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THE PROPER INTERPRETATION OF THE INTERNAL ENERGY OF FORMATION USED IN THERMODYNAMIC EQUILIBRIUM CALCULATIONS

ANTHONY J. KOTLAR

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13. ABSTRACT <i>(Maximum 200 words)</i> The interpretation of the internal energy of formation as it applies to thermodynamic equilibrium calculations of gun propellants is clarified. The enthalpy of formation is compared to values of the internal energy and enthalpy based on an assigned, "absolute", energy scale. In general, at the standard pressure (0.1 MPa) and reference temperature (298.15 K), $H^{\circ} = \Delta H_f^{\circ}$, $U^{\circ} \neq \Delta U_f^{\circ}$, and in most cases, for condensed phases, $U^{\circ} \approx H^{\circ}$.			
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1. Introduction

There is a longstanding controversy in the ballistic community, largely undocumented, regarding the correct value of the internal energy of formation of condensed phases as it is used in thermodynamic equilibrium calculations. Recently this problem again surfaced at a meeting of NATO AC/225 (Panel IV/SD.2)WP9 which was working on the standards agreement, STANAG 4400, "Derivation of Thermochemical Values for Interior Ballistic Calculations" (France, September 1990). At this meeting the results of thermodynamic equilibrium calculations using two differing interpretations (Freedman 1988; Volk and Bathelt 1990) and corresponding values of the internal energy of formation were presented. This particular problem does not arise for constant pressure calculations where the relevant thermodynamic quantity, the enthalpy of formation, is tabulated and can be used directly. Equilibrium calculations in which the reactant is a gun propellant, however, are usually performed for a constant volume system, since this situation is assumed to better simulate the conditions in a gun. The internal energy of the propellant is then the quantity that is usually required; it is presumed necessary, therefore, to calculate the internal energy of formation from the tabulated value of the enthalpy of formation, and it is here that the problem is encountered. The purpose of this report is to clarify the meaning of the internal energy of formation as it relates to thermodynamic equilibrium calculations, especially as it pertains to calculations which employ the standard computer programs now in use.

2. Background

Two examples from the literature are selected below to illustrate the divergent views. Before proceeding with these examples, some basic definitions and conventions are first reviewed.

The enthalpy, H , is a convenience function applicable to constant pressure processes and is defined as

$$H \equiv U + PV \quad (1)$$

where U is the internal energy, P is the pressure, and V is the volume. From this it follows that

$$\Delta H = \Delta U + \Delta(PV)$$

and

$$\Delta H_f = \Delta U_f + \Delta(PV)$$

where ΔH_f and ΔU_f are the enthalpy and internal energy of formation respectively.

The *energy*[†] that is customarily tabulated is the standard enthalpy of formation (sometimes called the heat of formation), ΔH_f° , defined (Chase et al. 1985) as "... the increment in enthalpy associated with the reaction of forming the given compound from its elements in their reference states, with each substance in its thermodynamic standard state at the given temperature." By this convention, for all elements in their standard state, $\Delta H_f^\circ=0$ at all temperatures. The superscript denotes that the pressure is for the standard state. In older tabulations, the standard state (pressure) is usually 1 atm; more recent compilations, e.g. the JANAF tables (Chase et al. 1985), use a pressure of 1 bar (= 0.1 MPa) for the standard state, and convert the older values based on 1 atm to this new standard state pressure. It is also customary and convenient to designate an "absolute" *energy* scale by assigning the enthalpy of the stable form of elements at the standard pressure, hereafter designated as p° , and a reference temperature, usually 298.15 K, a value of zero, i.e., $H^\circ_{298.15} = H(298.15 \text{ K}, p^\circ) = 0$. There actually is no absolute *energy* scale; however, by designating the standard state (p°) and the reference temperature, and assigning the enthalpies of the reference elements, an effective "absolute" scale is created. Hereafter in this report, the designation *assigned*^{††} applied to either an enthalpy or internal energy will mean an *energy* which is based on this chosen reference state, i.e., $H(298.15 \text{ K}, p^\circ)=0$ for elements in their stable form of aggregation at 298.15 K and p° . For a change of state, a negative value of ΔU or ΔH means that energy is evolved by the process.

