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MEASUREMENT OF THE TEMPERATURE OF THE THERMALLY IRRADIATED SURFACE OF ALPHA CELLULOSE

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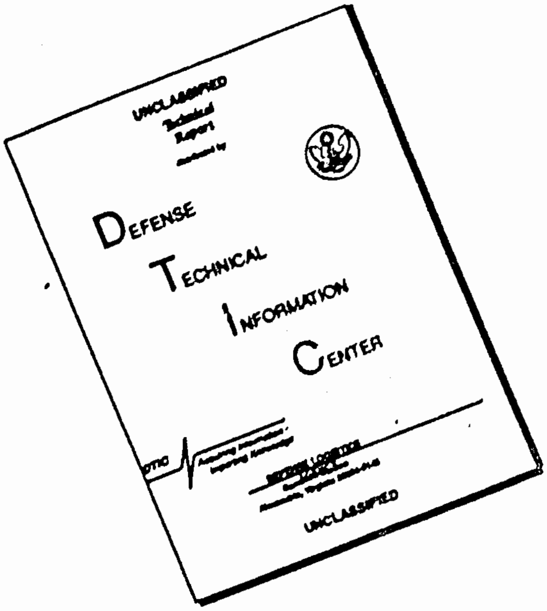

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ABSTRACT

The transient temperature histories of exposed surfaces of specimens of alpha cellulose, irradiated by a carbon arc image furnace are radiatively measured with a liquid nitrogen cooled, lead sulfide detector, behind a 3.4 micron, narrow band-pass interference filter. The output of the detector is displayed on an oscilloscope and is photographed by a Polaroid Land Camera. The overall time response of this system is 30 milliseconds. Reflected arc radiation off the surface of the sample at 3.4 microns is eliminated by inserting a Plexiglas filter between the arc and the specimen. A mirror is used to focus the detector onto an area of 0.21 square cm on the surface of the specimen. The detector is calibrated by substituting a standard black-body at the position normally occupied by the specimen during the measurement period.

For irradiances of approximately 7 and 14 cal $cm^{-2}sec^{-1}$, the temperature of the surface at the instant of ignition was measured to be 600°C for samples thicker than 0.02 in. However, due to the possibility of small errors in the emittance and to a small but finite radiating depth over which a temperature gradient exists, the surface temperature may be somewhat greater than 600°C but not over 650°C.

SUMMARY

The Problem

To measure the temperature of the thermally irradiated surface of blackened alpha-cellulose.

Reactions which precede and accompany the ignition of fine fuels by intense fluxes of thermal radiation are directly dependent upon the temperature of the exposed surface. Methods of temperature measurement which employ contact type sensors have been found to be unreliable.

The Findings

A method is described for optically monitoring the temperature of the thermally irradiated surface of blackened alpha cellulose and other fine fuels. Temperature histories of the exposed surface of several samples of blackened alpha cellulose of different thickness are presented.

CONTENTS

LIST OF ILLUSTRATIONS

LIST OF TABLES

INTRODUCTION

The Radiation Effects Branch of the Naval Radiological Defense Laboratory has been studying the ignition of fine fuels by thermal radiation for the past several years. The material used for this research is pure alpha cellulose, in the form of thin sheets, blackened by the addition of various percentages of carbon black to the initial pulp mix. The samples are irradiated in a carbon arc image furnace where the time to ignition, the depth of char, the weight loss, and the relative concentration of pyrolysis products have been measured as a function of irradiance level and time of exposure.

Since the temperature history of the thermally irradiated surface is fundamental to the understanding of the ignition process of alpha cellulose, many attempts have been made to measure this function. The most common procedure is to attach or contact a thermocouple to the surface of the sample. However, it is difficult to interpret data obtained in this manner since it is not known what part of the recorded signal is due to the direct absorption of arc radiation by the thermocouple. It is also difficult to insure that the temperature of the thermocouple junction is the same as the surface due to conduction along the thermocouple leads. Another method is to place thermocouples at various known depths in the sample. The signal from these thermocouples is simultaneously recorded, and the measured temperature profile is extrapolated to the surface. This method has unfortunately yielded inconclusive results. Since contact devices and extrapolating techniques do not yield satisfactory results, it becomes necessary to look to other means of temperature sensing, the most obvious being optical methods. The temperature measurements described herein are on specimens of alpha cellulose which range in thickness from 0.01 in. to

0.03 in. and contain about 2.0% carbon black by weight. In appearance, these specimens look quite black.

EXPERIMENTAL ARRANGEMENT

The carbon arc image furnace, used for the front surface temperature measurements, utilizes two opposing 36 inch paraboloidal reflectors to concentrate the energy from the arc into a spot 3/8 inches in diameter with an irradiance of 100 cal $cm^{-2}sec^{-1}$. The time of exposure is controlled by an air driven shutter with an opening and closing time of about 4 milliseconds[1]. The carbon-arc radiation of wavelengths longer than 2.4μ is filtered out of the beam with a 3/4 inch thick slab of Plexiglas which is placed in the parallel beam next to an aluminum grid attenuator. With this configuration, the irradiance of the imaged spot on the alpha cellulose is reduced to about 7 cal $cm^{-2}sec^{-1}$. The energy radiated normally from the surface of the sample is directed through a 4 inch hole in the 36" reflector to the radiation detector by means of a folded optical path which includes a 14 inch paraboloidal reflector and a plane mirror. A 0.2 inch diameter spot in the irradiated area of the surface of the sample is imaged onto the total sensitive area of the detector which is a liquid nitrogen cooled lead sulfide photoconductive cell. An interference filter which peaks at 3.41 microns is located directly in front of the detector. The signal from the detector is fed into the differential amplifier of an oscilloscope and is photographed by a Polaroid Land Camera.

