimated by scaling it by the TL intensity induced in the same fraction by a known test dose. Since the ED values depend slightly on test dose, due to some non-linear processes, two test doses were used in the present work: ~9 Gy (low dose) and 500 Gy (high dose). In case of ALHA 76008 and ALHA 77282 the acquired dose were estimated by additive dose method as well. In this method, several identical discs made from the same virgin sample (having their NTL) are given different beta irradiation (0 to 100 Gy). The beta irradiations were made using a ^{90}Sr - ^{90}Y plaque. Weight normalisation was done for different discs and typical disc to disc variation in the TL signal was 10-15%. A plot of the TL level of these discs versus the given dose was made and the intercept on the dose axis gave the natural dose [ED]. As shown in Table 2, the acquired dose computed from additive beta method and the value based on low dose and high dose irradiations agree within ±30 %. This is the dominant source of error in the age estimation.

TABLE 1. DOSE RATE DATA FOR ANTARCTIC METEORITES AND RECENT FALLS

METEORITE	CLASS	Latitude, Longitude	Altitude (M)	U[1] (ppm)	Th[1] (ppm)	Internal Dose rate (mGy/a)	K(%) in ambient environment	Gamma Dose rate (mGy/a)	Cosmic Dose rate (mGy/a)	Total D_T (mGy/a)
BANSUR	L6	27°42'N,76°20'E	65	0.014	0.042	0.014	1.5	1.62	0.26	1.89
DHAJALA	H3	22°22'40''N 71°25'N38''E	65	0.011	0.041	0.012	1.5	1.62	0.26	1.89
JILIN	H4	44°N,126°30'E	65	0.011	0.041	0.012	1.5	1.62	0.26	1.89
ALHA 76008	H6	76°45'S,159°20'E	2000	0.011	0.041	0.012	0.00004	$4.3x10^{-5}$	0.79	0.80
ALHA 77256	DIO	76°45'S,159°20'E	2000	0.007	0.015	0.006	0.00004	$4.3x10^{-5}$	0.79	0.80
ALHA 77257	UREI	76°45'S,159°20'E	2000	0.0005	0.003	0.0007	0.00004	$4.3x10^{-5}$	0.79	0.79
ALHA 77231	L6	76°45'S,159°20'E	2000	0.014	0.042	0.014	0.00004	$4.3x10^{-5}$	0.79	0.80
ALHA 77231	L6	76°45'S,159°20'E	2000	0.014	0.042	0.014	0.00004	$4.3x10^{-5}$	0.79	0.80
ALHA 77282	SHERG	76°15'S,156°30'E	2000	0.11	0.47	0.132	0.00004	$4.3x10^{-5}$	0.79	0.92
EETA 76001	L6	77°35'S,160°19'E	2000	0.014	0.042	0.014	0.00004	$4.3x10^{-5}$	0.79	0.80
META 78028	L6	79°41'S,155°45'E	1500	0.014	0.042	0.014	0.00004	$4.3x10^{-5}$	0.62	0.63

[1] Representative values of the meteorites class

TABLE 2. TL DATA AND THE ANTARCTIC SURFACE AGES ESTIMATED BY THE FUSION CRUST METHOD

Meteorite	Acquire Dose (Gy) second glow normalization		Acquire Dose (Gy) Additive beta	TL Age T_s (years)	Cosmogenic Age T_E (years)
	Low Dose	High Dose			
BANSUR	0.61	0.71		323	96+
DHAJALA	-	0.16		85	11+
JILIN	0.35	0.36		185	11+
ALHA 76008	8.37	5.7	9.48	$1.0x10^4$	$3x10^4$
ALHA 77231	43.3	37.0		$5.4x10^4$	$3.1x10^4$
ALHA 77256	-	42.0		$5.3x10^4$	$1.1x10^4$
ALHA 77257	-	123.0		$1.5x10^5$	$3.7x10^4$
ALHA 77282	21.2	17.5	20.76	$2.6x10^4$	$3x10^4$-$1.6x10^4$
EETA 79001	-	57.0		$6.2x10^4$	$3.2x10^5$
MBRA 76001	17.1	14.0		$2.1x10^4$	$3.2x10^4$
META 78028	-	67.0		$10.6x10^4$	$3.3x10^4$

+ known ages

In view of the fact that the measurement of the TL levels could result in sensitivity or reflectivity changes, a few samples were repeatedly irradiated and read out. The results show that changes in TL sensitivity or reflectivity are $< 5\%$. The glow curves obtained for ALHA 77282 are shown in Fig. 1. The glow curves show a low temperature peak at ~250°C and a broad structure in the high temperature region as is typical of most ordinary chondrites[19−20].

