APENDIX B

Response to "Extension of Emission-Transmission Technique to Particulate Samples Using FT-IR" Response to "Comment on 'Extension of Emission-Transmission Technique to Particulate Samples Using FT-IR'" by D.J. Seery and J.D. Freihaut

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INTRODUCTION

Seery and Freihaut (SF) have commented on the Emission/Transmission (E/T) method of particle temperature measurement presented in a recent publication in Combustion and Flame (1). They suggest that such a method would be very important if it is truly laboratory independent. They have agreed that the method we have developed gives casonable results for particles exhibiting gray, or near black-body behavior, but they raise questions of the accuracy of the method when it is applied to particles which are not gray-bodies, particularly bituminous and lower rank coals. Certainly the temperature of non-gray-bodies is more difficult to determine by any technique. However, the technique we have developed is ideally suited to such measurements and has no subjective features which might make it laboratory dependent.

The theory of the technique is complicated, and answering the questions raised by SF provides useful amplification of points which may not have been clear in the paper. In particular, SF raised questions about the approximations used and their range of application and these are addressed. Most of our response to their comments concerns non-gray-bodies. In addition, there were several errors in the original paper which are considered in an erratum at the end of the paper. These errors do not have a significant impact on the matter under discussion.

TEMPERATURE MEASUREMENT METHODS

Measurements of temperatures for gray-bodies can be made in several ways. By definition, such bodies will emit radiation which has the spectral dependence of a black-body, but at a reduced amplitude. The temperature can be determined by comparing the shape of the radiance spectrum to the shape of a theoretical black-body spectrum (i.e., the Planck function). This can be done by comparing the radiance amplitude at a minimum of 2 frequencies, i.e., 2 color pyrometry. Increased accuracy can be achieved by using more frequencies. In the work reported in Ref. 1, the spectrum is obtained using 8 cm⁻¹ resolution from 500 to 6500 cm⁻¹ at 2000 data points, i.e., 2000 color pyrometry.

Another method to measure temperature for gray-bodies is to determine the amplitude of the radiance, normalized to the emitting surface area at one frequency, and to compare this to the amplitude of the Planck function at that frequency, i.e., one color pyrometry. To do this, the frequency independent emittance of the body must be known. Additional accuracy can be obtained by using multiple frequencies.

In the method reported in Ref. 1, measurements are made of both the radiance R, and the transmittance \mathcal{T}_{v} through a homogeneous sample of particles suspended in a gas stream. For large particles and high frequencies, $1-\mathcal{T}_{v}$ represents the fraction of the viewing area obscured by the geometric areas of the particles. The normalized radiance $\mathbb{R}^{n}_{v} = \mathbb{R}_{v}/(1-\mathcal{T}_{v})$ is thus the radiance normalized to the emitting surface area.

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The use of the E/T method, thus allows the measurement of temperature to be performed by comparison of the normalized radiance to the Planck function in shape, or amplitude, or both.

For non-gray-bodies, either one or more color pyrometry can be performed so long as the emittance is known for each frequency. In Ref. 1, the application of the E/T method was considered for the case of hot particles and cold walls, (e.g., the HTR described in Ref. 1). The temperature is obtained by comparing the normalized radiance to the Planck function. From Eq. 19 of Ref. 1,

$$\mathbf{R}_{\mathbf{v}}^{\mathbf{n}} = (\boldsymbol{\varepsilon}_{\mathbf{v}} / \mathbf{F}_{\mathbf{v}}^{\mathbf{t}}) \mathbf{R}_{\mathbf{v}}^{\mathbf{b}} (\mathbf{T}_{\mathbf{p}}).$$
(1)

Thus, to determine temperature from the measured normalized radiance, it is the ratio $\varepsilon_{J}/F_{J}^{L}$ which must be known at one or more frequencies.

