LOOKING BACKWARDS TO LOOK FORWARDS: A FIFTY-YEAR EXPERIMENT IN THE KINETICS OF THERMOLUMINESCENCE OF LUNAR SAMPLES AND THE APOLLO NEXT GENERATION SAMPLE ANALYSIS PROGRAM (ANGSA).

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Introduction: When the Apollo samples were returned there was considerable interest in their thermoluminescence (TL) and related cathodoluminescence properties (CL) [1-5]. This interest was prompted in some part by the observations of light being emitted by the lunar surface, the so-called "transient lunar phenomenon" (TLP) [6]. It was quickly realized that sample luminescence was not efficient enough to explain the TLP [6a] and interest turned to using TL to estimate radiation and temperature related processes, like radiation profile [7] and temperature profile [8] in the regolith, surface temperatures of the regolith [9] and duration of boulder emplacement [10]. For quantitative purposes, these applications require a knowledge of the kinetic parameters of the thermoluminescence process, in particular the activation energy (*E*, also referred to as the "trap depth" in reference to the band energy model for luminescence) and Arrhenius factor, *s*, describing the probability of a critical part of the process [e.g. 13]. These parameters are poorly known and the earlier work relied on laboratory experiments to determine *E* and *s* kinetics lasting minutes or hours. In more recent years, it has been proposed that TL measurements (or related luminescence measurements) could be the basis of an instrument for in situ determination of sediment (ejecta) emplacement on the Moon (and Mars) [11,12]. While *E* and *s* values are not required for this application, an improved quantitative understanding of the decay process would surely help.

NASA recently decided to make several new samples from the Apollo 17 mission available and these included 17 soil samples that had been stored in a freezer at -20ºC, whereby 6 of them were recovered from a drill core up to 291 cm in depth. Thus we have an almost fifty-year experiment in which some regolith samples were stored at -20ºC while splits were stored at room temperature. We have a unique opportunity to observe and quantitatively determine the low-temperature decay of characteristics of thermoluminescence.

Samples: The samples available and some information are listed in Table 1. Apollo 17 landed in the Taurus-Littrow valley (TLV) and these regolith samples were taken in the shadow of boulders on the north and south side of the valley at stations 6 (EVA 2) and 2 (EVA 3). We also include a sample of a representative boulder collected near the landing site (Station 1A).

Table 1. Samples available for this experiment

Sample	Wt (g)	Ref
Rock, boulder Station 1A		
71036,0	118.4	14
Permanently shadowed soil Station 2		
72320,1	18.91	
72320,2	6.622	15
72320,4*	0.131	
72320,5	0.035	
Permanently shadowed soil Station 6		
76240,5	19.67	
76240,6	4.523	
76240,22*	0.099	15
76240,33	0.025	
76240,34	0.04	

* These samples were the subject of earlier TL measurements [9]

Measurements: The unique value of these samples is that they enable time-dependent low-temperature processes to be measured with a precision not previously available.

Anomalous fading. One of the first observations upon the return of samples from the Moon was that the Apollo samples all showed anomalous fading [16], a phenomenon first reported for terrestrial volcanic rocks [17]. This is the uniform loss of signal at all heating temperatures, contrary to classical TL theory that predicts that loss of signal decreases with heating temperature. While theories exist for explaining this phenomenon, very little is actually known. Based on short term experiments at elevated temperatures higher the fraction remaining, *f*, after time *t* is given by the expression [18]:

$$\text{Log}(f) = m\,\text{Log}(t) + c$$

The new samples enable us to measure the amount (if any) anomalous fading at low temperatures and relatively long durations.

Thermal fading: The fading of natural TL can be described by the equation:

$$dI/dt = -s\,I_o\,\exp(-E/kT)$$

where E and s were defined earlier, I is TL intensity, t is time and T is temperature. There are multiple trapping sites in silicate samples, each with a characteristic E and s, but to date all seem to be second order. E and s can be determined by laboratory measurements [13] but uncertainties are large due to overlap between the TL from multiple sites. Curve fitting is another approach, but it is not clear that this provides unique solutions [19]. A fifty-year experiment will enable direct measurement of E and s over a much longer time than previously available.

Applications: The advantage of refining E and s values for lunar samples enables us to provide more accurate and more reliable temperature estimates of the sample and sample locations history over the last several million years. Since natural TL process is directly related to the radiation environment, parameter refinement will also tell us about the radiation environment over the same time span.

The natural TL of Apollo 17 sites has previously been only measured for two permanently shadowed soils, we will revisit the obtained temperature measurements [9] with the refined parameters and more detailed understanding for the TL process itself. We also will apply the newly derived parameter to obtain novel temperature estimates of the remaining Apollo 17 samples to understand the thermal history of the TLV.

The expression for the equilibrium level of natural TL, where loss of TL due to thermal draining in is balanced by build-up due to irradiation, is:

$$\phi = \phi_s / \{1 - [s / \alpha R \exp (E/kT)]\}$$

where ϕ (Gy, absorbed dose) is the level of natural TL, ϕ_s (Gy) is the value of TL at saturation, α is the reciprocal of the mean dose (the dose to fill 1/e of the traps, Gy^{-1}), R is the dose rate (Gy/s), and k is Boltzmann's constant (eV/K). The dose observed is determined from I by laboratory calibration while saturation dose, and α are determined from laboratory measurements. Durrani et al [9] rearranged the equilibrium expression to determine the temperature of the surface of the Moon under boulders at the Northern and Southern Massif at the Taurus-Littrow region of the Moon. Additionally, by the dividing the dose absorbed, ϕ, by the dose rate, R, it is possible to determine the time taken to reach equilibrium which is the minimum duration of the shadowing.

Implications: Ultimately, studies of the natural TL of future Artemis returned samples will inform us about the thermal and radiation history of the next generation of samples returned from the lunar surface. Especially with regards to in situ resource utilization (ISRU) for a sustainable human presence on the lunar surface, understanding the thermal and radiation history is essential.

Understanding the surface condition on the Moon, especially in areas shadowed by boulders, is helpful in designing surface operations. It also helps us to understand the possible distributions of volatiles on the Moon, the most important of which of course is water. However, volatile trace elements like Cd, Hg, C, and N have been considered [20-24], although probably more important are H and ^{3}He [25].

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