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The Thermodynamics of Interior Ballistics and Propellant Performance

by Anthony J. Kotlar

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14. ABSTRACT

The equilibrium thermodynamic properties of propellants are routinely calculated by standard computer codes such as NASA-Lewis, BAGHEERA, or BLAKE. This thermodynamic information is also "packaged" by the interior ballistic (IB)-oriented codes BLAKE and BAGHEERA and transferred to IB performance codes via several parameters (mainly γ, impetus [I], and the covolume). A related quantity called the chemical energy [$CE = I/(\gamma - 1)$] is also usually calculated, since it is sometimes viewed as one of the traditional measures of propellant performance. In cases like the electrothermochemical (ETC) gun, where energy (electrical) is added to the system, the CE appears not to adequately account for the additional energy. This study takes a critical look at the concept of CE as it is calculated for use in IB applications. Using a JA2 + 1-kJ/g system as an example, the missing energy, as reflected in the value of the CE, is found to be tied up in a composition change of the system; the energy scale used for the CE and IB calculations is incapable of directly accounting for this chemical effect. The relationship of the IB energy scale to the thermochemical energy scale is discussed. Using BLAKE (which is based on the thermochemical scale), the energies of several changes of state are calculated as alternatives to (as opposed to substitutes for) the CE. Also, for an often-used idealized gun model—constant breech pressure before burnout, Lagrange gradient, no losses—the processes delineating propellant performance (and the conditions at muzzle) are calculated via BLAKE using full variable-composition equilibrium thermodynamics.

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1. Introduction

Equilibrium thermodynamics have traditionally provided the parameters that delineate the energetics that are needed by interior ballistic (IB) calculations or to quantify propellant performance (*1*, *2*). These equilibrium thermodynamic properties of propellants are routinely calculated by standard computer codes such as NASA-Lewis (now NASA-Glenn) (*3*), BLAKE (*4*), or BAGHEERA (*5*). BLAKE, in particular, is widely used in the ballistic community and has been customized for gun-oriented calculations. In the traditional approach, the complete thermodynamic information is "packaged" by BLAKE for subsequent transfer to IB codes via several parameters (mainly the ratio of the "frozen" heat capacities γ, impetus I, and the covolume). A related quantity called the chemical energy [$CE = I/(\gamma - 1)$] is also usually calculated since it is sometimes viewed as a quantity that is an independent measure of propellant performance. The detailed level at which the thermodynamic codes calculate a propellant's thermodynamic parameters is, consequently, condensed and reformulated as several select quantities, some as "lumped" parameters that are routinely used in interior ballistic applications.

This report explores the implications and limitations of using these traditional IB thermodynamic quantities. In doing so, it utilizes thermodynamic quantities that are evaluated at the level of the BLAKE (truncated virial equation of state [EOS]) code itself. Two different energy scales are encountered, and their relationship is explored, especially regarding the role of propellant chemistry. A survey of well-defined propellant energetics is proposed for comparison with the CE. Two topics are presented to illustrate this use of the full-code-level variable-chemistry thermodynamics: (1) an energy-accounting anomaly exhibited by the CE in electrothermochemical (ETC) systems (*6*), where additional energy is added to the propellant, is clarified, and (2) propellant performance is explored in the context of an often-used, idealized, constant breech pressure before-burnout Lagrange pressure gradient, no-loss, model gun system (*7*).

2. The Thermodynamic Quantities of IB

2.1 Thermodynamic Packaging

IB codes, such as NOVA (*8*), IBHVG2 (*9*), CONPRESS (*10*), and typical closed-bomb reduction programs (*11*), use the standard lumped thermodynamic parameters. The choice of these parameters appears to be dictated in part by the acceptance in IB calculations of the Noble-Abel (covolume) EOS, $P(V - b) = nRT$, where b is the covolume. The other parameters are $\gamma \equiv C_P/C_V$, the ratio of the frozen specific-heat capacities at constant pressure and volume, respectively, and the impetus (or force constant) $I \equiv (W_{gas}/W_{Total})(RT/M)$, where M is the average molecular weight of the gas-phase combustion products and W refers to the weight (mass) of the propellant. These parameters are usually evaluated for some assumed, average, or generally accepted loading density (commonly 0.2 g/cm^3) under conditions of constant volume and energy. This is referred to as the "chamber" condition, where the propellant is all burnt. IB codes typically use the quantities b and γ as invariant over the course of the ballistic cycle and fixed at the chamber values. Thus, the combustion products are an inert working fluid (i.e., no subsequent chemistry transpires) and, furthermore, there is no variation of b and γ with temperature. In contrast to this "abridged" picture, the "full" thermodynamics of BLAKE uses a truncated virial EOS, has complete temperature variation of γ, and can allow for further reactions to take place in the working fluid. The contrasting approaches are shown schematically in figure 1.

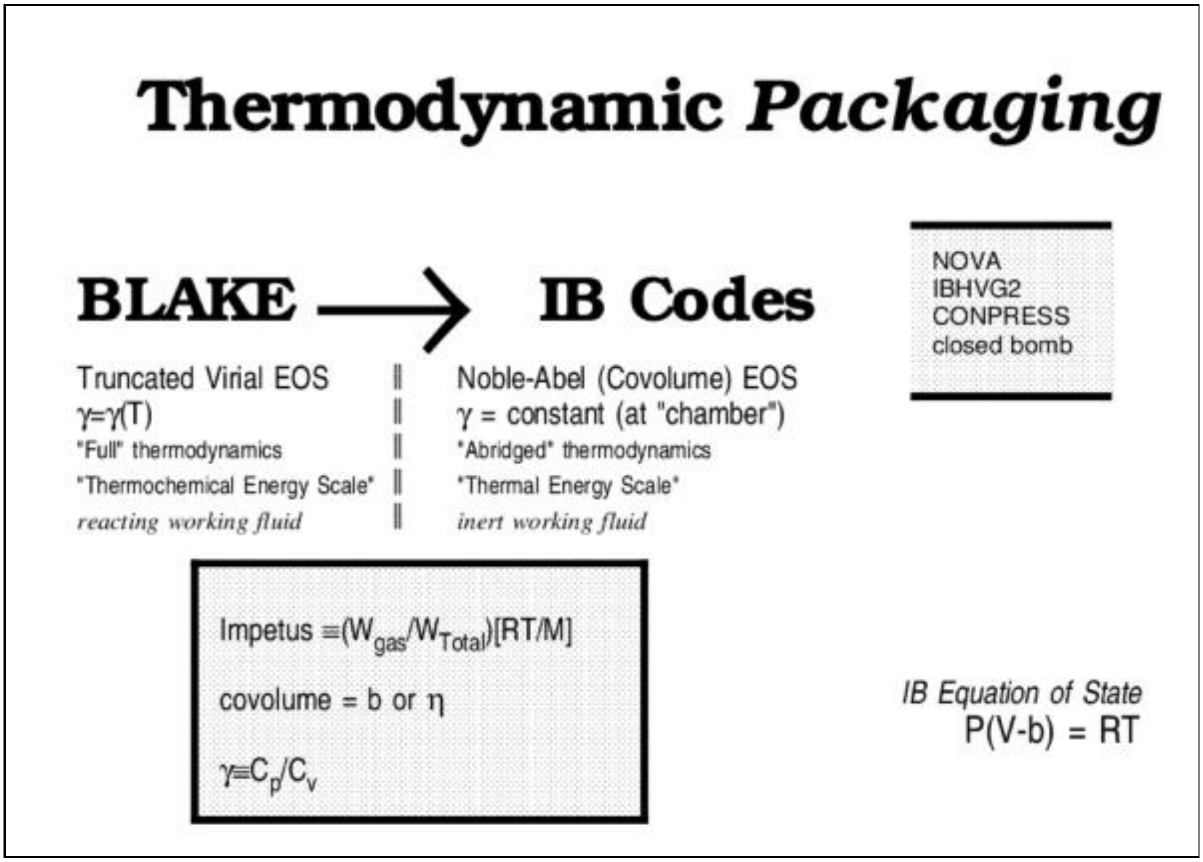

Figure 1. Relationship of BLAKE-level thermodynamics to the standard IB thermodynamic description based on the Noble-Abel EOS.