The simplest situation for the temperature determination of non-gray-bodies would be one in which the chemical composition of the particles does not change with temperature. In that case a measurement of normalized radiance at a known particle temperature T_p , would suffice to determine ϵ_v/r_v^t since,

$$\sum_{n} / F_{\nu}^{\mathrm{L}} = R_{\nu}^{\mathrm{n}} / R_{\nu}^{\mathrm{b}}(\mathrm{T}_{\mathrm{n}})$$
⁽²⁾

With the assumption that the particle size does not change appreciably with temperature, this determination would need to be done at only one value of temperature. Subsequent measurement of the amplitude of the normalized radiance at a different temperature would be sufficient to determine that temperature. This determination would utilize a set of black-body amplitudes determined during calibration with a black-body cavity. This procedure is valid for amplitudes measured at just one value of wavenumber; complete spectra would not need to be measured. However, added accuracy could be obtained using several frequencies.

For particles which change composition with exposure to high temperature, the procedure becomes much more complicated. Then, a measurement of the normalized radiance at a known temperature will determine the value of ε_v/F_v^t of the particles at that temperature, and at that composition. In a temperature determination it is important that the value of ε_v/F_v^t used is that for particles of the same composition.

TEMPERATURE MEASUREMENTS FOR COAL AND CHAR PARTICLES

Measurements for coal and char show that the emittance can vary considerably with the extent of pyrolysis (see Fig. 10 of Ref. 1). Because of this, it may seem that the measurement of coal particle temperatures is a hopeless task. Our early observations (2-4) suggested, however, that for particles larger than $35 \,\mu$ m, the emittance of all coals, and non-graphitized chars, is not significantly different from 0.9 in the wavenumber region from 1200 to 1600 cm⁻¹ (excluding water vapor lines). More precisely, what was measured in Refs. 1-4 was the ratio $C_{\downarrow}/F_{\downarrow}^{L}$ which was equated to E_{\downarrow} in the limit of large particles for which $F_{\downarrow}^{L} \approx 1$. For now we will assume $F_{\downarrow}^{L} = 1$. This assumption does not affect the temperature measurement in such a way that errors due to the departure of F_{\downarrow}^{L} from unity will cancel. This insensitivity of the temperature measurement to F_{\downarrow}^{L} is also important with regard to the question of laboratory independence, since, as discussed in (1), F_{\downarrow}^{L} depends on the optics of the spectrometer which can vary among laboratories.

The observation that there is a band width over which $\epsilon_{\gamma}/F_{\gamma}^{t}$ is constant, independent of pyrolysis state or particle size, indeed simplifies the task of temperature determination. For large coal particles, high rank coals, and coal chars, other regions of the spectrum may also have ϵ_{γ} , and such regions can be used for added accuracy. For example, Fig. 6 of Ref. 1 shows a char for which ϵ_{γ} , 0.87 over the whole spectrum.

Because of the importance of the constant value of $\epsilon_{\gamma}/F_{\nu}^{L}$ in the 1200 to 1600 cm⁻¹ region, we wish to demonstrate this observation from a theoretical as well as experimental perspective. In Figure 1 are KBr pellet spectra recorded for chars of a Zap North Dakota lignite in differing states of pyrolysis, formed in the heated tube reactor (see Ref. 1) at an asymptotic tube temperature of 800°C. From these spectra we can determine the imaginary part of the complex index of refraction, k, as a function of wavenumber as discussed in Ref. 2 to 4. The 1200-1600 cm⁻¹ region is one in which scattering is small in the pellet spectra, so the absorption coefficient can be accurately determined from these data.

In Figure 2a we have plotted (open circles) an average value of k in the 1200 to 1600 cm⁻¹ region measured from the char spectra (Fig. 1), as a function of distance, D, traveled in the tube reactor. Here, the extent of pyrolysis increases monotonically with increase in D. Also plotted (closed circle in Fig. 2a) is the value for the raw coal of k = 0.007 at 2000 wavenumbers.

Taking a constant value of 1.6 for the real part of the index of refraction, n, we have used the Mie Theory program in Bohren and Huffman (5) to calculate the emittance of particles of different size, and different k values (Fig. 2b).

