A PRECISION METHOD OF MEASURING THE RATIO OF RELEASED ENERGIES AND ITS PRACTICAL APPLICATIONS

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A method has been developed for determining the ratio of released energies, based on measuring the time after which the temperature curves plotted for the two components of a calorimetric system will intersect.

In experimental physics it often becomes necessary to determine the ratio of released energies, especially in relative measurements (in specific heat measurements, for example). In such cases one compares the energy released in the test specimen with that released in the reference specimen (see [1], for instance).

The author has developed a method of measurements which is based on the "overtake" effect and which he then used in various applications. The gist of this method and the meaning of the term "overtake" are as follows. Let us consider a calorimetric system where the absorber and the sheath are made of different materials. After a definite ratio of energies released in these two components has been reached, there prevails a situation where the sheath temperature for some time exceeds the absorber temperature. Such a situation is depicted in Fig. 1 for two values of the energy ratio K. The difference between the respective heating rates is called the "overtake" effect and the duration of this effect has been called the "overtake" time τ^*.

It is quite evident that such a situation can occur when

$$W_1/c_1 > W_2/c_2, \tag{1}$$

with W denoting the power release per 1 g of substance and c denoting the specific heat. Subscripts 1 and 2 refer to the sheath and to the absorber, respectively.

An analysis of this effect indicates that time τ^* is a unique function of the ratio $W_2/W_1 = K$. As W_1 increases, for instance, the heating rate of the sheath increases too (the heating rate of the absorber changes relatively little) and the temperature curves intersect later. An analogous reasoning applies to the decrease in W_2.

The basic factors (besides the released energies) affecting the "overtake" time τ^* are: the weight of the sheath G_1 and of the absorber G_2, their respective surface areas S_1 and S_2 (we will assume a sufficiently thin sheath with a negligible difference between its inside and outside surface areas), the specific heat of each material c_1 and c_2, finally the referred heat-transfer coefficients α_1 and α_2. The time τ^*, as a function of all these quantities, has been calculated in [2]. For a broader generality, we introduce the dimensionless variables: temperature

$$T_1 = \frac{t_1 - t_0}{t_{1\infty} - t_0}; \quad T_2 = \frac{t_2 - t_0}{t_{2\infty} - t_0},$$

time (criterial homochronicity number)

$$U_1 = \tau \frac{\alpha_1 S_1}{c_1 G_1}$$

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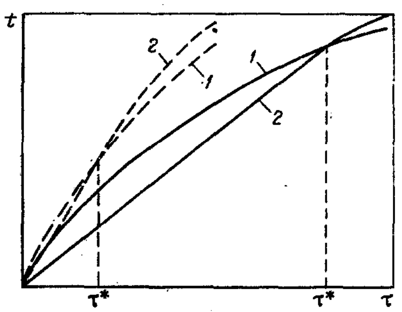

Fig. 1

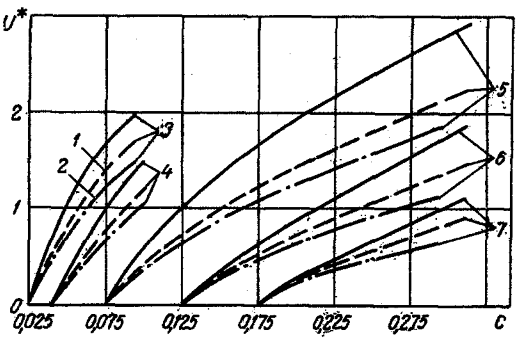

Fig. 2

Fig. 1. Temperature of calorimeter components as a function of time, for two values of K: 1) sheath; 2) absorber.