2.2 “Chemical” Energy: Ballistic Energy

Another quantity used in IB is the so-called “chemical” energy. The CE has been associated with the work performed by the chamber gases in expanding to infinite volume along a reversible adiabatic path (*12*). CE as defined previously is also just C_V T; this implies an energy change $C_V \Delta T = C_V (T - T_{ref})$, where the reference temperature $T_{ref} = 0$. This establishes the IB energy scale as based on the energy being zero at absolute zero of temperature. Hence, the condition for calculating the CE based on maximum work (infinite volume or T = 0) corresponds to the “absolute” energy of the IB scale, which is arbitrarily referenced to T = 0. Conveniently, then, the energy of the reference state or the final state of the system need not be specified since its value is zero. And, since the final state of the system along the reversible adiabatic path is at the absolute zero of temperature, the energy difference ΔU identically equals the maximum work. Thus, within the traditional IB formalism, the CE of the chamber gases is also a measure of the maximum work that can be done by those gases; however, the final state is unattainable. Also, there is a bias in the CE in that the energy associated with the temperature range from absolute zero to ambient is also included, energy that is never actually used and, with the final state having T = 0 K, is only formally applicable.

3. The Two Energy Scales

The identification of U = 0 at T = 0 establishes an absolute energy scale that is implicit in most IB calculations. However, this scale is not used in the standard thermodynamic equilibrium codes. If the IB scale can be considered a “thermal” scale, then the energy scale used by BLAKE, for example, can be considered a “thermochemical” energy scale. To compare these two energy scales, and in the calculations that follow, a system’s composition is specified as either:

- “Static” (i.e., frozen, nonequilibrium, inert, nonreacting, infinitely slow kinetics designated by [ ]) or
- “Dynamic” (i.e., equilibrium, reacting, infinitely fast kinetics designated by { }).

A constraint on the composition (static or dynamic) and two thermodynamic state variables (e.g., P, V, T, U, H, S) are sufficient to specify the state of the system. The system here is either all the matter comprising the propellant if the volume is not held constant or everything inside a specified constant volume. Ambient conditions are taken to be 0.1 MPa ≈ 1 atm and T = 298.15 K; these also correspond to the standard state pressure and the reference temperature of the thermochemical energy scale.

3.1 IB Energy Scale

The IB scale is illustrated in figure 2. A propellant at ambient conditions (its temperature and heat capacity determine its energy) and a specified loading density (or specific volume) goes to chamber conditions at the same loading density. All energies are referenced to the state $T = 0$. As indicated, the conditions for the change of state are constant volume and adiabatic (this corresponds to a GUN calculation in BLAKE). In this picture, the chamber gases are at a higher energy, qualitatively, than the propellant. Also, since the reference energy is zero for all compositions (which are always static), the absolute energy is just the change in energy (i.e., $U = \Delta U$). Therefore, on this IB energy scale, it is not necessary to explicitly denote the reference state since its energy value is always zero for all compositions. In addition to a selection of energetics, the choice of a loading density is also the choice of a composition. Unfortunately, this can obscure the fact that even on a designated absolute energy scale an energy difference involving a final state of the system is still implied.

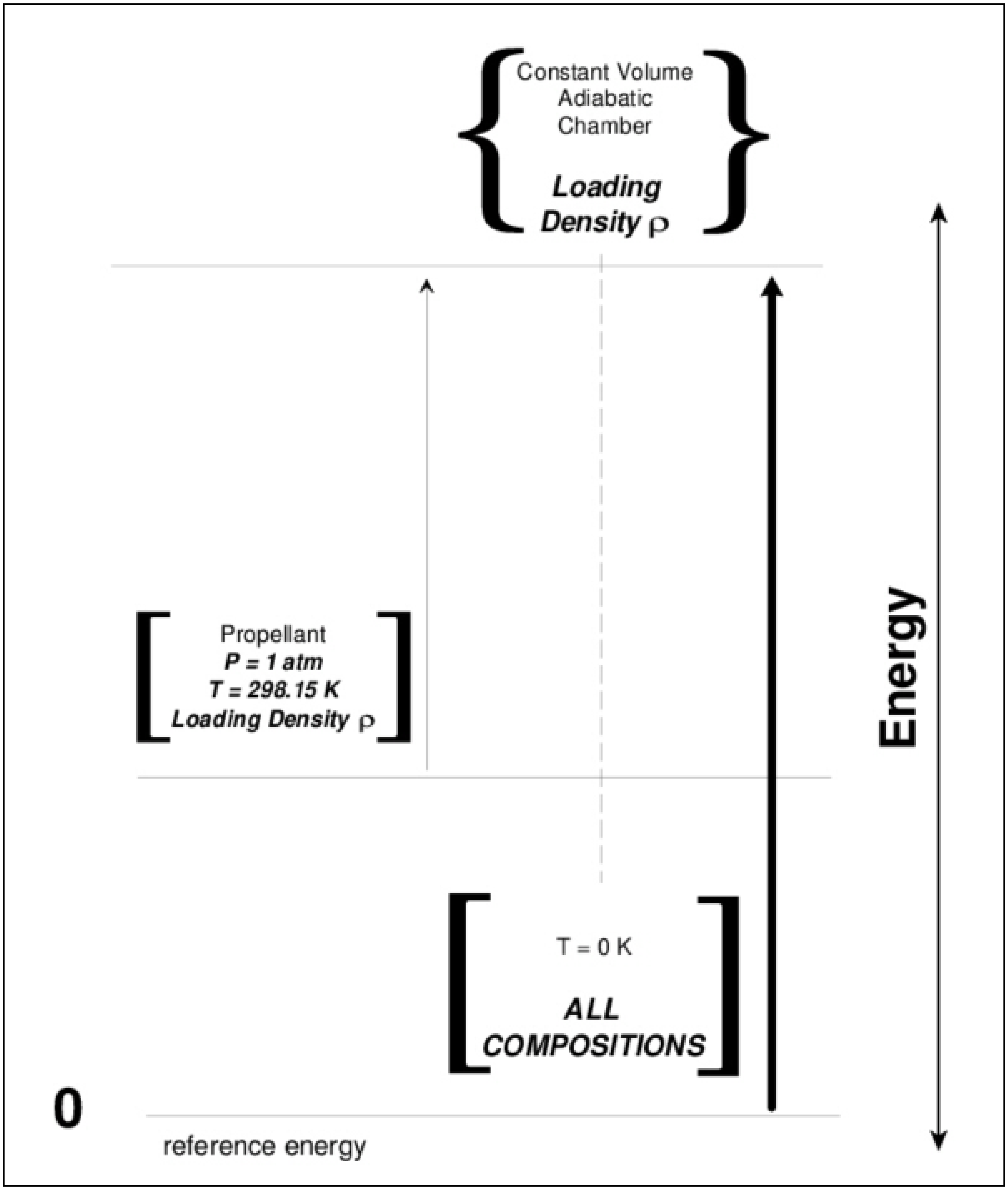

Figure 2. Traditional IB energy scale.