Using Figures 2a and 2b it can be seen that for all states of pyrolysis, the value of k determined for the chars in the region between 1200 and 1600 cm⁻¹ are such that the value of the emittance, ϵ_{ν} , is constant and is within 10% of 1.05 for particle diameter, d, greater than 20 μ m. The actual value depends slightly on particle size, but is practically independent of pyrolysis state. At 2000 cm⁻¹ the values of ϵ_{ν} for coals are particle size dependent and significantly less than unity for all particle sizes computed. As can be seen in Fig. 2b, any increase in k with extent of pyrolysis will affect ϵ_{ν} at 2000 cm⁻¹. The Mie theory calculations are for spheres, rather than coal-particles, but evidence, described below, supports these values.

There is a stage in pyrolysis beyond which $\mathcal{E}_{\sqrt{2}}$ in the 1200 to 1600 cm⁻¹ region starts to vary. This occurs when the coal starts to graphitize and both the real and imaginary parts of the complex index of refraction (m = n +ik) start to increase. In Fig. 3 we display contour plots of emittance in the n-k plane for 55,4 m diameter spheres. That part of the figure which contains the n and k values appropriate to coals in the infrared region of the spectrum is shown shaded, on the left hand side of the figure. In a similar manner, the region for graphite is shaded on the upper right hand side of the figure. During pyrolysis, the emittance in the 1200 to 1600 cm⁻¹ region (where k ≈ 0.1) drops from a value greater than 1, towards the value for graphite. In other regions of the spectrum where k<0.1, $\mathcal{E}_{\sqrt{2}}$ first increases and then decreases as k increases. Our experimental observations suggest that for residence time on the order of 1 sec, n and k increase significantly only above 1000°C, and it is above this temperature that $\mathcal{E}_{\sqrt{2}}$ is observed to decrease. For a highly graphitized char $\mathcal{E}_{\sqrt{2}}$ can be as low as 0.7.

In order for (\in_{v}/F_{v}^{t}) to be constant as a function of temperature, F_{v}^{t} must also be temperature independent. Mie theory calculations show that F_{v}^{t} is insensitive to the variations in k expected during coal pyrolysis. Some calculations of F_{v}^{t} are presented

in Fig. 4. In Fig. 4a we show calculations of F_{L}^{L} for our instrument, using the optical constants we have derived for a Montana subbituminous coal (2). The Nicolet 7199 FT-IR spectrometer, with a 4" diameter, 9.5" focal length collection mirror, has a semi-cone acceptance angle of 12° which was used for the calculations in Fig. 4a. It can be seen that F_{L}^{L} is a smooth function of wavenumber, even though k changes substantially across the spectrum. Therefore, for non-swelling coals $\mathcal{E}_{J}/F_{L}^{L}$ in the 1200 to 1600 cm⁻¹ range should be independent of temperature, and the temperature measurement is straight forward. For swelling coals, F_{L}^{L} must be measured at each temperature because it varies with particle size. Indeed, the shape of the F_{L}^{L} , spectrum has been made the basis for a method of particle size determination (4). The shape of F_{L}^{L} can, however, be measured using Eq. 9 of Ref. 1, and for particles above 40 μ m diameter, F_{L}^{L} at 6500 cm⁻¹ is relatively insensitive to size being equal to 1.1 + 0.10. The temperature measurement can therefore be made on swelling coals as well.

The sensitivity of the spectral shape of F_{v}^{t} to particle size distribution makes the measured F_{v}^{t} spectra useful monitors of the size distribution. Particle break-up, or swelling can be easily detected.

The calculated values of \mathcal{E}_{v} from Fig. 2b (close to 1.05), are about 17% higher than the value we reported in Refs. 1-4 (0.9 ± 0.05). This difference is a result of the approximation that F_{v}^{L} is unity for particles of the size we used. Estimating F_{v}^{L} to be 1.25 at 1600 cm⁻¹ for 55 μ m diameter particles, the corrected values of emittance becomes $\mathcal{E}_{v} = 1.13 \pm 0.05$, in agreement with the calculations. This agreement of these two completely independent determinations of \mathcal{E}_{v} gives considerable support to the application of the emission-transmission technique to particle streams.

We believe that this discussion clarifies the main question raised by SF; that of the value of particle emittance appropriate to a temperature determination for coal.