Although the time of ignition can be observed easily from this signal, a flame detector was installed in order to confirm it. The flame detector consists of two probes which are placed approximately 1/8 inch apart, directly above the image spot of the carbon arc. A potential of 900 volts d.c. is impressed across the probes. At the instant the alpha-cellulose ignites, the ionized gases create a conduction path between the probes. This electric pulse is monitored by another oscilloscope and photographed.

A phototube was installed in the sample mount for the purpose of triggering the oscilloscopes simultaneously the instant the shutter opens.

Figure 1 is an overall view of the experimental apparatus showing the 36 inch carbon arc image furnace to the right, and the detection system to the left. Figure 2 is a closeup of the sample mount, with a sample of alpha cellulose in place. The triggering photocell and the flame detector probes can be seen in this figure. Figure 3 includes the detection system showing the optical path of the measured radiation, the detector mount, and the recording oscilloscopes. Figure 4 is a close-up of the internal configuration of the detector mount, showing the position of the lead sulfide cell and the filter mount. Figure 5 is a schematic diagram of the optical paths of interest and also the circuit diagram of the lead sulfide detector.

The area of the sensitive surface of the lead sulfide cell is 0.04 square inches. It is located in a permanently evacuated dewar behind a sapphire window. The wavelength sensitivity of this cell extends to 4.5 microns when cooled to 77°K. At this temperature, the peak sensitivity is located at 3.2 microns. The time constant was determined with a neon lamp to be 30 milliseconds. The wavelength sensitivity was restricted to 3.41 ± 0.02 microns by means of an interference filter. This wavelength region is practically free from H_2O and CO_2 absorption bands, it is close to the peak wavelength sensitivity of the lead sulfide cell and does not include any of the arc radiation which is restricted to a long wavelength limit of 2.4 microns by the Plexiglas filter. It is the wavelength at which a 580°C black body would emit the most energy.

Figure 6 is a composite of three curves which illustrate the wavelength characteristics of the carbon arc radiation limited by the

Plexiglas filter, the wavelength sensitivity of the filtered lead sulfide cell, and the radiation curve of a 600°C black body. The amplitudes of these curves are plotted in arbitrary units.

CALIBRATION AND PROCEDURE

The lead sulfide cell used in this experiment was calibrated in its normal mode of operation. The calibrations were accomplished by placing a black body source having a 0.2 inch diameter orifice at the position normally occupied by the specimens of alpha cellulose during the measurement sequence. The temperature of the black body source was varied from 200°C to 1000°C in 100°C increments. The output of the lead sulfide cell was recorded and a calibration curve was drawn. The black body source was then replaced by the sample holder, and radiation from the arc was focused onto the surface of the alpha cellulose sample. The signal from the lead sulfide cell, created by the transient temperature rise of the surface of the alpha cellulose was fed into the oscilloscope and photographed. The deflection of the measurement pulse was then compared to the temperature calibration and the resultant temperature data was plotted as a function of time.

This procedure appears to be straightforward, except for one disturbing factor. The sensitivity of the lead sulfide cell changed from day to day. This fact made it necessary to repeat the calibration procedure for every set of samples that were exposed to the arc. A current stabilized glow bar was used as a secondary standard and was placed adjacent to the detector. Radiation from the glow bar was reflected to the sensitive area of the detector by means of a small front surface mirror before and after each exposure. The enhancement of the lead sulfide cell's sensitivity was in the order of a factor of four over the five months that this experiment was in operation. A survey of the literature concerning the operation of lead sulfide cells indicated that this effect had been previously observed, and was not peculiar to this experimental setup[2]. Any optical determination of temperature requires a prior knowledge of the emittance of the surface of the material being measured. In order to satisfy this requirement, a measurement of the emittance of alpha cellulose at a wavelength of 3.41 microns was made at a temperature of 207°C. This measurement was made by firmly securing a specimen of alpha cellulose to the surface of a laboratory hot plate. The exposed surface was then placed at the focus of the carbon arc furnace. The current to the hot plate was turned on and the temperature of the exposed surface of the sample was brought up to an equilibrium temperature of 207°C. This temperature was monitored with a platinum, platinum 10% rhodium thermocouple at various points across the sample's surface. The detector registered an output from the heated alpha cellulose of 6.1 millivolts. The hot plate apparatus was then replaced by the black body furnace, which was heated to a temperature of 207°C. The signal recorded by the detector was again 6.1 millivolts. The comparison between the two signals indicates a total normal emittance of unity at this temperature.