The relationship between the standard enthalpy of formation, as it is defined, and the assigned enthalpy is illustrated in Figure 1. $(\Delta H_f^\circ)_T$ is the enthalpy of formation at some temperature T for which the reactant elements of the formation reaction have some arbitrary enthalpy value. Also shown is the same $(\Delta H_f^\circ)_T$ on the assigned scale, where it is displaced by an amount equal to $(H^\circ_{\text{reactants}})_T - (H^\circ_{\text{reactants}})_{298.15\text{K}} = (H^\circ_{\text{reactants}})_T$. The quantity $(\Delta H_f^\circ)_T$ has the same value on any *energy* scale; in general, however, the assigned enthalpy $H^\circ_T \neq (\Delta H_f^\circ)_T$ for a temperature other than the reference temperature, in this case $T = 298.15 \text{ K}$, of the assigned scale. [The situation illustrated in Figure 1 is typical for $(\Delta H_f^\circ)_T$ when $T > 298.15\text{K}$]. Also note that, owing to its definition, discontinuities can occur in ΔH_f° if the stable form of aggregation of an element passes a phase-transition temperature (Chase et al. 1985).

U and H can be expressed as functions of any two other state properties. Choosing the appropriate natural variables gives

$$U=U(T,V) \quad (2a)$$

and

$$H=H(T,P). \quad (2b)$$

For ideal gases, H and U are rigorously only functions of T. In general, however, as indicated in Eq. 2, H and U also have a pressure and volume dependence, respectively; if the initial conditions

† The term *energy* (italicized) will be used in this report to refer to either internal energy or enthalpy.

†† This designation is also used in the NASA-Lewis documentation (Gordon and McBride 1971).

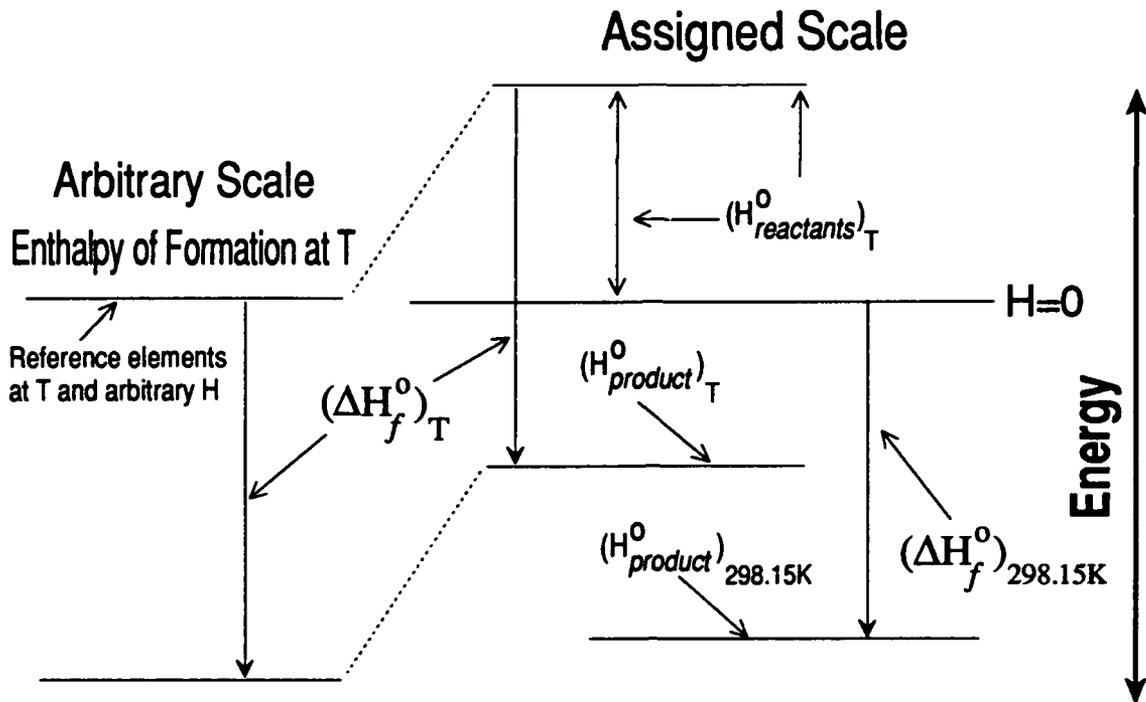


Figure 1. Schematic energy diagram showing the relationship of the enthalpy of formation at some arbitrary temperature, T, to the assigned energy scale. The enthalpy of formation at 298.15K is also shown for comparison.