IV.1. Suitability of the fusion crust for TL dating

As the fusion crust consists of a deep glassy zone, which are suspected of exhibiting the phenomena of anomalous (non-thermal) fading, they were studied for TL fading behaviour and also for sensitivity. The TL sensitivity was found to be 10-50% of the value of the interior material of the meteorite but the light levels were adequate for reliable estimation of dose except for recent falls. The thermal decay and the presence of anomalous fading was examined after irradiating the discs with a known test dose followed by measurement of TL level for storage periods up to 100 days at room temprature. These were then normalised to the TL levels from the same disc read out immediately (after ~2 minutes). Fig. 2 gives the fading plot for Dhajala and ALHA 77231 which suggests negligible fading at high temperatures (> 360°C). It was found that most of the fading occurs in the first day and subsequent fading is negligible. Even extrapolation over the terrestrial ages of meteorites (10^3-10^6 years), the anomalous fading may be negligible. Thus it can be concluded that the fusion crust is quite suitable for dating.

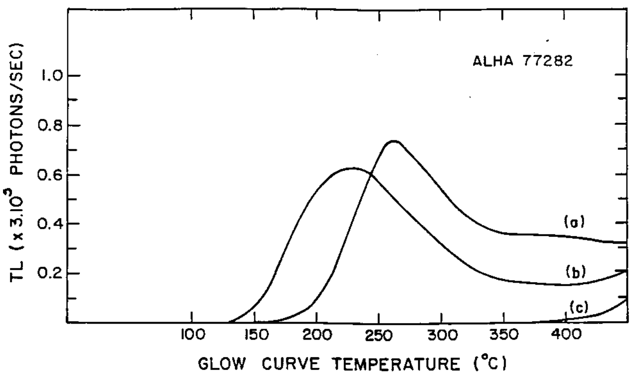

Figure 1. The (a) natural, (b) beta induced and (c) the black body curves for ALHA 77282 fusion crust sample.

IV.2. Resetting of TL during the formation of fusion crust

As discussed above a basic assumption using the natural TL levels (NTL) for the estimation of terrestrial ages is that it is completely erased during the formation of the fusion crust. Hence the NTL levels in the crust of freshly fallen meteorites should be small or close to zero. The acquired dose (ED) estimates for various meteorites are summarised in Table 2 which indicate that recent falls like Dhajala and Jilin chondrites ($T_E \sim 11$ years) have low NTL values corresponding to low ED values. The ED values when coupled to the indoor dose rate of 1.6 mGy/a[21] provide upper limits to their ages of 85 and 185 years respectively, compared to the known ages of 11 years. Even for Bansur (fell 1892), the estimated age is 3.5 times the actual terrestrial age. The discrepancy is due to the fact that the dark current of the photomultiplier tube used is appreciable, giving about 100 counts/sec and the signal for such low ages of meteorites (11-96 years) are too low to be measured accurately. We need a minimum signal corresponding to ~ 4 Gy to measure the TL levels properly. From this discussion, it is clear that the fusion crust method is not suitable for dating meteorites with terrestrial ages lower than about 500 years but the accuracy improves as the terrestrial age increases.

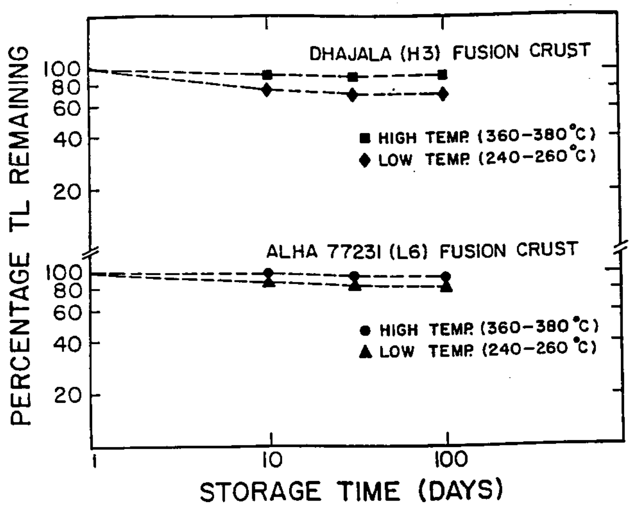

Figure 2. The fractional TL remaining in the fusion curst samples of Dhajala and ALHA 77231 for storage periods of ten, thirty and hundred days respectively kept at room temperature.