Fig. 2. Dimensionless "overtake" time $U^* = \tau^*[(\alpha_1 - S_1)/c_1G_1]$ as a function of the ratio of thermal capacities $C = c_2G_2/c_1G_1$: solid lines) $A = 0.1$; dashed lines) $A = 0.5$; dashed-dotted lines) $A = 1.0$; 1) $A = 0.245$; 2) $A = 0.5$; 3) $B = 0.025$; 4) $B = 0.040$; 5) $B = 0.075$; 6) $B = 0.125$; 7) $B = 0.175$.

and the simplex groups

$$A = \alpha_2 S_2/\alpha_1 S_1; \quad B = W_2G_2/W_1G_1; \quad C = c_2G_2/c_1G_1.$$

In these variables, then, the equations of heat balance with zero initial conditions will be

$$\partial T_1/\partial U_1 = (A-1)T_1 + \left(A + \frac{1}{B+1}\right)T_2 + \frac{1}{B+1}, \tag{2}$$

$$\partial T_2/\partial U_1 = \frac{A}{C}\left[\frac{1}{1+\dfrac{1}{A(1/B+1)}}T_1 - T_2 + \frac{1}{A(1/B+1)+1}\right]. \tag{3}$$

The solution for $U^* = \tau^*(\alpha_1 S_1/c_1G_1) = f(A, B, C)$ cannot be obtained in explicit analytical form and is, therefore, found graphically by plotting the solutions to Eqs. (2) and (3). In dimensionless variables, the instant of "overtake" corresponds to the ratio

$$T_1/T_2 = 1 + \frac{1}{A(1/B+1)}. \tag{4}$$

As a result of evaluating approximately 400 variants of solutions to Eqs. (2) and (3), with machine solutions obtained on a model USM-1 analog computer, we arrive at the following empirical relation for U^*:

$$U^* = 1.65\left(\frac{C}{B} - 1\right)^{0.74} \exp\left(-0.58A^{0.46}\right)\left[1 - \exp\left(-26.7\,C/A^{0.05}\right)\right], \tag{5}$$

where $C/B \geq 1$.

Within the range $0.1 \leq A \leq 1.0$, $1.0 \leq C/B \leq 4.0$, and $0 \leq C \leq 0.30$, Eq. (5) yields values of τ^* with an error not larger than 5-10%. When $C > 0.10$, the factor inside the square brackets may be assumed equal to unity.

The relation $U^* = f(C)$ is shown in Fig. 2 for $0.1 \leq A \leq 1.0$ and $0.025 \leq B \leq 0.175$. Evidently, τ^* becomes longer as the quantity c_2/c_1 decreases and becomes equal to zero when $c_2/c_1 = K$. As the thermal resistances increase, τ^* becomes longer and this trend follows rather clearly from physical considerations. Particularly distinct is the $\tau^* = f(K)$ curve shown in Fig. 3 (for a polyethylene absorber and a graphite sheath). For two identical materials this curve can shift appreciably as a result of geometrical changes in the system, but in any general case the curve will intersect the axis of abscissas at the point $K = c_2/c_1$.

The initial conditions are quite important here. Because of unwieldiness, the problem was not solved here for zero initial values. Generally, time τ^* becomes shorter than with zero initial conditions when the calorimetric system is initially above the ambient temperature and becomes longer than with zero

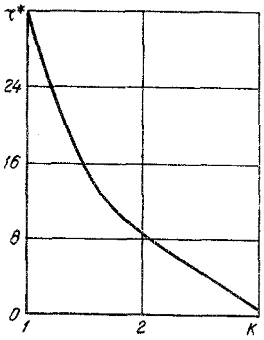

Fig. 3. "Overtake" time τ (min) as a function of the ratio K of released energies, for one particular calorimeter design (with a polyethylene absorber and a graphite sheath).

initial conditions when the calorimetric system is initially below the ambient temperature. According to the $\tau^* = f(K)$ curve shown in Fig. 3, with $W_2 = 0.03$ W/g and $K = 1.5$ a departure $\Delta t_0 = 0.5$°C from zero initial conditions causes τ^* to change by 28 sec. Apparently, the ratio $\Delta\tau^*/\Delta t_0$ decreases as W increases.