for a calculation are other than those of the tabulated standard enthalpy of formation, then its value or the corresponding internal energy must be adjusted, as necessary, for the specified initial conditions of temperature, pressure, or volume. For example, on the assigned energy scale, for standard pressure p° , and reference temperature T_{ref} ,

$$H^\circ(T) = \Delta H_f^\circ(T_{\text{ref}}) + \int_{T_{\text{ref}}}^T C_p^\circ(T) dT$$

where $C_p^\circ(T)$ is the standard state heat capacity. However, for $P \neq p^\circ$,

$$H(T, P) = H^\circ(T) + \int_{p^\circ}^P \left(\frac{\partial H}{\partial P} \right)_T dP$$

Unless otherwise noted, in the following discussions all compounds will be considered to be in the standard state, *i.e.*, $P=p^0$, and at the reference temperature $T=298.15$ K. The two representative approaches are now summarized. Both cases have the same expressed purpose: deduce the value of the internal energy of formation from the enthalpy of formation for a condensed phase.

2.1 Case 1

In this approach (Freedman 1988), it is noted that the internal energy, U , is the appropriate variable for constant volume calculations, and that the internal energy of formation, ΔU_f , is the "correct input datum", it is pointed out that the difference between the ΔH_f and ΔU_f *i.e.* the PV contribution, is small, and is less than 120 cal/mol for typical propellants; this value is calculated for a pressure of 1 atm and a maximum value of 0.005 m^3 for the molar volume of a propellant ingredient.[†] Furthermore, it is argued, this small a difference is less than the uncertainties (*ca.* 0.3-2 kcal/mol) of tabulated values of the enthalpies of formation. For condensed phases, therefore, it is correct to use the value of the enthalpy of formation for the internal energy of formation when the latter quantity is required.

2.2 Case 2

In the other approach (Volk and Bathelt 1990), the need for the energies of formation, rather than the enthalpies of formation, when performing constant volume calculations for gun propellants is also noted. The energy of formation, ΔU_f , is calculated using the standard formula:

$$\Delta U_f = \Delta H_f - \Delta n R T = \Delta H_f - \Delta n (0.59248) \text{ (kcal/mol)},$$

where Δn is the difference in the number of moles of gases, *i.e.* $n_{\text{products}} - n_{\text{reactants}}$, R is the universal gas constant, and T is the absolute temperature. [$RT=0.59248$ kcal/mol for $T=298.15\text{K}$]. The term ΔnRT is a standard approximation for $\Delta(PV)$ assuming, as in the first case, that the PV product of condensed phases is negligible. A sample calculation is performed using this approach for the internal energy of formation of dibutyladipate, a liquid, at $T=298.15$ K, chemical formula $\text{C}_{14}\text{H}_{26}\text{O}_4$, $\Delta H_f=-181.7$ kcal/mol, for the formation reaction



Since $\Delta n = 0 - 15 = -15$, the ΔU_f is readily calculated to be -172.8 kcal/mol. The difference, in this approach, between ΔU_f and ΔH_f is $\Delta nRT = 8887.6$ cal/mol.

[†] To take an actual example for comparison, at $P=1$ atm, for HMX which has a density $\rho=1.9 \text{ g/cm}^3$, a molecular weight $\text{MW}=296.17 \text{ g/mol}$, and using $V=\text{MW}/\rho$, the PV product is just 3.78 cal/mol.

The two methods for calculating ΔU_f° above give results for the related quantity *enthalpy of formation - internal energy of formation* which, when compared, are seemingly in sharp disagreement. Case 1 estimates a maximum difference of 120 cal/mol for a typical propellant. The method used in Case 2 gives, for a liquid, 8887.6 cal/mol. Since both methods are dealing with a condensed phase substance, this large a disagreement, 120 cal/mol versus 8887.6 cal/mol, is not expected.