Because of progressive thermal decomposition of the specimen, measurements of the emittance of alpha cellulose above this temperature were impossible by this technique. It was assumed, however, that the emittance of the surface of the samples remained essentially unchanged over the entire temperature range of the ignition experiment. The basis for this assumption is that the time of exposure of the experimental samples is of such a short duration that the products of thermal decomposition do not perturb the optical properties of the original surface. Even if this were not the case, the very thin carbonaceous char, which forms on the irradiated surface of the sample, in combination with the alpha cellulose substrate would indeed, have an emittance very close to unity. This latter view is supported by the data of Pfund(3) and Kozyrev and Vershinin.[4]

In order to determine whether or not radiation was emitted from within the samples, a measurement of the transmission, at 3.41 microns, of the blackened alpha cellulose was made by placing large, thin sheets, of blackened alpha cellulose between the detector mount and an intense source of infrared energy. Several sheets of alpha cellulose, which ranged in thickness from 0.03 inch to 0.002 inch were placed, one at a time, directly in front of the detector. The only detectable signal observed during these measurements was for sheets of alpha cellulose 0.004 in. thick or less. The 0.004 in. thick sample transmitted 0.8 percent of the total energy, and the 0.002 in. thick sample transmitted 1.6% of the total energy. From these data it was concluded that the extinction coefficient would be no smaller than 800 cm^{-1}. Using this value and the calculated temperature gradient at the time of ignition, the effect of radiation originating in the cooler region behind the surface could cause a reduction of no greater than 30 degrees C in the observed ignition temperature. The details of this calculation are covered in the Appendix.

It is difficult to insure that the computed absorption coefficient is due strictly to the bulk transmission of blackened alpha cellulose, since specimens with a thickness of 0.008 in. or less contain a myriad of microscopic pin holes which undoubtedly contribute to the transparency of the samples. The resultant error determined by consideration of the measured transmission can only be construed to be an outside limit.

Most of the work was performed at an average irradiance level of 7 cal $cm^{-2}sec^{-1}$, but a few exposures were made at an irradiance of about 14 cal $cm^{-2}sec^{-1}$. This change in irradiance was accomplished by increasing the size of the openings in the aluminum grid attenuator.

RESULTS AND DISCUSSION

The temperature rise of the surface of the alpha cellulose as monitored by the lead sulfide cell was recorded by the oscilloscope camera. Figure 7 is a photo-reproduction of four scope-camera prints showing two typical temperature traces for a 0.02 in. and 0.03 in. sample, and the corresponding flame detector traces for both exposures. The vertical scale of the Polaroid prints is 20 mv/cm, and the horizontal scale is 0.2 sec/cm. From the figure it can be seen that both the temperature traces are shaped similarly, each with a definite change in slope at about one half the amplitude and each with a very sudden change of slope near the top of the curve. It is at this latter point of the temperature history curve that the alpha cellulose ignites. This is confirmed by the presence of a voltage pulse on the flame detector trace, which is exactly coincident with the occurrence of the abrupt change of slope. Above this point, the curve increases regularly until the shutter cuts off the arc radiation.

The pronounced change in slope that occurs near the middle of the trace, could be caused by either a change in thermal conductivity, by the transpiration of gases, or by endothermic reactions. The enhancement of the trace after the ignition point is reached, is probably due to emission from incandescent soot particles, which are present in the flame.

Figure 8 is a plot of the temperature-time history of eight samples of alpha cellulose, showing the variation of the temperature of ignition with sample thickness. The thickness of the measured samples was 0.01 in., 0.012 in., 0.02 in., and 0.03 in., respectively and data from two samples of each thickness are included in the curve. The average ignition temperature varied from 560°C for the 0.01 in. samples, to 600°C for the 0.03 in. samples. This variation is probably due to geometric factors, e.g., in the case of the thinner samples, the back surface temperature increases significantly before ignition can occur on the front surface. Thus the average temperature of the thinner samples is probably higher than the average temperature of the thicker samples for the same irradiance level. If this is the case, then the volumetric evolution of volatile gases would be greater for the thinner specimens, which might in effect reduce the time and temperature of ignition.

The 0.02 in. and 0.03 in. samples ignite at essentially the same temperature. Here the surface temperature is not affected by the presence of a back surface, i.e., for all practical purposes it is infinitely thick.

Figure 9 is a photo-reproduction of two oscilloscope traces which are the result sample exposures made at an irradiance of about 14 cal $cm^{-2}sec^{-1}$. The vertical scale of these traces is 50 mv/cm and the horizontal scale is 0.1 sec/cm. Measurements were made at the higher irradiance level, to determine whether there is an effect upon the ignition temperature caused by a change in irradiance. The left trace was from the exposure of a 0.02 in. sample, and the right trace was from a 0.03 in. sample. The measured ignition temperature was 602°C and 608°C. In the case of the higher irradiance, the point of changing slope has moved closer to the inflection point at which ignition occurs. From these data alone, we cannot reach any definite conclusion regarding the cause of the change in slope.

Table 1 is a collection of all the ignition temperature determinations and ignition time values measured during this research. The average temperature of ignition for samples of each thickness is listed for the low irradiance measurements.