3.2 Thermochemical Energy Scale

In contrast, the same change of state on the thermochemical energy scale is shown in figure 3; energy values are provided for the propellant JA2, which is used later as an example. On this scale, the reference state is the enthalpy of the stable form of the elements in their standard state (0.1 MPa); and, for convenience, a reference temperature is also chosen (usually 298.15 K). This represents another absolute energy scale. It is the "assigned" energy scale (*13*) used by the NASA-Lewis, BLAKE, and BAGHEERA codes, and most other chemistry-oriented codes. Except for the designation of the reference temperature, it is also the energy scale of the JANAF tables (*14*). A change of state involving the reference condition as reactant going to product at constant temperature and pressure is just the formation reaction for which the enthalpy change is the familiar standard enthalpy ("heat") of formation, $\Delta H^o{}_f$. Based on this energy scale, the process of the propellant going to products at chamber conditions adiabatically and at constant volume (the same situation illustrated previously on the IB scale) is isoenergetic, that is, the propellant and the chamber gases have the same total energy. This energy manifests itself differently; however, in the case of the propellant, it resides in the chemical bonds and, in the combustion gases, it is primarily in the form of thermal energy. Also, for this process, all gas-phase loading densities are at the same energy; however, different compositions are produced depending on the specified value of the loading density. Thus far, in contrast to the IB approach, the choice of loading density is only a selection of the composition; since the final state of the system has not been specified, the value of the energy, ΔU, cannot be determined.

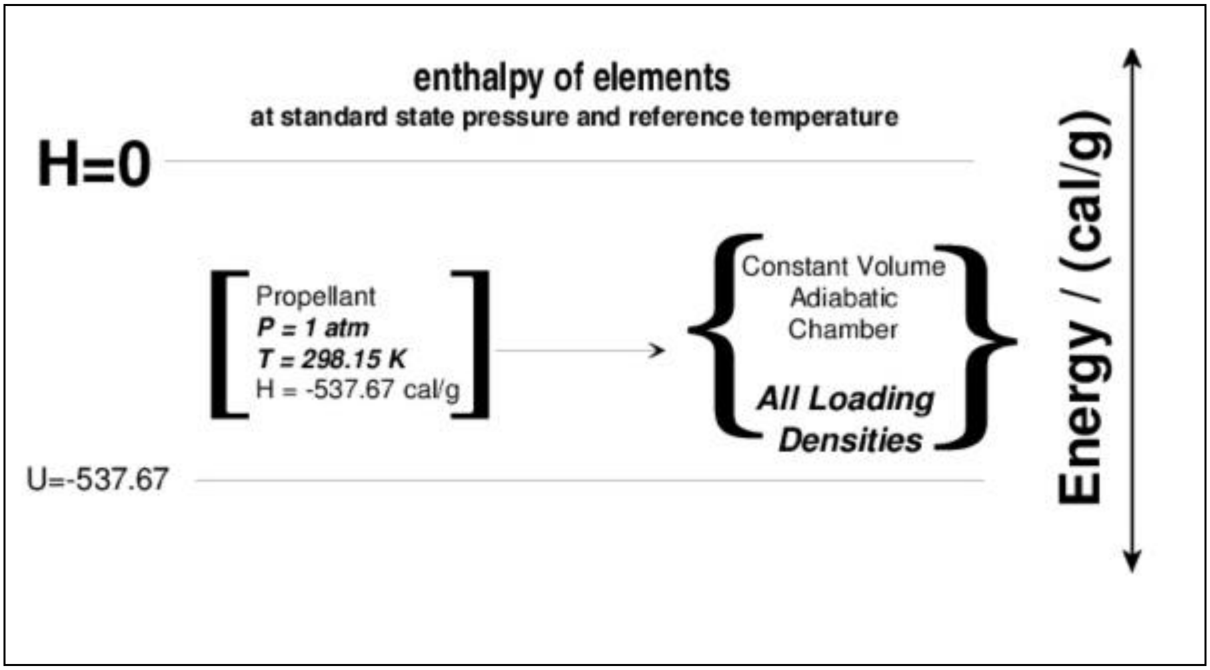

Figure 3. Thermochemical energy scale.

To evaluate propellant energetics on this scale it is necessary to construct the final state of the system, as shown in figure 4. The chamber composition is specified as either static or dynamic, and two state variables are given to fix the final state of the system. Unlike the IB energy scale case, where the final state effectively disappears from consideration, on this scale, the necessity of choosing meaningful final states is of utmost importance. The energy of the change of state, ΔU, is the difference in energy of the two states that denote the process. As shown, this would be either the propellant or the isoenergetic chamber composition going to the static or dynamic composition with two thermodynamic state variables specified.

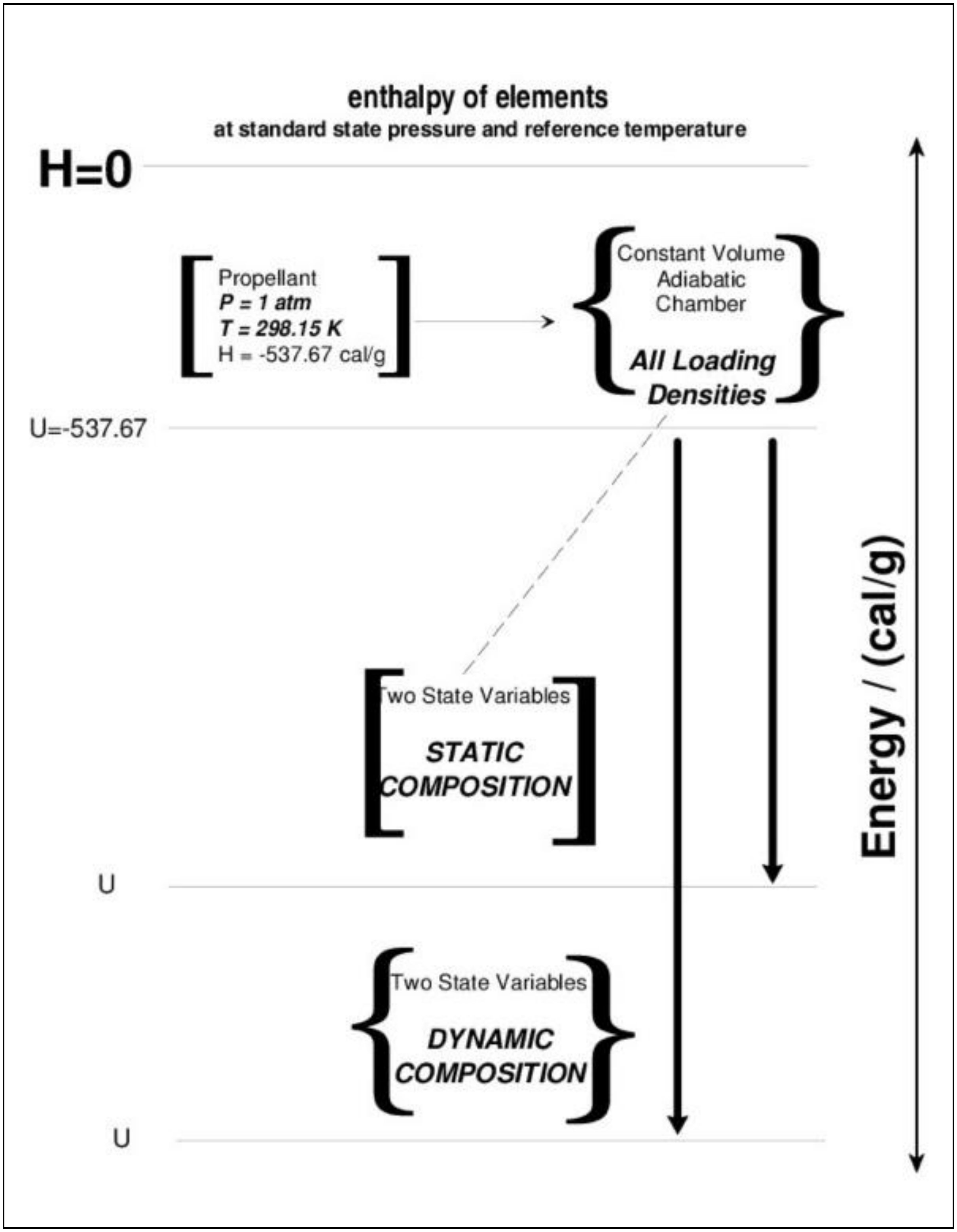

Figure 4. Selection of the final state on the thermochemical energy scale.