3. Discussion

The resolution of this discrepancy can best be addressed by reference to a schematic energy level diagram. For convenience, and by way of illustration, the internal energy and enthalpy of dibutyladipate, which was calculated in the second example above, will be used. In Figure 2 the

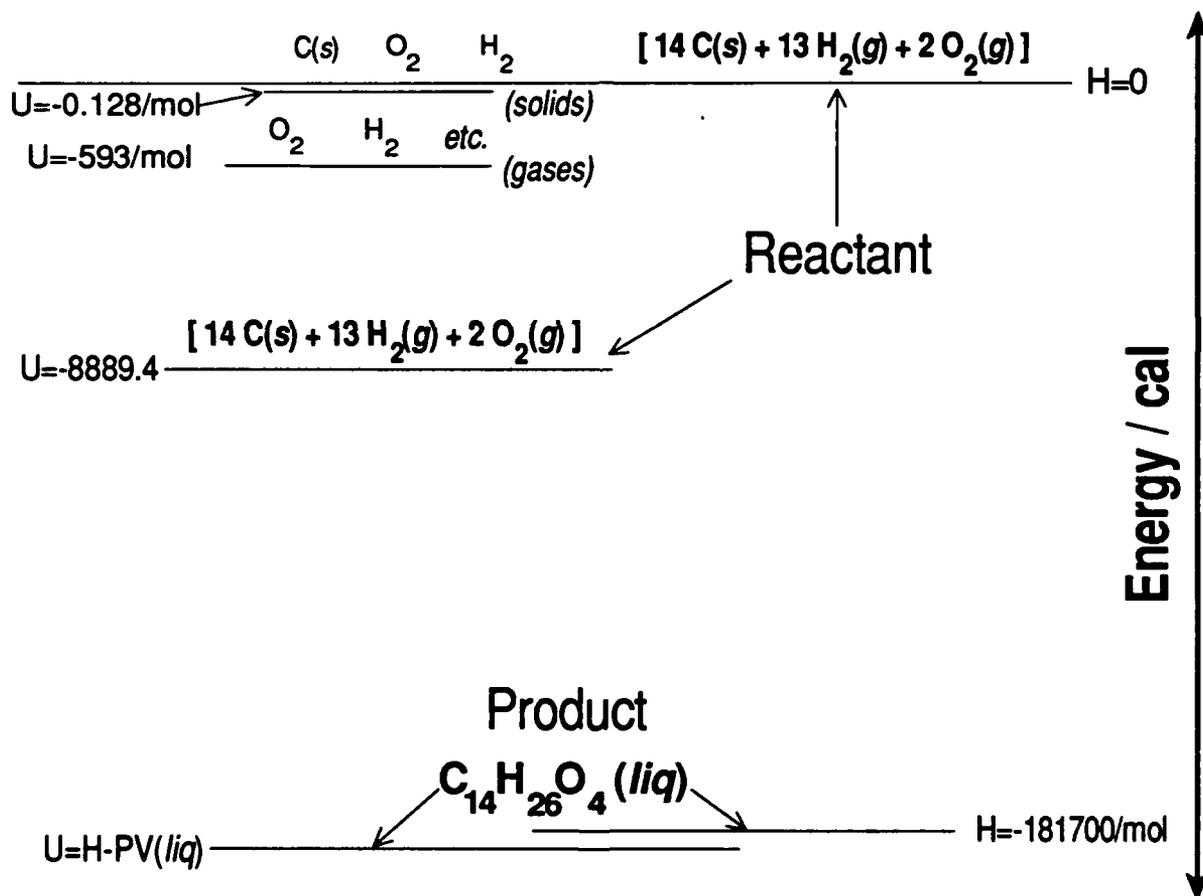


Figure 2. Schematic energy diagram showing the enthalpy and internal energy states involved in the formation dibutyladipate.