The temperature-history data shown have an estimated variance of ±10%. The variations in times to ignition listed in table 1 for samples of the same thickness indicate that the irradiance of the carbon arc image furnace fluctuated significantly. Also the wide spread in the data in the lower temperature portion of Figure 7 points to a variation of the arc radiation. Variations in the moisture content of the alpha-cellulose samples could change the shape of the surface temperature curve but neither the fluctuation in the moisture content or in the irradiance should affect the ignition temperature. The high degree of reproducibility of this measurement is reflected by the small standard deviation for the mean of a large number of determinations, i.e., 2-3°C, even with some scatter in the data due to the instability of the sensitivity of the lead sulfide cell. The accuracy of the measurement is influenced by some uncertainty in our knowledge of the optical properties of the surface. A change in emissivity from the assumed value of 1 to 0.9 would cause the measured temperatures to be some 20° lower than actual temperatures. Diathermancy at 3.4μ would introduce an error of no greater than 30 degrees in the same direction. From published optical properties combined with a measurement of the emissivity of cellulose at 200°C, it is concluded that the temperature of the surface at ignition is greater than 600°C but less than 650°C.

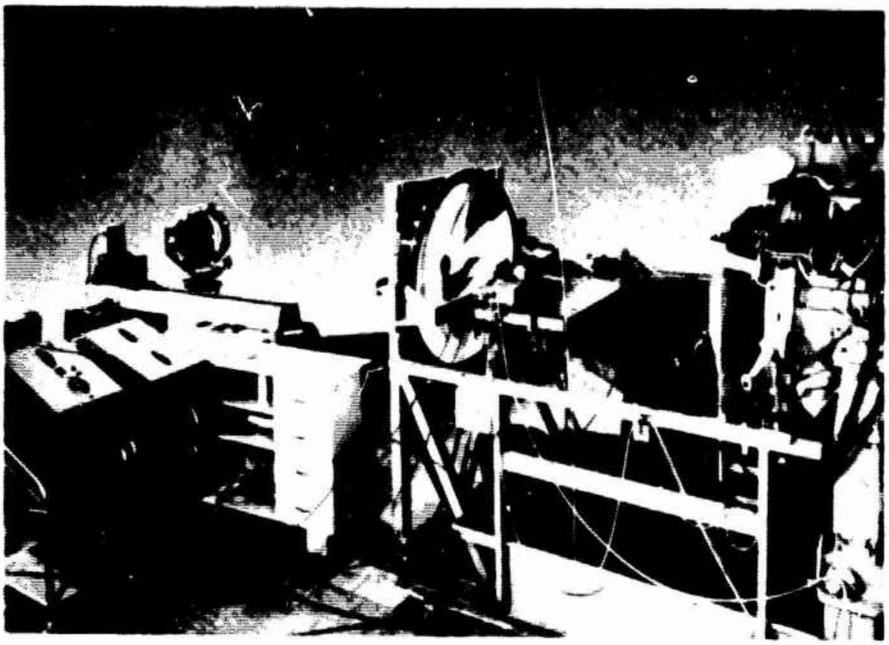

Fig. 1. Overall Photograph of Apparatus for Measurement of the Surface Temperature of Alpha Cellulose.

Fig. 2. Photograph of the Sample Mount for Alpha Cellulose and its Position Relative to the Carbon Arc Image Furnace.

Fig. 3. Photograph of the Detection Apparatus used for the Measurement of Surface Temperature.

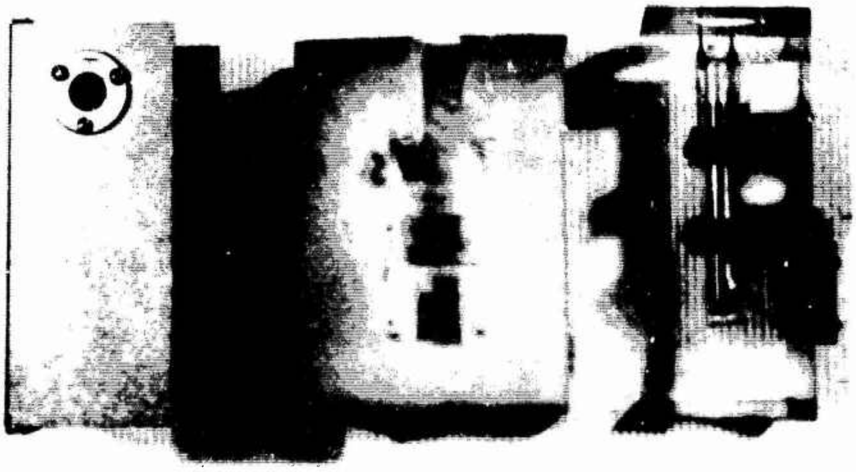

Fig. 4. Detail Photograph of the Internal Configuration of Detector Mount.