IV.3. Estimation of acquired dose

In order to evaluate possible errors in estimation of the acquired dose it has been calculated in three different ways in two of the meteorites (Table 2) . The estimates at low doses and the dose calculated from additive beta method agree within 10% but the estimate based on high dose is lower by 15-30%. The differences seen for low and high dose may possibly be because of non-linear growth of TL. Considering these difficulties and the stability of the TL signals at low temperatures as well as to avoid the effect of black body contribution at high temperatures, the ages have been computed in the stable plateau region of 360-380°C as shown in Fig. 3.

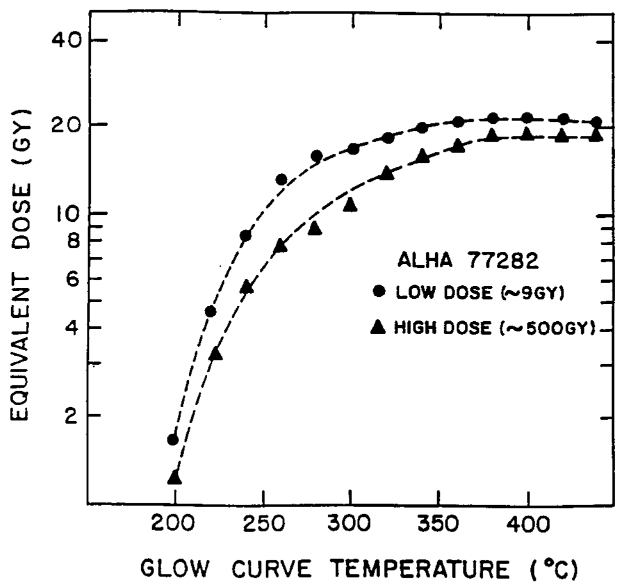

Figure 3. The variation of acquired dose with the glow curve temperature for ALHA 77282 fusion crust at low dose (~9 Gy) and high dose (~500 Gy) used for normalisation.

IV.4. Surface residence times of Antarctic meteorites

Using the comic ray dose rate at the Antarctic altitudes and geomagnetic latitude we have calculated T_S for the eight Antarctic meteorites. The ages have been calculated obtained based on the low dose values wherever available. The values range between 10,000-100,00 years and are summarised in Table 2. The overall error in the ages are estimated to be ±20%.

IV.5. Comparison with other methods

In case of some meteorites, terrestrial ages based on cosmogenic isotopes are available[7,8,22]. These values are given in Table 2 for the sake of comparison. It is seen that the T_S based on TL is of the same order as the T_E based on cosmogenic radionuclides. T_E based on the cosmogenic radionuclides (eg. ^{26}Al, ^{53}Mn and ^{14}C) is independent of the burial history of the meteorite whereas the TL method gives only the surface residence time T_S. Apart from this reason, the differences may be due to errors in estimating the ages by the two methods. When more accurate estimates are available, by studying terrestrial ages from cosmogenic radioisotopes and fusion crust TL, one can determine the surface and burial residence times. These data can thus be also useful in estimating long term ice flow rates. From the data presented in Table 2, it appears that the fusion crust method offers a rapid method of estimating T_S with reasonable accuracy.

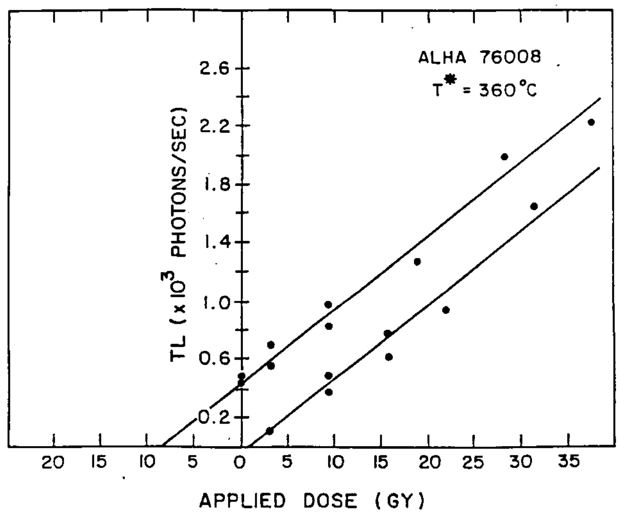

Figure 4. The additive beta growth and the supralinear correction for the ALHA 76008 fusion crust sample at a glow curve temperature of 360°C.

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