energies of the various compounds are drawn schematically on a diagram that is only intended to convey large and small differences in the quantities, rather than provide a precise scale. The elemental species needed to form the dibutyladipate are listed at the top of the figure at the zero energy reference state. In addition, the reactant composition for the formation reaction is also

indicated, and is included in brackets for identification; it also has an $H=0$, which is just the sum of its component enthalpies. Just below this level, at -0.128 cal/mol, is a representative U for an elemental solid, where the particular value chosen is for carbon. This and subsequent U values are all calculated using Eq. 1. The small difference between the internal energy and enthalpy of the solid carbon is just the PV product term; this was calculated using (Lange's Handbook of Chemistry 1979) a density, $\rho=2.267$ g/cm³, a molecular weight $MW=12.011$ g/mol, and the relation $V=MW/\rho$. The third level down, $U=-593$ cal/mol, corresponds to the $PV(=RT)$ of one mole of an ideal gas. The next (fourth) level at $U=-8889.4$ cal is the total internal energy of the reactants needed to form dibutyladipate. Since, as noted above, $H=0$ for this reactant composition, U is just the negative of the sum of the PV product of each species, where each mole of (assumed ideal) gas contributes $RT=593$ calories and each mole of solid carbon contributes 0.128 calorie. These four levels schematically represent all the internal energy and enthalpy states of the reactants.

The next two levels represent the enthalpy and internal energy for the product compound dibutyladipate. The upper level of the two, at $H=-181.7$ kcal/mol, is derived from the tabulated value² of the enthalpy of formation and the change of state given in Case 2 above, where, for the dibutyladipate, $H = \Delta H_f + H_{reactants} = -181.7 + 0.0 = -181.7$ kcal/mol. The next slightly lower level represents the corresponding internal energy value of dibutyladipate. For this level $U=H-PV(liq)$, where $V(liq)$ is the molar volume of the liquid phase dibutyladipate. As implied in both cases presented above, at pressure p^0 , the value of $PV(liq)$ is expected to be much less than $H_{product} = -181700$ cal/mol. Neglecting the small $PV(liq)$ contribution gives $U=H$.

The levels in Figure 2 represent the reactant and product assigned *energy* states for the formation of dibutyladipate. These levels are duplicated in Figure 3. Additionally in Figure 3, vertical arrows are drawn to indicate the states involved in forming the ΔU_f and ΔH_f for dibutyladipate. Referring to Figure 3, and using the states involved in the formation reaction of Case 2, it follows that, for the product, $H=\Delta H_f$ exactly, however, $U=\Delta U_f + (PV)_{reactants}$. Because of the choice of reference state, the assigned value of the product enthalpy, $H_{product}$ and the enthalpy of formation, ΔH_f , are always equal. In general, however, the assigned internal energy, $U_{product}$ and the internal energy of formation, ΔU_f , are not equal, differing by $(PV)_{reactants}$ which is not usually negligible if gaseous reactants are involved.

In summary, for compounds which are used as a reactant component in an equilibrium calculation, at the standard state pressure and reference temperature (here 298.15K), on the assigned *energy* scale, $H^0 = \Delta H_f^0$ and $U^0 \neq \Delta U_f^0$; also, for condensed phases only, usually, $U^0 \approx H^0$.

Two commonly used thermodynamic chemical equilibrium codes, NASA-Lewis⁴ and BLAKE (Freedman 1981), calculate a standard enthalpy, denoted $H^0(T)$, which is a function of the temperature and a set of coefficients. These $H^0(T)$ values are assigned enthalpies from which assigned internal energies are calculated. These codes, therefore, use an assigned *energy* scale for referencing both the enthalpy and internal energy of reactants and products; the input data, *i.e.* the