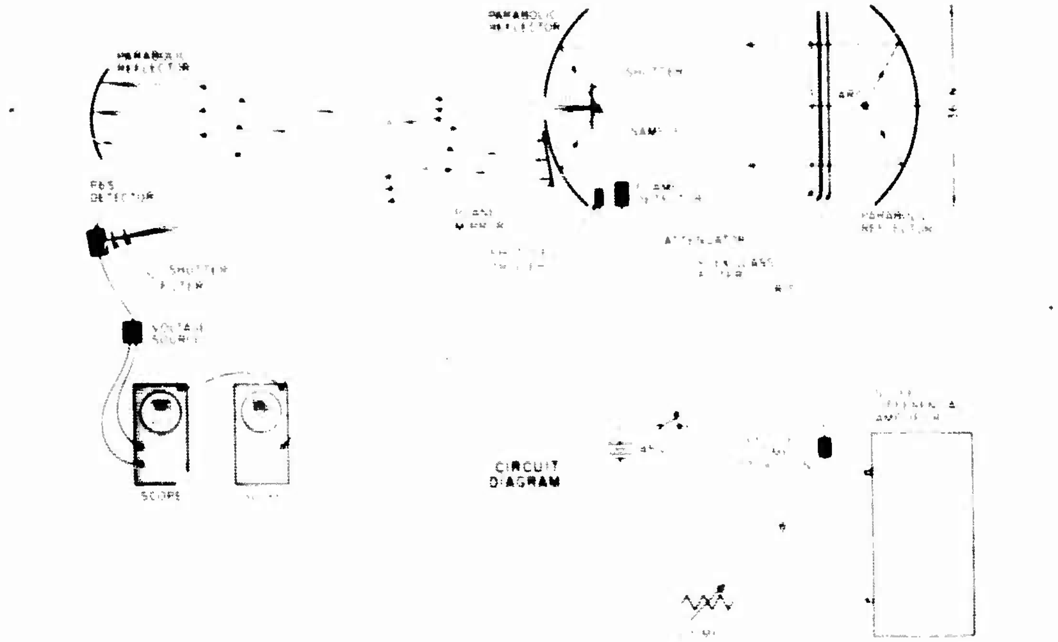

Fig. 5. Schematic Diagram of Apparatus for Measurement of Surface Temperature and the Circuit Diagram of the Electrical System of the Lead Sulfide Cell.

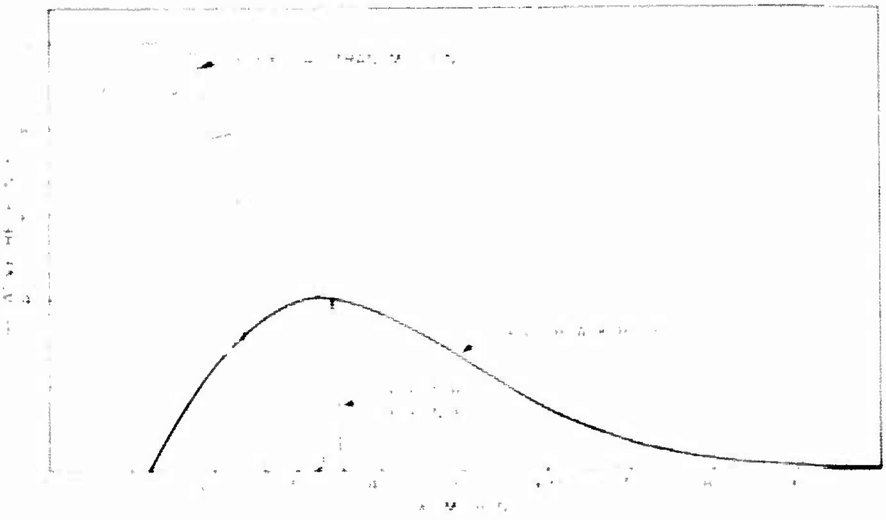

Fig. 6. Schematic Diagram of Spectral Relationship of Source Radiation and Detector Response.