3.3 The Use of a Thermodynamic Equilibrium Code

The means of actually performing the calculations must be chosen before proceeding to discuss a survey of propellant energetics. The methods and formalism used throughout this work are general considerations of thermodynamics. To quantify this formalism the apparatus for performing the calculations must be equally general. The code BLAKE is employed here because it is widely used, it is familiar to many in the ballistics community, and it has the necessary generality inherited from the TIGER (*15*, *16*) code, although it has also been customized (the GUN option) for IB applications. Examples of calculations for the remainder of this work are performed using BLAKE and illustrated using BLAKE commands as concrete examples of the required computations. It must be emphasized, however, that the methods are general and are not code specific. Any general thermodynamic code could be used, and, in fact, BLAKE itself was not modified in any way to perform the calculations. The general requirement is that the equilibrium state be calculated for either specified static or for dynamic compositions (MELT or FREEZE commands), that the final state of one part of a process be used in a subsequent calculation as the initial state of the next part of the process, and that a good selection of two state variable combinations be available for use (ISOLINE and POINT commands and their list of variable combinations).

4. Survey of Energetics

When the approach shown in figure 4 is used, a survey of energetics for several processes is calculated and the results are given in table 1. The chamber composition was calculated for the typical value of loading density (ld = 0.2 g/cm^3) used in IB. This state can be calculated using the GUN command or, for example, as a POINT command, where the U and V are specified and constant as $U = \Delta H^o_f$ and $V = 1/ld$. This state is designated as the chamber state in the table. This chamber composition is then allowed to react (dynamic, MELT) or is fixed or frozen (static, FREEZE), and the state is recalculated for the two specified thermodynamic variables. Three sets of variables were used to specify the final state: VT, TP, and SP. For each of these the final state was calculated for both dynamic and static compositions. Two other entries, the CEs, are calculations of the traditional “chemical energy” and are included for comparison and discussed later.

In the table 1 example, a JA2 formulation was used and well-posed energetics are rigorously calculated from the energy state function, which comes directly from the code as output. The table of results is followed by an explanation of each entry. For the states in the table, ambient is 1 atm, 298.15 K. The process is:

INITIAL CHAMBER FINAL

Propellant (298.15 K, 1 atm, V_o) → Adiabatic Composition (T_{ad}, P_{ad}, V_o) → Final State (T, P, V).

Table 1. A survey of propellant energetics illustrated for JA2 at a 0.2-g/cm^3 chamber loading density.

Thermodynamic State	**P (atm)**	**V (cm^3/g)**	**T (K)**	**H (cal/g)**	**U (cal/g)**	**U (J/g)**	**−ΔU (J/g)**	**S (cal/K)**
Chamber	2844.63	5.00	3448	−193.22	−537.67	−2249.61	0	2.213
VT Dynamic	49.44	5.00	298	−1914.51	−1920.49	−8035.33	5785.72	1.186
TP Dynamic	1.00	748.62	298	−1877.85	−1895.98	−7932.78	5683.17	1.531
CE Standard Static	0.0	∞	0	—	—	—	5134.50	—
CE New Static	0.0	∞	0	—	—	—	4408.00	—
VT Static	131.59	5.00	298	−1538.51	−1554.45	−6503.82	4254.21	1.488
TP Static	1.00	983.98	298	−1509.58	−1533.41	−6415.79	4166.18	1.984
SP Dynamic	1.00	2850.95	892	−1422.39	−1491.43	−6240.14	3990.53	2.213
SP Static	1.00	2281.74	690	−1385.43	−1440.69	−6027.85	3778.24	2.213

Since the initial and chamber states are isoenergetic, the change in internal energy is the same for INITIAL → FINAL and CHAMBER → FINAL. In this approach the final state is the key to the energetics, and as stated before, it is the essential concept that is inadvertently lost or obscured in the CE. The relevant final state is best identified by considering the operating conditions of the gun system. Here, an independent measure is sought, if possible, for propellant performance; so, the final state is, instead, not permitted to go below ambient. This is a limiting condition but it is a reasonable limit; a gun or device to measure performance is not expected to operate into a regime that is below ambient (T, P). The first step of the process (adiabatic, constant internal energy, constant volume) gives the working fluid's initial state (chamber condition) including the all important composition. That working fluid then goes to some well-defined final state for which the unspecified thermodynamic state functions are readily calculated by the code. It can go to the chosen final condition (1) as an inert fluid (i.e., static composition) or (2) allow the composition to react (i.e., dynamic). BLAKE calculates these states exactly and directly, as shown next.

The entries in table 1 are in decreasing order of the difference in internal energy (−ΔU) between chamber or propellant and final states. This is the key quantity (i.e., the amount of energy that is released [−ΔU > 0; it is written this way to conform to the sign convention used for the CE] by the chamber composition or the propellant in going to the final state).

4.1 Table Explanation

The first two letters indicate the two state variables that define the unique state, except for CE; these are followed by the constraint on the composition. If static, the composition is fixed as that of the chamber condition; if dynamic, the constraint is that the composition is at equilibrium for the given state variables. For the two CE calculations, the compositions are static only. It must be emphasized that the specified initial volume, V = 1/ld, is the physical volume of the chamber and not the volume occupied by the solid propellant.

4.1.1 VT Dynamic {VT}

The volume V is constant and no work is done; the energy is the heat. This is the constant volume analog of the "heat of reaction," which in this case, is an internal energy of reaction. One variation is that the final composition is the equilibrium composition at the specified conditions and not the composition of the chamber. This ΔU is the heat (internal energy) of reaction for Propellant (T = 298.15 K, V) → Equilibrium Composition (T = 298.15 K, V).

4.1.2 TP Dynamic {TP}

The system (the working fluid) is returned to ambient with a new equilibrium composition, and it also has expanded to a new volume. This returns the system to ambient, which is the initial condition of the propellant: Propellant (T = 298.15 K, 1 atm) → Equilibrium Composition (T = 298.15 K, 1 atm).

4.1.3 CE Static

This is the traditional CE ($I/(\gamma - 1)$) with T = 0 for the final state. The composition is always static as that of the chamber condition, and the thermodynamics are invariant over the specified range of T and P.

4.1.4 CE New

A second value using a new approach (*17, 18*) for calculating CE (vide infra) is also listed.

4.1.5 VT Static [VT]

This is similar to VT Dynamic, but it defines a different reaction since the composition is static, that is, Propellant (T = 298.15 K, V) → Chamber Composition (T = 298.15 K, V). This is an exact calculation of the CE-type quantity, which is adjusted to T = 298.15 K.

4.1.6 TP Static [TP]

This is similar to TP Dynamic, but the final composition is that of the chamber: Propellant (T = 298.15 K, 1 atm) → Chamber Composition (T = 298.15 K, 1 atm).

4.1.7 SP Dynamic {SP} and SP Static [SP]

The system expands to ambient P at constant entropy. The final temperature is above ambient in each case. The state with ambient T would have a P below ambient, which is excluded from consideration, as previously stated.

4.2 Calculation of Work

As mentioned previously, the process that best measures the CE of the chamber composition in the sense of the IB concept of CE is VT Static. The reason is that the state function, ΔU, under these conditions, is the measure of the heat evolved by the reaction

$$\text{Propellant (298.15 K, } V_{\text{Chamber}}\text{, 1 atm)} \rightarrow \text{Chamber Composition (298.15 K, } V_{\text{Chamber}}\text{, P),}$$

since no work is done. However, it is also reasonable to measure propellant performance by the amount of work that can be done (i.e., another possible measure of propellant performance is found in the SP calculation). Since the entropy is constant for this process, ΔS is 0; but, by definition, $dS = \delta Q$ (reversible)/T, so Q (reversible) is also 0; therefore, by the first law, ΔU is itself a measure of the reversible work (i.e., the maximum work).