In terms of testing applications, the relation $\tau^* = f(K)$ (or, which is equivalent, $U^* = f(B)$) has one drawback: a short temperature "run" owing to the temperature dependence of specific heat and thermal resistances. Generally, the temperature "run" may be either positive or negative. Even for the earlier mentioned pair of materials (polyethylene and graphite), with a long temperature "run" for the specific heat of polyethylene, time τ^* changes by approximately 1.5% per 10°C change in the average test temperature within the 20-50°C range. Disregarding the effect of the temperature "run," which is permissible in the absolute majority of cases, one may say that the length of time τ^* does not depend on the released power but is a unique function of the powers ratio – as a consequence of Eqs. (2) and (3).

The main advantage of this proposed "overtake" method is the high accuracy with which the ratio of released powers can be measured by means of a rather simple instrumentation. Any calorimeter operating with the "overtake" effect comprises an absorber and a sheath made of appropriate materials and surrounded by an isothermal jacket. Isothermality of this outer sheath (jacket) during the "overtake" period is an absolutely necessary condition for correct measurements. Equations (2) and (3) have been derived on this premise. The absorber and the sheath are both equipped with calibrating heaters and coupled through a differential temperature probe (differential thermocouple or battery of thermocouples).

For measurements one records the initial zero (or near zero) difference between the thermocouple emfs as well as the time in which this difference goes through a maximum and returns to the initial magnitude (the "overtake" time). A preliminary calibration is made during which electric power is applied to the absorber and to the sheath under given conditions. The calibration yields a $\tau^* = f(K)$ curve. In the case of a temperature "run," the calibration is made at various initial system temperatures.

The high accuracy of this method is due to precise measurements of such quantities as time and to the relative measurements of all other quantities (currents, resistances, and weight). The differential temperature probe does not have to be calibrated. The systematic error of this method is 0.6-0.9%; its convergence is 1% or better.

APPLICATIONS

I. Measurement of Radiation Parameters

The described method is very convenient for measuring the absorbed ionizing-radiation energy and quantities related to it (for example, the mass coefficient of γ-ray energy absorption), inasmuch as here the energy is released directly in the absorber and sheath materials.

1. Determining the Composition of Absorbed Energy in the Case of Reactor Radiation. The radiation from a reactor consists essentially of two components: a γ-quanta flux and a wide-spectrum neutron flux. In several applications such as radiation physics, radiation chemistry, radiation materials science, etc., it is important to know not only the total energy released but also its composition, i.e., all its components individually. For such cases one uses materials variously interacting with at least one component [3]. Then the contribution of the neutron component, for example, to the energy released in material 1 is

$$m = W_1^n / W_1 = \frac{K_n}{K_n - K_\gamma}(1 - K_\gamma / K),$$

where K_n, K_γ are, respectively, the ratio of neutron energies and the ratio of γ-energies released in the two materials; and K is the ratio of total energies released. The accuracy with which m is determined depends mainly on the accuracy of the K measurement [4]. The application of the "overtake" effect to a K measurement under such conditions has been described more thoroughly in [2].

2. Measuring the Mass Coefficient of γ-Ray Energy Absorption. The absorption of γ-ray energy from a monochromatic radiation source is described by the relation

$$W = \mu_a/\rho\, \Phi E, \tag{6}$$

where Φ denotes the γ-quanta flux (γ-quanta); E denotes the energy (MeV per γ-quantum); and μ_a/ρ denotes the mass coefficient of energy absorption (cm^2/g). For two sufficiently thin specimens irradiated at the same field point by γ-rays we have

$$W_1/W_2 = (\mu_a/\rho)_1/(\mu_a/\rho)_2 = K. \tag{7}$$

If one of these coefficients is known accurately enough (for the material of the reference specimen), then, with the calorimetric absorber-sheath pair made of the reference and the test material, respectively, the "overtake" method will immediately yield the μ_a/ρ ratio for the test material. In this way, the author obtained K = 1.149 ± 0.011 for the polyethylene–graphite pair in the field of a Co^{60} γ-source. In the technical literature [5] is found the value K = 1.141 ± 0.004 at $E_\gamma = 1.25$ MeV. The technique of measuring the coefficient μ_a/ρ will be described more thoroughly at a later date.