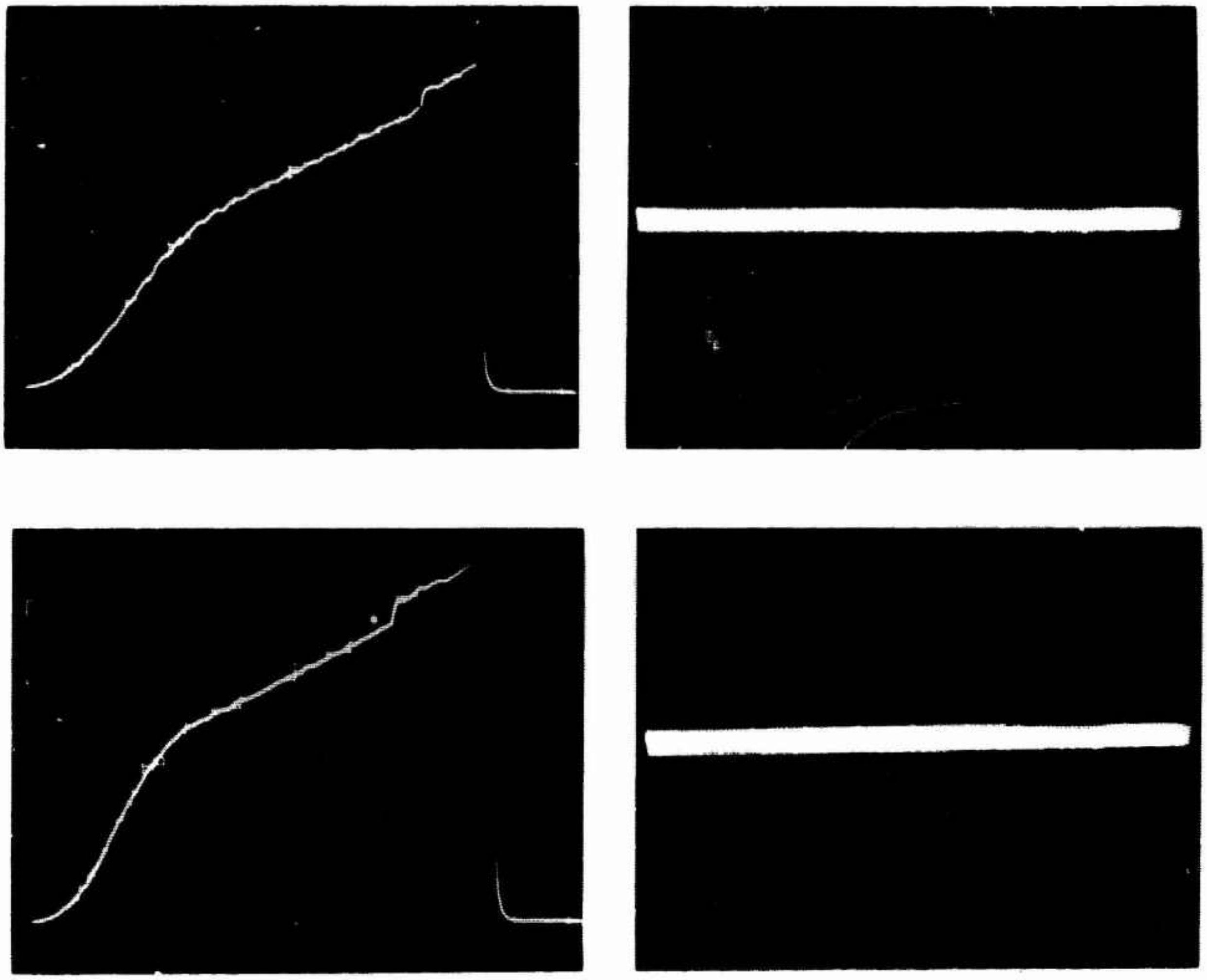

Fig. 7. Two Photo Reproductions of Polaroid Recordings of the Surface Temperature Trace and Flame Detector Trace at an Irradiance of Approximately 7 cal cm^{-2} sec^{-1} for a 20 mil sample at the top of the Figure, and a 30 mil Sample at the Bottom. Note the Correspondence of the Voltage Pulse of the Flame Detector and the Trace Discontinuity.

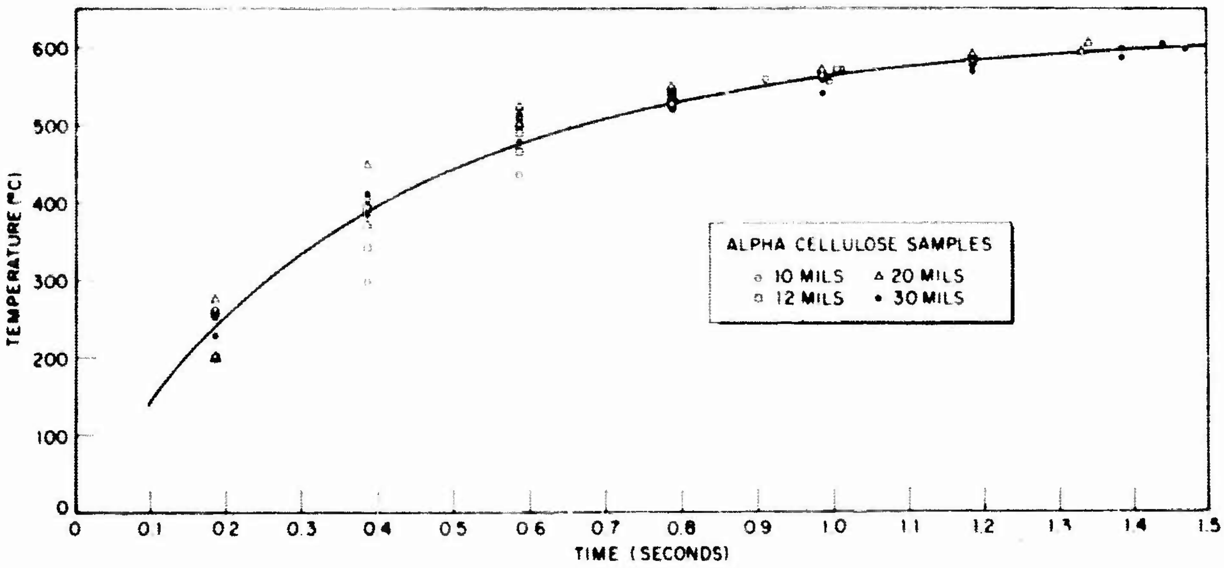

Fig. 8. Curve of the Temperature History of the Front Surface of Several Samples of Blackened Alpha Cellulose.