4.3 Example of an Inaccessible Final State Along the Path of Maximum Work

The [SP] and {SP} states give the maximum work, but they do not give the best energetics in the previous cases. Why not just go back to ambient and realize a greater energy yield? The distinction is that it cannot be done along the path that gives the maximum amount of work (i.e., usable energy). The concept of CE or, more properly heat (internal energy in this case) of reaction, is based solely on the first law. The useful energy, the propellant's performance in the nature of the chamber gases doing work, also depends on the second law. For this JA2 sample system, the combined ambient conditions are inaccessible along the path of reversible work (figure 5). For example, to go from the final SP state to ambient, the temperature must be lowered at constant pressure. To do this heat must be removed from the system, and this heat is lost for doing work.

Actually, the utility of the thermodynamic state functions is that the details and the nature of the energetics can be easily determined if the states are known. For example, to go from the SP Static state (1 atm, 690 K) to ambient, the Q is just $\Delta H = -1{,}510 - (-1385) = -125$ cal/g. The corresponding ΔU for the same change of state is $\Delta U = -1533–(-1441) = -92$ cal/g. The difference $\Delta U - \Delta H = 33$ cal/g is Δ(PV), which, since P is constant, is just PΔV, that is, the work that must be done on the system as it is compressed at the constant pressure of 1 atm to its final volume. Then the system can go from Chamber to SP Static state with Q = 0,
$W = -3778$ J/g $= -903$ cal/g (done by the system on surroundings), then to the TP STATIC state by $Q = -125$ cal/g (removed from the system) and $W = 33$ cal/g (done on the system). There is actually 33 cal/g less work done by the system on the surroundings in going to the TP Static state than the higher temperature lower energy SP Static state. (This subtlety is lost in the CE because with T = 0 as the final state temperature, all the energy can be recovered as work.)

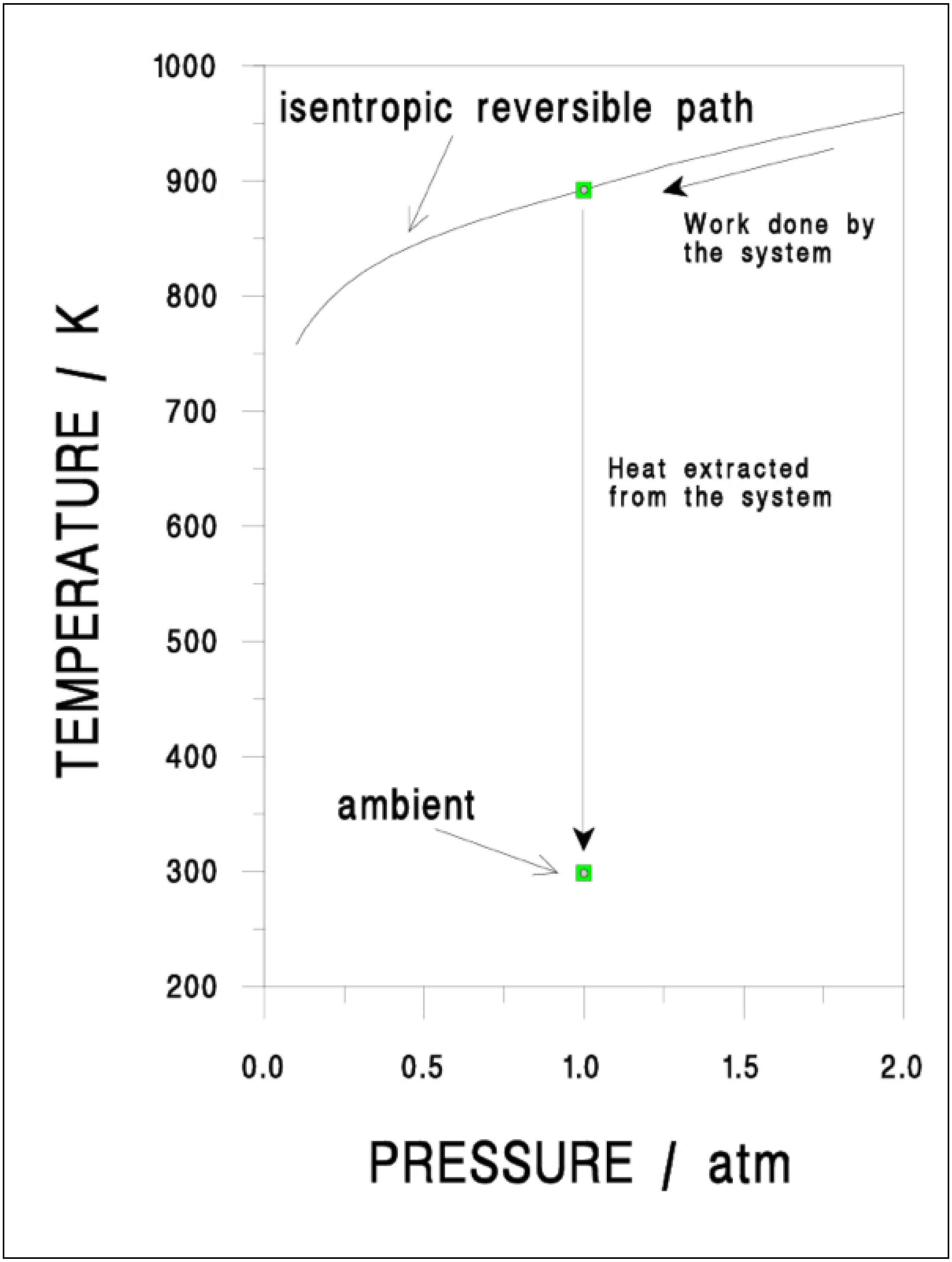

Figure 5. Illustration showing that the ambient state is inaccessible along the reversible adiabatic path.

Another check can be made for consistency: the overall energy difference (TP Static-Chamber) from table 1 is −1533 − (−538) = −995 cal/g as compared with a sum of the heat and work terms just discussed, which is 0 − 903 − 125 + 33 = −995 cal/g also. Another check is to do a different calculation of the "lost adiabatic work" of 33 cal/g. The work, using the volumes of the two states explicitly and the constant pressure of 1 atm, is $P\Delta V = 1(2282 - 984) =$ 1328 atm cm^3/g = 32 cal/g (presumably the same value except for code roundoff).

4.4 Mixed Dynamic-Static

It is possible to go from chamber to some (T, P) with dynamic compositions then (FREEZE) and go the rest of the way with static compositions, as shown in table 2. The energy in this case, 3883 J/g is, as expected, between the previous cases of either completely dynamic, where additional energy is produced from the equilibration and phase changes of the components, or completely static, where the working fluid does not react at all. This can be used, for example, to simulate the cessation of reactions at a known "freeze-out temperature."

Table 2. Example of mixed Dynamic-Static calculation.

Thermodynamic State	**P (atm)**	**V (cm^3/g)**	**T (K)**	**H (cal/g)**	**U (cal/g)**	**U (J/g)**	**−ΔU (J/g)**	**S (cal/K)**
Chamber	2844.63	5.00	3448	−193.22	−537.67	−2249.61	—	2.213
SP Dynamic	40.00	132.72	1599	−1077.69	−1206.25	−5046.95	2797.34	2.213
SP Static	1.00	2393.92	726	−1407.79	−1465.76	−6132.74	3883.13	2.213

4.5 Summary

In summary, a rigorous calculation of an alternative to the traditional CE can be readily performed and is basically equivalent to a properly defined heat (i.e., either internal energy or enthalpy, depending on conditions) of reaction. The reversible work done by the initial chamber gases in going to a well-defined final state, which reflects the range of operating conditions of the system, is also a plausible independent limiting (maximum) measure of propellant performance based on work during adiabatic expansion. This work is calculated as the difference between the internal energy (U) of the state having the same S as the chamber state and with some other state variable (e.g., either T, P, V, U, H) specified, and the state with U of the chamber (which also equals the U of the propellant on the thermochemical scale). The process may involve either dynamic or static compositions or an overall change of state involving both.