On the basis of measurements made inside reactors, one may use formula (7) for estimating the mean energy of the γ-radiation spectrum.

3. Measuring the Remanent Energy of Ionizing Radiation. After ionizing-radiation energy is absorbed, most of it will be converted to heat and dissipated into the ambient medium. However, some very small portion of it (a few percent or a fraction of one percent), called the remanent energy, will be expended on forming chemical bond, producing defects in the crystal lattice, etc.

If during an "apparent" release of energy W_1 (relation (7)) there is still more unaccounted for remanent energy W_1' present, then we have

$$K = W_1/W_2 < \frac{(\mu_a/\rho)_1}{(\mu_a/\rho)_2} = \frac{W_1 + W_1'}{W_2} = K(1 + W_1'/W_1). \tag{8}$$

Inequality (8) can be used for determining at least the upper limit of W_1', but only when K has been measured very accurately, since $W_1' \ll W_1$. Applying the "overtake" method, the author has established 0.7% as the upper limit of remanent γ-radiation energy in polyethylene. The technique of measuring the remanent ionizing-radiation energy will be described at a later date.

II. Measurement of Other than Radiation Parameters

1. Quantitative Chemical Analysis. Generally, one uses here known mass coefficients of absorption for various elements. Based on the additive law for a chemical compound of the A_mB_n type, we have for the weight fraction X of element A

$$X = (\mu_a/\rho)_B \frac{(\mu_a/\rho)_{A_mB_n}/(\mu_a/\rho)_B - 1}{(\mu_a/\rho)_A/(\mu_a/\rho)_B - 1}. \tag{9}$$

The accuracy of the X determination depends on how large the difference is between $(\mu_a/\rho)_A$ and $(\mu_a/\rho)_B$ with respect to the energy of the γ-source used in the test.

The author determined the fraction of lead in grade AG-1500-S05 graphite (the 5% tin admixture to lead did not introduce an appreciable error in the final result) by measuring the "overtake" time for this graphite in combination with polyethylene. The ratio of mass coefficients of absorption is 1.32 for lead and graphite at $E_\gamma = 1.25$ MeV. This ratio has been found to be 1.05 for polyethylene and grade AG-1500-S05 graphite, but 1.149 for polyethylene and pure graphite. The lead content in the graphite specimen was 30 ± 1.2%. V. P. Savina measured the specific heat of pure graphite and of grade AG-1500-S05 graphite, using published data on the specific heat of lead and by using the additive law, and obtained the same lead content as determined from the mass coefficients of absorption.

It was possible by the "overtake" method to first record and then measure, on the basis of data pertaining to the flux density of thermal neutrons, the boron content in one calorimeter insulation component (glass-fiber insulation tape) after irradiation from a reactor. The weight content of boron was approximately 0.15% referred to the weight of the graphite sheath (about 20 mg). The current density of thermal neutrons did not exceed $1.6 \cdot 10^{12}$ n/cm$^2 \cdot$sec.

2. Measuring the Specific Heat of Solids. If the specific heat of one component in the calorimetric system is known with sufficient accuracy, then an application of the "overtake" principle will immediately yield the specific heat of the other component. Indeed, according to the method described here, the "overtake" effect ceases when $K = c_2/c_1$. A rough estimate of the unknown specific heat c_{20} is usually given beforehand. Measurements are started with $K = c_{20}/c_1$. By gradually varying K, one eventually reaches the condition where $\tau^* = 0$. Then $c_2 = Kc_1$. The specimen temperature necessary for such measurements is usually obtained by means of a fluid coolant which under normal conditions serves also to keep the outer jacket isothermal. A determination of the specific heat at room temperature does not require the use of such an isothermal fluid.

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