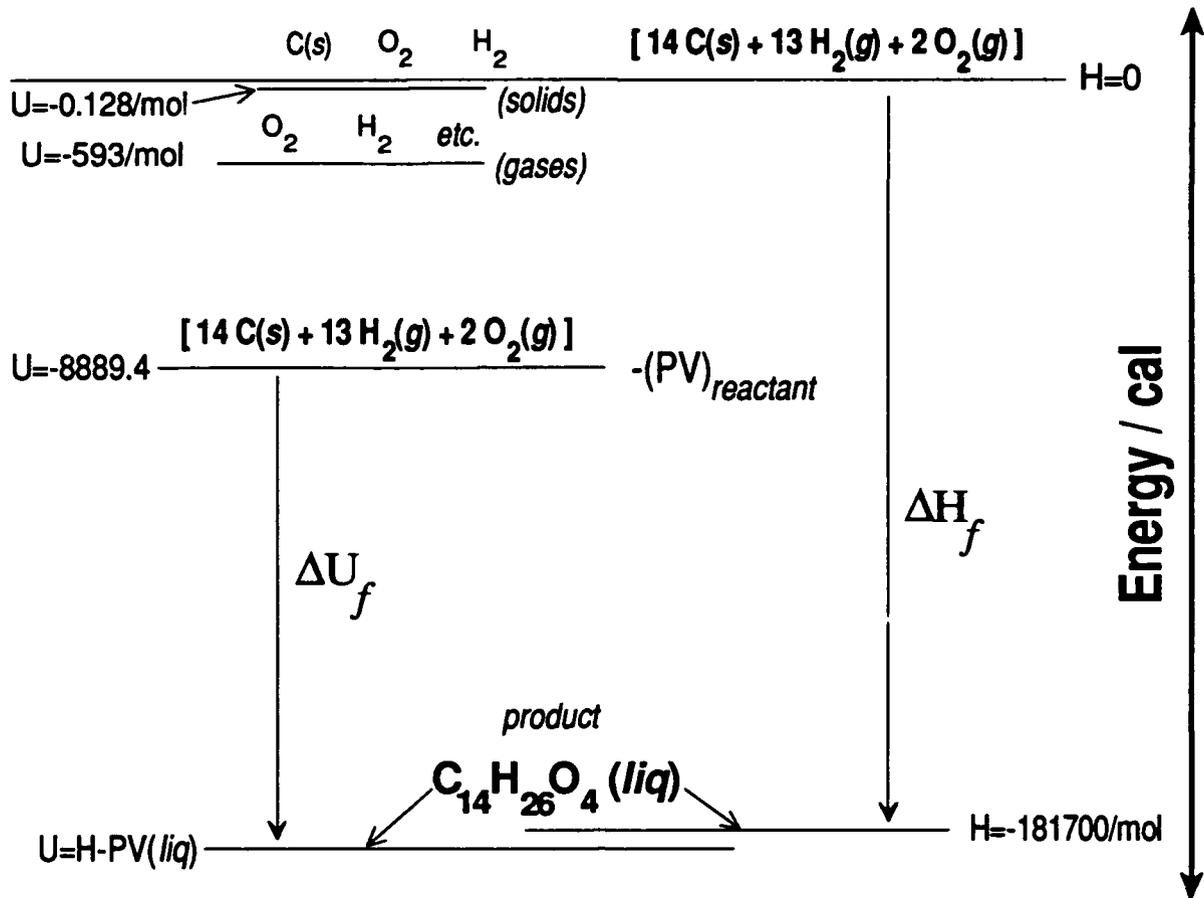


Figure 3. Same energy levels as Figure 2, but with ΔH and ΔU also indicated.

internal energy or enthalpy of the propellant components, must correspond to this scale. Formally, U and H represent these assigned *energies* while ΔU and ΔH do not.

4. Conclusions

1. Because the choice of reference state is, by convention, based on the reactants of the formation reaction having an assigned enthalpy value of zero, it follows that, at the reference temperature, for the product of formation, $H^\circ = \Delta H_f^\circ$.
2. The quantity $(\Delta H_f^\circ)_T$ has the same value on any *energy* scale; in general, however, the assigned enthalpy $H^\circ_T \neq (\Delta H_f^\circ)_T$ for a temperature other than the reference temperature
3. Because of this assigned *energy* scale, in general, for the same product also at the reference temperature, $U^\circ \neq \Delta U_f^\circ$.
4. The approach in Case 2 above correctly calculates ΔU_f° , but this is not the quantity that is likely to be required by a thermodynamic equilibrium code; for instance, it is not the quantity required as input by two widely used codes, BLAKE and NASA-Lewis.

5. The relationship between the internal energy and enthalpy of condensed phases as stated in Case 1 is also correct, however, it pertains to the assigned *energies*, U° and H° , and not to ΔU_f° and ΔH_f° .
6. In general, for condensed phases referenced to the conventional *energy* scale having $H^\circ=0$ for elements in their standard state, $U^\circ \approx H^\circ$ and $\Delta U_f^\circ \neq \Delta H_f^\circ$.

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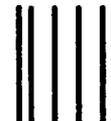
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