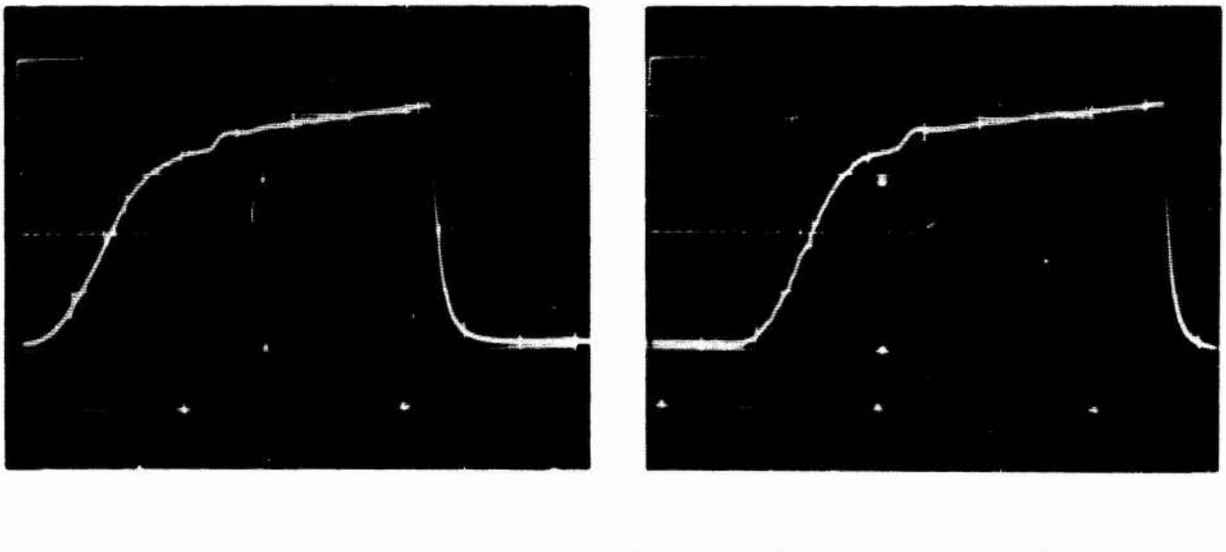

Fig. 9. Two Photo Reproductions of Polaroid Reproductions of the Surface Temperature Trace at an Irradiance of about 14 cal cm^{-2} sec^{-1}.

TABLE I

LISTING OF IGNITION TEMPERATURES AND IGNITION TIMES FOR ALL SAMPLES OF ALPHA CELLULOSE MEASURED DURING THIS EXPERIMENT

Low Irradiance	Date	Ignition Time(sec)	Ignition Temperature °C 10 mil	12 mil	20 mil	30 mil
	14 June 63	0.99			580°	
	19 June 63	1.17				602°
		0.81		557°		
		0.81		552°		
	3 July 63	1.11			590°	
	13 July 63	.91	558°			
		.99	556°			
		1.05		571°		
		1.08		572°		
		1.33			593°	
		1.34			605°	
		1.44				604°
		1.47				598°
	29 Aug 63	1.36			591°	
		1.43				609°
	23 Sept 63	1.32				603°
		1.22				603°
		1.11			574°	
		1.24			587°	
		1.19			570°	
	24 Sept 63					
	24 Sept 63	1.13			594°	
		1.22			588°	
		1.21			594°	
		1.22			589°	
		1.32				606°
		1.24				593°
	(Average Ignition Temp.)		557°	563°	588°	602°
High Irradiance	8 Oct 63	0.28			602°	
		0.34				608°

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APPENDIX A

Computation of the correction to the measured surface temperature of a specimen of semi-transparent alpha cellulose.

SYMBOLS

T_o	Measured surface temperature °K
T_x	Temperature as a function of depth at time of ignition °K.
T_t	True temperature °K
t	Time (sec)
k	Thermal conductivity
ρ	Density
C	Heat capacity at constant pressure
ierfc	An integral of the error function
α	Thermal diffusivity
x	Thickness
I	Intensity
I_o	Incident intensity
β	Absorption coefficient
H	Radiant energy from sample
C_1	First radiation constant
C_2	Second radiation constant
λ	Wavelength (3.41 microns)
ϵ^*	Effective emissivity of specimen
F_o	Incident radiant flux to specimen

The temperature within a sample of blackened alpha-cellulose, at the time of ignition is given by;[5]

$$T_x = F_o\left(\frac{t}{k\rho C}\right)^{1/2} 2\text{ierfc}\left(\frac{x}{2(\alpha t)^{1/2}}\right). \tag{1}$$

at $\frac{x}{2(\alpha t)^{1/2}} = 0$, the function 2ierfc = 1.128, therefore:

$$\text{at } x = 0,\ T_x = T_o = F_o\left(\frac{t}{k\rho C}\right)^{1/2} 1.128 \tag{2}$$

$$\text{at } x \neq 0,\ T_x = F_o\left(\frac{t}{k\rho C}\right)^{1/2} 1.128\ \frac{2\text{ierfc}\left(\frac{x}{2(\alpha t)^{1/2}}\right)}{1.128}, \tag{3}$$

or

$$T_x = T_o\ \frac{2\text{ierfc}\left(\frac{x}{2(\alpha t)^{1/2}}\right)}{1.128}. \tag{4}$$