4.6 Critique of CE

Relative to the aforementioned analysis, the traditional CE is biased in several ways:

- It contains a contribution from energy associated with the temperature range from ambient to absolute zero of temperature.

- It is calculated using a γ evaluated at chamber condition (high temperature) and taken to be constant to the absolute zero of temperature.
- It implies a composition fixed at the chamber condition and invariant.
- It obscures the role of the final state in specifying a meaningful energy value that can be an independent indicator of propellant performance.

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5. Application to an ETC-Gun-Related Problem

Previous comments regarding the traditional calculation of the CE indicated several shortcomings in the quantity. In fact, it has been previously noted in ETC calculations (*6*) where energy (electric in this instance although the nature of the energy is not at issue) is added to the propellant that there appears to be an incomplete accounting for all the added energy. Table 3 shows the result of two BLAKE calculations of JA2. In one instance 1 kJ/g of energy is added to the propellant, while in the other there is no addition of energy. Two methods were used to calculate the CE:

- The standard method shows an energy difference of 922.10 J/g leaving 77.9 J/g unaccounted.
- The newer improved method (*17*, *18*) gives an energy difference of 802.3 J/g, leaving 197.79 J/g unaccounted.
- The improved method takes into account the variation of the thermodynamic properties with temperature and uses tabulated values of the enthalpy for the temperature range of 500–0 K.

Seemingly, the improved calculation exhibits a poorer accounting for the full 1 kJ/g that is added.

Table 3. Effect of adding energy.

Summary of Propellant Thermo Properties								
Rho/ (g/cm^3)	**Temp (K)**	**Press (MPa)**	**I (J/g)**	**Molecular Weight Gas**	**Covolume (cm^3/g)**	**Frozen Gamma**	**Standard CE (J/g)**	**New CE (J/g)**
... add 0 kJ/g to propellant			1154.70	24.82	0.99	1.22	5134.50	4408.00
0.20	3446	288.20						
... add 1 kJ/g to propellant			1342.50	24.61	1.00	1.22	6056.60	5210.30
0.20	3973	335.80	Δ ENERGY =				922.10	802.30
					Missing		+77.90	+197.70

To resolve the discrepancy, BLAKE calculations were performed for specific states yielding full thermodynamic information. This is shown in figure 6 using the code's thermochemical energy scale. The same two calculations given previously are indicated, with two different resulting equilibrium compositions being denoted as C0, for 0 kJ/g added energy and C1, for 1 kJ/g added energy; in both cases, the change of state is from propellant at ambient to chamber at constant energy and volume. The internal energy values indicated, U = −1250 J/g and U = −2250 J/g for the C1 and C0 compositions, respectively, which are reported by the code as output values, clearly account exactly for the 1 kJ/g added energy. An additional state is calculated for static composition C1 at V = 5 cm^2/g and T = 3446 K, that is, for the unchanged system volume and the equilibrium temperature of the C0 composition. This allows the formulation of the isothermal constant volume change of state C0 → C1, for which the ΔU = −2062 − (−2250) = 188 J/g is the energy of reaction; the positive value of this quantity means that an additional 188 J/g is tied up in the C1 composition. Thus, it would be expected that approximately 188 J/g would not show up on an energy scale that was basically thermal and does not take into account the possibility of chemical reactions.

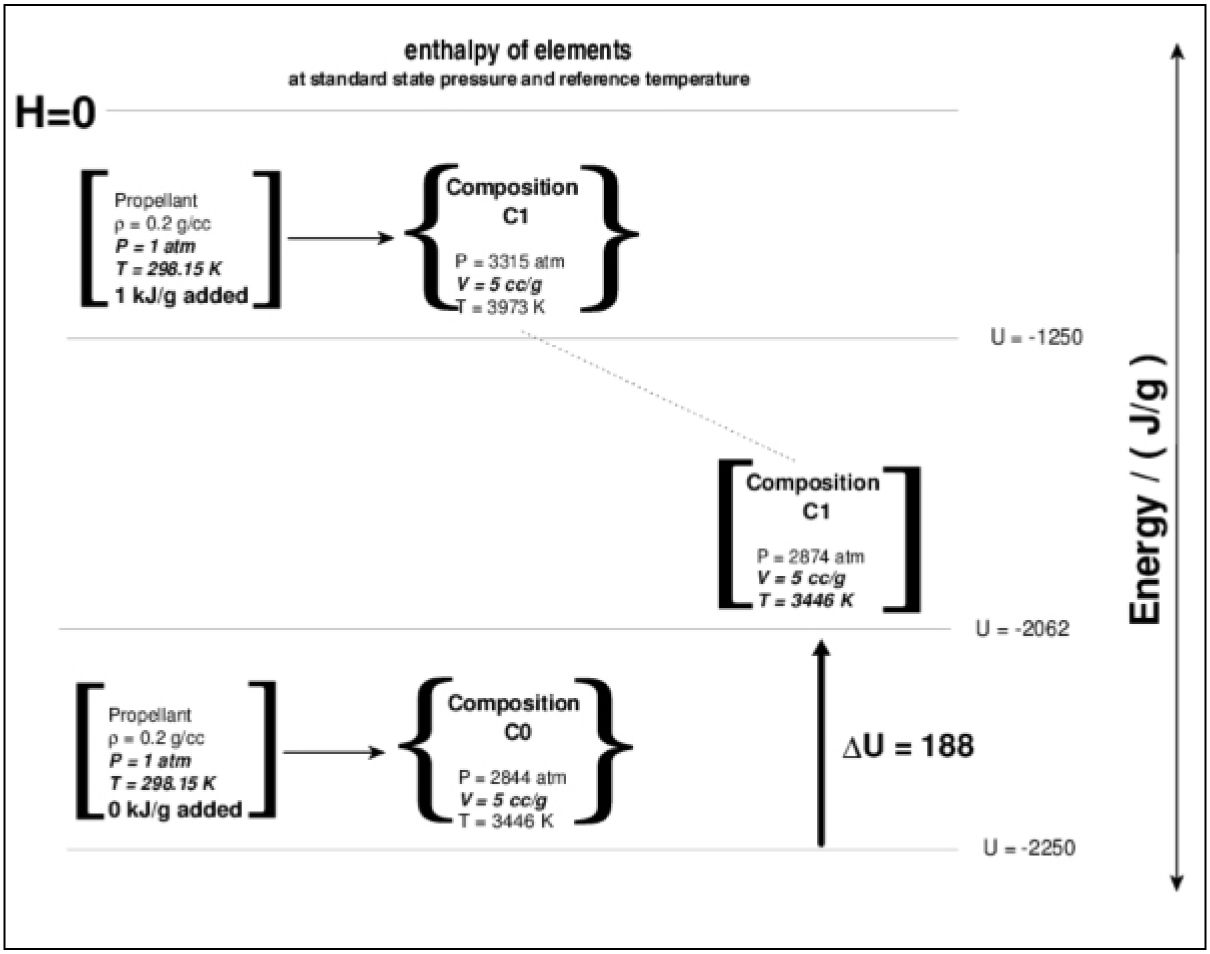

Figure 6. Calculation of an energy of reaction.