ANSWERS TO QUESTIONS IN SF COMMENT

1. The wavenumber region which is employed to determine temperature for coal is from 1200 to 1600 cm⁻¹ (8.33 to 6.25 μ m). As discussed above, the best region for temperature determination is where the absorption coefficient is large and ϵ_{ν} is close to unity. The 1200 to 1600 cm⁻¹ region contains the C-C and C-O absorption bands of coal which typically have the highest values of absorption coefficient. Figure 1 shows the KBr pellet spectra for a lignite and several chars. Figures 9a of Ref. 1 shows a KBr pellet spectrum for a subbituminous coal. As discussed in Refs. 2-4, the sloping background is mostly due to scattering in low rank coals. The region where the absorption coefficients is largest is the 1200 to 1600 cm⁻¹ region. As shown in Figures 1 and 2a, the absorption in this region remains high even during pyrolysis.

At the resolution of our measurement (8 cm⁻¹), spectral features due to water vapor can be readily distinguished from features in the char spectra. This distinguishability is most useful in studies of combusting systems, for which temperature determinations have been made for individual gas species and condensed phases, respectively.

More important than water is tar and soot. When pyrolysis starts, tar or soot surrounding the particles could influence the temperature determination and such effects must be carefully considered. The presence of soot can be recognized by the characteristic shape of its extinction spectrum. For the furnace temperatures used in Ref. 1, no soot was observed. Possible errors due to tar evolution are discussed in question 3 below.

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2. We have determined that coal of pulverized size is not a gray or black-body. Furthermore, we have observed that the emittance changes with the extent of pyrolysis. Fitting the shape of the radiance spectrum to a black-body curve is, therefore, questionable. The use of the E/T method allows one to fit the black-body in both shape and/or amplitude. Thus, it is possible to derive a temperature from one spectral region if the value of ϵ_{v}/F_{v}^{t} is known (i.e. one-color pyrometry). Only the amplitude information is utilized in this case. In Ref. 1, the value of ϵ_{v}/F_{v}^{t} is determined experimentally, and its value used in the temperature determination. The insensitivity of ϵ_{v} to particle size and extent of pyrolysis in the 1200 to 1600 cm⁻¹ region makes the method applicable to coal particles greater than 20 /m in diameter.

Other regions of the spectrum are useful. For example, \in in the high frequency region of the spectrum should also be close to unity for large particles, high rank coals, or when sufficient pyrolysis has occurred.

The "goodness of fit" refers indirectly to the reproducibility of the method, and its accuracy, topics we discuss later.

The method is not restricted to temperatures with black-body peaks near 1600 cm⁻¹. While the sensitivity of the apparatus we used for the work described in Ref. 1 falls off rapidly beyond wavenumbers of 6500 cm⁻¹, with associated deterioration the signal-to-noise ratio, other detectors and beam splitters are available which extend the method to the visible region of the spectrum. With a MCT detector, particle temperatures have been measured between 100 and 1600°C.

3. $(1 - \tau_{y})$ spectra of devolatilizing coal particles are displayed in Ref. 6. No additional $(1 - \tau_{y})$ spectra were presented in Ref. 1, because we did not wish to extend the length of the paper unnecessarily.

The spectra corresponding to Fig. 10 of Ref. 1 are shown in Fig. 5. Changes in level of these $1-\tau_v$ spectra with pyrolysis mainly reflect change in velocity of the particles, and hence the average number of particles in view at one time. The emission-transmission technique is insensitive to this variation.

Prior to the onset of pyrolysis, the spectra are mostly flat and uninteresting. When pyrolysis is initiated, some structure can be observed for the evolved species. In the 1200 to 1600 cm⁻¹ region, the tar has increased the extinction by up to 20%, (Fig. 5d). If the tar is at the same temperature as the coal particle, the radiance will also be increased by the same percent and the normalized radiance will be unchanged. If the tar is 100°C hotter than the coal, the normalized radiance will be a weighted average of the two temperatures, or ~ 20 °C hotter than the particle temperature. A detailed analysis can be made employing the whole spectrum to identify separate temperatures for the particle and the volatile species. It is noted that the presence of tar signals itself by increased extinction in this wavenumber region.

The pellet spectrum in Figure 9a, and discussed on pages 58, 60,62, and 63 of Ref. 1, was presented to show the correspondence between absorption bands in the KBr pellet spectrum and the emission bands in the normalized radiance spectrum.