For the purpose of integration it is desirous to express this relation in a linear form. For the small values of $\left(\frac{x}{2(\alpha t)^{1/2}}\right)$ which are comparable to the penetration depth of radiation, it is possible to approximate equation (4) by

$$T_x = T_o\left(\frac{1}{1 + bx}\right) \quad \text{or} \quad \frac{1}{T_x} = \frac{1}{T_o}(1 + bx), \tag{5}$$

where

$$(1 + bx) = \frac{1.128}{2\text{ierfc}\left(\frac{x}{2(\alpha t)^{1/2}}\right)}$$

at $\frac{x}{2(\alpha t)^{1/2}} = 0.05$, the function 2ierfc = 1.032.

thus $(1 + bx) = \frac{1.128}{1.032} = 1.09$

$bx = 0.09$,

also at $\frac{x}{2(\alpha t)^{1/2}} = 0.05$, $x = 3.16 \times 10^{-3}$

where $\alpha \approx 10^{-3}$ and $t \approx 1.0$ sec.

$$b = \frac{9.0 \times 10^{-2}}{3.16 \times 10^{-3}} = 28.5 \quad .$$

The temperature over any small distance x, can therefore be represented by,

$$\frac{1}{T_x} = \frac{1}{T_o}(1 + 28.5\ x)\ . \tag{6}$$

In order to determine the absorption coefficient, β, of the blackened alpha cellulose, a direct measurement of transmission was made at 3.41 microns. A specimen 5.0×10^{-3}cm thick transmitted 1.6%. The fraction of absorbed intensity in a depth, dx, is given by

$$\frac{dI}{I} = \beta dx\ . \tag{7}$$

The intensity at any depth x is;

$$I = I_o e^{-\beta x} . \qquad (8)$$

Therefore;

at

$x = 5 \times 10^{-3}$cm,

$$\frac{I}{I_o} = 0.016 = e^{-\beta(5.0 \times 10^{-3})} ,$$

and

$$\ln 0.016 = -\beta(5.0 \times 10^{-3})$$

so that

$$\beta = \frac{\ln 4.14}{5 \times 10^{-3}} = 828 \text{ cm}^{-1} .$$

The emissivity of a layer of material of thickness dx, can also be represented by the term βdx, thus the energy radiated by this layer is;

$$H(dx) = \beta dx C_1 \lambda^{-5} e^{-\frac{C_2}{\lambda T}} .$$

The Wein's law approximation is valid here since $\lambda T \approx 0.3$, which means that the Wein's approximation is within one percent of Planck's relationship.

The fraction of energy which reaches the surface of the specimen is $e^{-\beta x}$, thus the integrated intensity at the surface is;

$$H = \int_0^\infty C_1 \lambda^{-5} e^{-\frac{C_2}{\lambda T x}} e^{-\beta x} \beta dx . \qquad (9)$$

Substituting for $\frac{1}{T}$ from equation (5);

$$H = \int_0^\infty C_1 \lambda^{-5} e^{-\left(\frac{C_2}{\lambda T_o} + \frac{C_2 bx}{\lambda T_o} + \beta x\right)} \beta dx \quad , \tag{10}$$

$$H = \beta C_1 \lambda^{-5} e^{-\frac{C_2}{\lambda T_o}} \int_0^\infty e^{-\left(\frac{C_2 b}{\lambda T_o} + \beta\right)x} dx,$$

$$H = \left(\frac{\beta}{\frac{C_2 b}{\lambda T_o} + \beta}\right) C_1 \lambda^{-5} e^{-\frac{C_2}{\lambda T_o}} \quad .$$

Thus an effective emissivity (ϵ^*) can be defined as,

$$\epsilon^* \equiv \frac{1}{1 + \frac{C_2 b}{\beta \lambda T_o}} \tag{11}$$

which takes into account the radiation from the cooler layers beneath the surface. In this case;

$\beta = 828\ \text{cm}^{-1}$, $C_2 = 1.44$, $b = 28.5$

$\lambda = 3.41 \times 10^{-4}\ \text{cm}$, $T_o = 873^\circ\text{K}$

so that

$$\epsilon^* = 0.86 \quad .$$

The true surface temperature, T_t, of the specimen including the effect of diathermancy is determined from the relationship

$$\epsilon^* \, e^{-\frac{C_2}{\lambda T_t}} = e^{-\frac{C_2}{\lambda T_o}} \qquad (12)$$

where T_o = measured surface temperature (873°K). the solution for T_t is as follows;

$$\epsilon^* = e^{-\frac{C_2}{\lambda T_o} + \frac{C_2}{\lambda T_t}}$$

$$\ln \epsilon^* = \frac{C_2}{\lambda} \left(\frac{1}{T_t} - \frac{1}{T_o} \right)$$

$$\frac{T_o}{T_t} = 1 + \frac{\lambda T_o}{C_2} \ln \epsilon^*$$

$$T_t = \frac{T_o}{1 - \frac{\lambda T_o}{C_2} \ln \left(\frac{1}{\epsilon^*} \right)}$$

substituting in, $\epsilon^* = 0.86$, $\lambda = 3.41 \times 10^{-4}$, $T_o = 873$, and $C_2 = 1.439$

$$T_t = 900^\circ K = 627^\circ C \; .$$

Thus the error in the surface temperature can be no greater than + 30°C due to diathermancy.