The energy of reaction is only an approximate measure of the CE if this is taken to mean the total energy associated with the conversion of one composition to another, which is not necessarily at the same temperature and pressure. The internal energy of a system on the thermochemical energy scale can be written as[*]

$$U(T,P) = U(T,P)$$

$$dU(T,P) = \left.\frac{\partial U}{\partial T}\right)_P dT + \left.\frac{\partial U}{\partial T}\right)_P dP$$

$$U(T,P) = U^o(298.15K) + \int_{298.15K}^{T'} \left.\frac{\partial U}{\partial T}\right)_P dT + \int_{P^o}^{P'} \left.\frac{\partial U}{\partial T}\right)_P dP$$

or

Internal Energy = standard state formation energy + energy terms from T & P changes,

which includes an energy of formation term and terms involving changes in temperature and in this example, pressure. Writing U′ for the last two integrals gives:

$$U(T, P) = U^o(T) + U'(T, P)$$

To isolate the chemical portion of the total energy, the energy term representing the T and P changes and

$$U'(T, P) = U(T, P) - U^o(T)$$

is identified.

Then, for the change of state previously considered, a temperature and pressure, T_Θ and P_Θ, are determined for the C1 composition

$$U(\{C0\}, V, T_{C0,}\ P_{C0}) \rightarrow U([C1], V, T_\Theta, P_\Theta) \qquad \Delta U_{chem.}$$

such that the energy difference, ΔU_{chem}, is the measure of the energy generated by the composition change only, that is, where the temperature and pressure terms contributing to the energy are equal:

$$U'(\{C0\}, T_{C0}, P_{C0}, V) = U'([C1], T_\Theta, P_\Theta, V)$$

This then simply gives

$$\Delta U_{chem} = U([C1], 298.15K, P^0) - U([C0], 298.15K, P^0)$$

Note that the volume is constant, and that it is the volume of the system and also the volume occupied by the combustion products of the propellant. Using values for the problem under consideration (see figure 6 for reference; the ambient temperature states, U[{C1}, 298.15K, P^0]

---

[*] Although the natural variables of *U* are *T* and *V*, one can still formally consider *U* as a function of *T* and *P* as done here.

and U[{C0}, 298.15K, P^0] correspond to this figure but are not shown) substituted into the previous equation gives:

$$\Delta U_{chem} = -6221 - (-6417) = +196 \text{ J/g}$$

Thus, the measure of the CE energy for a composition change is simply the difference in the standard-state formation energy of the products and reactants for the specified change of state at the reference temperature; that is, removing the temperature and pressure effects leaves only the difference between the energy of formation of the set of products and the set of reactants for the specified change of state as the measure of the CE. Referring to table 3, the calculation of the CE by the new method correctly reflects, to within 1.7 J/g, the value of the missing energy; the energy that is tied up in the composition change that the IB energy scale is unable to directly measure. This also indicates that the seemingly better value of the CE calculated by the traditional approach is actually in greater error.

Table 4 shows another way of illustrating the conclusion that the chemistry (i.e., the energy of the composition change) is not being reflected in either method of calculating the CE, but that the new method is at least correctly accounting for the thermal energy, while the traditional value of the CE is not. Here, the values of the previous calculation are repeated and it is also emphasized that the energy is added to the propellant and the resulting compositions are dynamic. The calculation is now repeated, but the 1-kJ/g energy is not added to the propellant but to the chamber composition. The system now proceeds to equilibrium again, with two different states specified. In the dynamic compositions case the system reaches the same state as before. It makes no difference whether the energy is added to the propellant or to the isoenergetic chamber composition, the same dynamic equilibrium state results, and the same values are reported by BLAKE.

In the next case the energy is also added to the chamber composition, but the chemistry is defeated. This means the composition is constrained to remain (static) at the composition of the chamber before the addition of the energy. If the energy difference between the 0-kJ/g chamber composition and the 1 kJ/g added to the 0-kJ/g static chamber composition is computed, the new value of the CE overestimates the added energy by only 1.8 J/g, while the standard method overestimates by 147 J/g. This also shows, in the cases of the new value of the CE, that it is able to account for the added energy if a composition change (the chemistry) is not permitted.

The relationship of the different values of the system energy at constant volume is shown in figure 7, where the energy is plotted vs. temperature as the difference from the energy at the adiabatic chamber state as a point of reference. The two curves labeled static and dynamic are the BLAKE calculations based on the thermochemical scale. The curve, allowing dynamic compositions relative to the curve of static compositions, shows the degree and regions where composition changes affect the energy. These curves stop at ambient (298.15 K) in keeping with the previously noted limit on this type of calculation. Also shown are the two points representing the CE values done by the standard and new method, where it is seen that the static (i.e., [VT] calculation, which is a CE-type quantity) is in line with the new method of calculating the CE. The remaining curve is the standard CE value scaled for temperature and shows the degree to which it differs for the (compatible) new CE and static values. Although, there are deviations at lower temperatures and in the two CE values themselves, the agreement in the neighborhood of the chamber condition is quite good over a fairly broad range of temperature. This suggests that energetics calculated for a system using the (biased) standard CE involving small temperature excursions from the chamber state are likely to adequately reflect the system's energy change. Figure 8 shows a similar comparison between the reversible work (isentropic expansion) for the fully variable thermochemistry BLAKE calculation and the corresponding results using the lumped invariant thermodynamic quantities and composition and the Noble-Abel EOS. Here too, and perhaps even more so, there is surprisingly good agreement over a fairly broad range of temperatures.

Table 4. Comparison of adding energy to propellant or to chamber composition.

Summary of Propellant Thermo Properties								
Rho (g/cm^3)	**Temp (K)**	**Press (MPa)**	**I (J/g)**	**Molecular Weight Gas**	**Covolume (cm^3/g)**	**Frozen (Gamma)**	**Standard CE (J/g)**	**New CE (J/g)**
... add 0 kJ/g to propellant								
0.20	3446	288.20	1154.70	24.82	0.99	1.22	5134.50	4408.00
... add 1 kJ/g to propellant								
0.20	3973	335.80	1342.50	24.61	1.00	1.22	6056.60	5210.30
						Δ ENERGY =	922.10	802.30
						Missing	+77.90	+197.70
... recompute 0-kJ/g to chamber								
0.20	3446	288.20	1154.70	24.82	0.99	1.22	5134.50	4408.00
... add 1 kJ/g to 0-kJ/g chamber - Dynamic								
0.20	3973	335.90	1342.50	24.61	1.00	1.22	6056.70	5210.60
... add 1 kJ/g to 0-kJ/g chamber - Static								
0.20	4093	342.40	1371.30	24.82	1.00	1.22	6281.50	5409.80
						Δ ENERGY =	1147.00	1001.80
						Missing[a]	–147.00	–1.80

[a] That is, unaccounted for on the basis of 1 kJ/g added energy.