4. We have performed many calculations of F_{ν}^{t} for a number of situations relevant to our own measurements using the appropriate collection angles for the optics we employ. For these calculations, we used the Mie theory program published by Bohren and Huffman (5). Some of the results of these calculations are displayed in Fig. 4a. They are accurate to better than 1%. Uncertainty in the amplitude of F_{ν}^{t} , then, is not a major source of error in the determination of E_{ν} , or of temperature in the EFR. As

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described in the Method Section, accurate temperature determination for the HTR can be made from $\mathcal{E}_{\mathcal{I}}/F_{\mathcal{I}}^{\mathsf{L}}$ independent of precise knowledge of the individual $F_{\mathcal{I}}^{\mathsf{L}}$ and $\mathcal{E}_{\mathcal{I}}$ values.

In those cases where F_{ν}^{L} is used to determine temperature, using Eqs. 19 or 24 of Ref. 1, for example, there are two particle sizes which distinguish different approximations for F_{ν}^{L} . These particle sizes depend on the angular aperture of the instrument. For the largest particle sizes, F_{ν}^{L} will be unity across the spectrum (as for the 250 μ m curve of Fig. 4a). For some intermediate size range, 40 μ m and up for our instrument, F_{ν}^{L} at 6500 cm⁻¹ will be approximately unity and this can be used to calibrate the whole F_{ν}^{L} spectrum, the shape of which is determined from the transmittance spectrum (Eq. 9 of Ref. 1).

The calculations in Ref. 24, of Ref. 1, show $E_{\mu} = 1.00$, 1.01 and 1.00 at 5000 cm⁻¹ for particles of d = 80, d = 50, and d = 30 μ m, respectively. The maximum error in assuming $F_{\mu}^{L} = 1$ at 6500 cm⁻¹ for particles greater than 35 μ m diameter is less than 15%.

In Fig. 4b we display spectra of F_v^t for 60 μ m diameter particles, for several different values of \mathcal{O} , the semi-cone acceptance angle of the spectrometer. For particles of this size, a large acceptance angle leads to a spectrum of F_v^t which is close to unity except for the low frequency region.

5. In our paper we discussed the work of all the authors cited by SF specifically to compare our results, and to point out the discrepancies in the reported values of k for coal. SF make no attempt to refute our discussion. The measurements of emittance for coal are discussed in more detail in Refs. 2-4. The emittance data presented in Refs. 1-4 are inconsistent with those of Foster and Howarth (7) and Grosshandler and Monterio (8). The results in Refs. 7 and 8 appear to be in error as discussed by us (1,2) and by Brewster and Kunitomo (9). Our data do support the work of Brewster and Kunitomo. Contrary to the statement of SF, Brewster and Kunitomo do not have data above 4000 cm⁻¹ from their particle extinction technique. The fact that Cashdollar and Hertzberg (9) see emission at wavelengths where we reported negligible emittance for a lignite, can be attributed to several factors. First, their starting sample is a bituminous coal which does show considerably greater emittance than lignite at the wavenumbers under discussion (2-4). Secondly, their emission is from an explosion, in which some pyrolysis will have taken place, and perhaps some soot has been formed. At the wavenumbers of interest the emittance of coals of all ranks increases with the extent of pyrolysis, and soot emits in this band, also.

ACCURACY OF METHOD

The final part of the SF comment has to do with the accuracy of the temperature measurement. The accuracy of the method can be considered in two ways. 1) By performing calibration experiments in which the predictions are compared to results obtained under well controlled conditions, where the particle temperatures are known. SF have not considered this. 2) By estimating the possible errors in the measurements, quantities and approximations in the analysis. SF have attempted to do this, but their estimates of error in our work are speculative. We consider both methods of estimating the possible errors.

Calibration Cases

References 1 and 4 present several cases where the particle temperatures are reasonably well known. The first case is for char discussed on pages 55 to 57 of Ref. 1. The case is for a char formed at moderate temperature, and which is a gray-body

with a high emissivity. Figure 6 of Ref. 1 shows that fitting the shape of the blackbody to the normalized radiance gives a temperature of 1000 K with black-body curves at 970 K and 1030 K giving inferior fits. The measured temperature is, therefore, 1000 K with +25 K as an upper limit of the uncertainty. This is in excellent agreement with the value of 983 K determined from thermocouple measurements (Fig. 7 of Ref. 1). Heat transfer calculations show that if the particles are in equilibrium with the tube and carrier gas at the tube exit (sufficient residence time in the hot tube was allowed for this to occur), then the maximum differences between the particles and gas temperature is 20 K.

For the best fit temperature, in Fig. 6c of Ref. 1, the emissivity required to obtain the proper amplitude was 0.87 which is in good agreement with measurements made on chars (8).

The same kind of calibration measurements have been made for $50 \ \mu$ m diameter coal particles (2-4). For these cases, the temperature determined by fitting the Planck amplitude in the 1200 to 1600 cm⁻¹ region gave temperatures within ± 40 K of the average thermocouple measurements.

Estimation of Random Errors

There are a number of sources of random errors in this experiment, the main ones being detector drift and variations in sample flow rate. The standard method to determine random error is by doing repeat measurements. When repeat measurements under nominally identical conditions have been performed, periodically over the last 2 years, the temperature variations obtained have been within +25 K for temperatures between 500 and 1100 K. With sufficient care, other workers should achieve the same level of reproducibility.

Estimation of Systematic Errors

SF estimated the errors in employing the equation

$$R_{v}/(1-\tau_{v}) = \epsilon_{v}R_{v}^{D}(\tau_{n})/F_{v}^{L}$$
⁽²⁾

based on a number of questionable or inaccurate assumptions. The following is the author's view of these uncertainties for a homogeneous sample based on experience in applying the method.

We consider first the LHS of Eq. 1. Errors in determining the radiance spectrum, R_{ν} , include: possible temperature variations in the hot cavity used as a primary blackbody standard (+30 K) and variations in the emissivity of a blackened stainless steel tube used daily as a more convenient secondary standard (+5%).

Errors in determining the transmittance, which will affect the amplitude of $\mathbb{R}^n_{\mathcal{Y}}$, are mainly random in nature, as mentioned above. We did detect, report, and eliminate two possible sources of systematic error, in Ref. 1. They were detector saturation (eliminated by neutral density filter or apertures); and errors due to spurious signals coming from the sample which are reflected in the interferometer back to the detector (such signals can be measured by turning off the source, and, using the intensity to correct the signal, or they can be reduced or eliminated with appropriate attenuation by apertures). It is emphasized that while the same flow rate of particles must be maintained in both emission and transmission measurements, the temperature determination is independent of the magnitude of that flow rate.

to the assumption of SF, uncertainty in the absolute value of the flow rate will not introduce any error in the determination of temperature.

The possible sources of error in the RHS of Eq. 1 are in \in_{V} and E_{V}^{t} . From Fig. 2, it is seen that $\in_{V} = 1.05 \pm 0.05$ in the 1200 to 1600 cm⁻¹ region so long as the particle diameter is greater than 35 μ m and the char is not highly graphitized. This value is in agreement with our corrected measured values for coals, which were found to be 1.13 \pm 0.05 for 55 μ m diameter particles. There is no basis for the \pm 50% estimate given by SF.

As for plasticizing bituminous coals, we have measured the emittance prior to pyrolysis. We have also measured k for char and thus, the emittance during pyrolysis can be calculated. Measurements should be performed to verify these predictions and to determine the effects of swelling. Such measurements are planned.

As discussed in our answer to question 4, we assume F_{ν}^{L} to be unity at the high frequency end of our spectra (i.e., 6500 cm⁻¹). From the data presented in that answer we conclude that a maximum error of $\pm 10\%$ will exist in F_{ν}^{L} , for particles $> 35 \mu m$ diameter. With knowledge of particle size and shape, this uncertainty could be reduced.

For swelling coals where \notin_{v} and F_{v}^{L} must be determined separately, adding all systematic errors, the amplitude of the normalized radiance at our chosen wavenumber region could differ from the Planck curve by $\pm 25\%$, or ± 75 K at 800 K. For nonswelling coals where ℓ_{v}/F_{v}^{L} can be measured to $\pm 5\%$, the temperature error will be ± 40 K. Again, from the calibrations reported above, the observed variations are ± 40 K for non-gray-bodies and ± 25 K for gray-bodies. These uncertainties are substantially lower than the ± 180 K estimated by SF.

The above analysis represents the state of the error estimates that we have used. In addition, commercial black-bodies are available with an emissivity of greater than 0.99 and an uncertainty in temperature of no more than ± 1 K. Under the most favorable conditions, then, the systematic error in this method of temperature determination could be reduced to less than 5% in \mathbb{R}^n or about ± 15 K.

ERRORS DUE TO NON-HOMOGENEOUS SAMPLES

An assumption made in estimating the error is that tar or soot are not interfering with the measurements. Both of these components have transmission spectra whose shape reveals their presence. The effects of soot, if present, can be corrected for. Possible error due to the presence of tar can be estimated from the relative contribution it makes to the transmission, and the temperature difference between the gas and condensed phases.

An important consideration not discussed by SF is that the analysis is for samples which are homogeneous along the line of sight. Care was taken in Ref. 1 to provide such samples. For samples which vary spatially, a tomographic reconstruction must be performed to obtain local spectra for which the homogeneous analysis can be applied. Such work is presently in progress.

Finally, SF stress laboratory independence. The only obvious laboratory dependent quantity is F_{ν}^{t} which depends on the spectrometer collection angle G_{ν} . As can be seen in Fig. 4b, for collection angles of 5° or larger, most of the variation with G_{ν} occurs in the low frequency end of the spectrum and values at 6500 wavenumbers are 1.10 ± 0.1 . With a knowledge of F_{ν}^{t} at one frequency, F_{ν}^{t} can be determined for an

individual spectrometer from Eq. 9 of Ref. 1, so the method should be laboratory independent. For non-swelling coals the temperature measurement does not require F_{y}^{t} to be separately determined. Of course, care can be taken to standardize the optics so that F_{y}^{t} can be compared among laboratories.

We expect that the validity of our method will be established by measurements at other laboratories and welcome any questions from the community concerning the setting-up of this method of temperature determination.

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ERRATUM

There were errors in three of the figures of the original paper. First, for Fig. 7, the numbers on the abscissa axis were printed without decimal points, they should go from -3.2 to 3.2 mm. Second, the results shown in Figure 9b and 9c are for a -500 mesh lignite sample. Third, the identification of the 1600 cm peak in Figure 10c should be C-C, rather than C-O.



Figure 1. Absorbance Spectra of Coal and Char in KBr-Pellets, as a Function of Distance in the Furnace at 800°C.



Figure 2. a) Average Value of k in the 1200 to 1600 cm⁻¹ Region Measured from the Char Spectra of Figure 1, as a Function of Distance Travelled in the Tube Reactor (open circles): Value of k at the Absorbance Minimum Between 1200 and 1600 cm⁻¹ (squares): k for the Raw Lignite at 2000 cm⁻¹ (closed circles): b) Calculated Emittance as a Function of k, for Spheres of Various Diameters, with n = 1.6.



Figure 3. Contour Plots of Constant Emittance Calculated by Mie Theory for Spheres of Various Values of n and k, for 55 μ m Diameter Particles. The Combinations of n and k Found for Coal in the Infrared are Shown Shaded on the Left Hand Side of the Diagram: Those for Graphite are Shaded on the Right (the values for graphite are taken from Foster and Howarth, Ref. 7). The Calculations are for Radiation of Two Wavelengths, as Shown.



Figure 4. a) Calculations of F_{ν}^{t} Spectra for our Instrument, for Particles of Various Diameters, and Having the Wavelength Dependent Optical Properties of a Montana-Subbituminous Coal (2). The Calculation for the 250 μ m Diameter Particle was done with Rayleigh Large Particle Theory. b) Calculations of F_{ν}^{t} Spectra for Instruments of Differing Half-Acceptance Angles, for Particles of Mean Diameter 60 μ m.



Figure 5. $(1 - \tau)$ Spectra for Subbituminous Coal Particles (mesh size -200 +270), Emerging from the HTR for Increasing Distances in the Tube at a Temperature of 1075K.