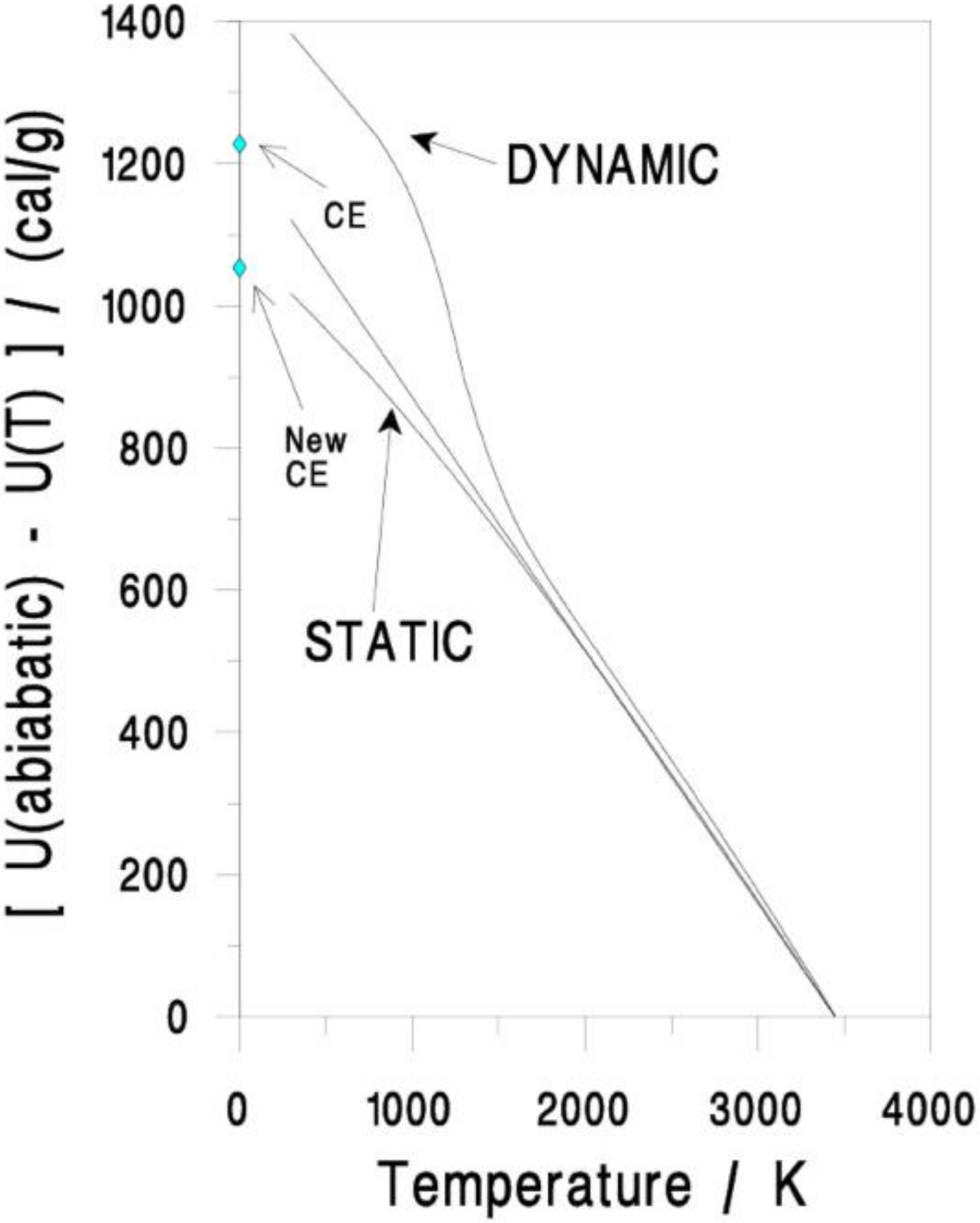

Figure 7. Comparison of different energetics.

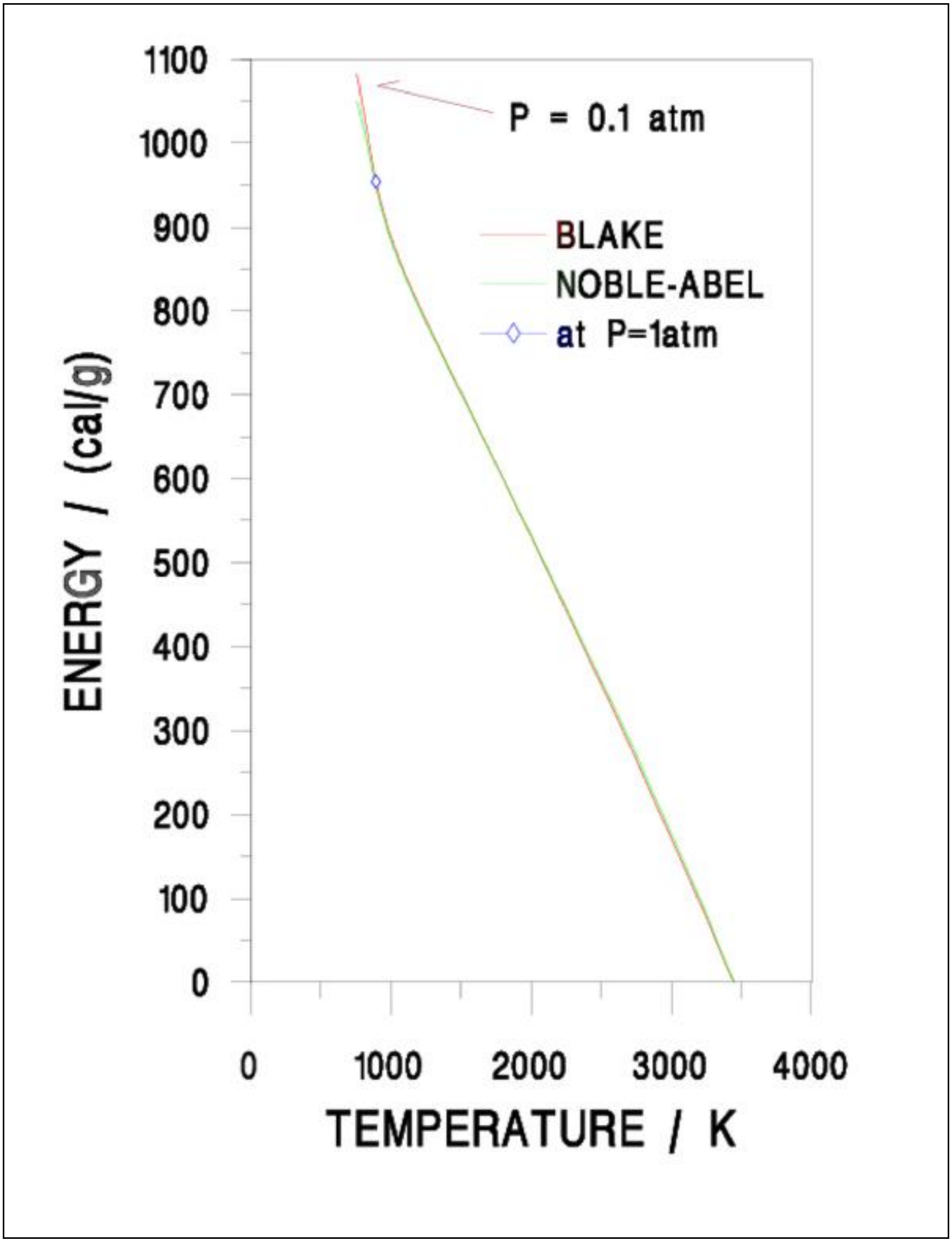

Figure 8. Comparison of reversible work.

6. Calculation of an Idealized Gun Using Variable Thermochemistry

One of the objectives of this work was to perform an assessment of propellant performance independent of the gun system, while fully realizing that the stated goal is an oxymoron. It turns out that it is quite common (IBHVG2 [9] and CONPRESS [*10*] are examples) to use as an IB model a constant breech pressure before propellant burnout, Lagrange pressure gradient, no-energy-loss gun. This gun's IB cycle can be calculated in the usual way of using BLAKE to estimate lumped thermodynamic variables, which are then used in conjunction with the traditional covolume EOS to get projectile velocity or kinetic energy (KE). The usual questions arise: Are the lumped thermodynamic values applicable over the range of the cycle? What effect would variable (equilibrium) chemistry play in the energetics? For this gun system it is possible to perform the IB calculation using BLAKE only (without alteration), thus including the full equilibrium thermodynamics and, in effect, performing an exact calculation of the model.

The calculation is straightforward once the states of the system have been identified. There are several steps that need to be considered to understand the process, but only three states of the system need to be calculated. The characteristic features of this system are a constant pressure portion that lasts from the time the operating pressure is reached (shot start) to the time the solid propellant is entirely consumed (burnout). This is followed by the reversible isentropic expansion of the gases until the end of the tube is reached (muzzle condition). The coordinates of the gun are shown in figure 9.

6.1 The Use of BLAKE

BLAKE is used to perform the calculations, but the method relies on basic thermodynamic principles and is not tied in any way to BLAKE itself. Results are not intended to reflect upon the accuracy of the code. BLAKE is chosen because it allows for real gas effects and has the apparatus in place to calculate the designated states.

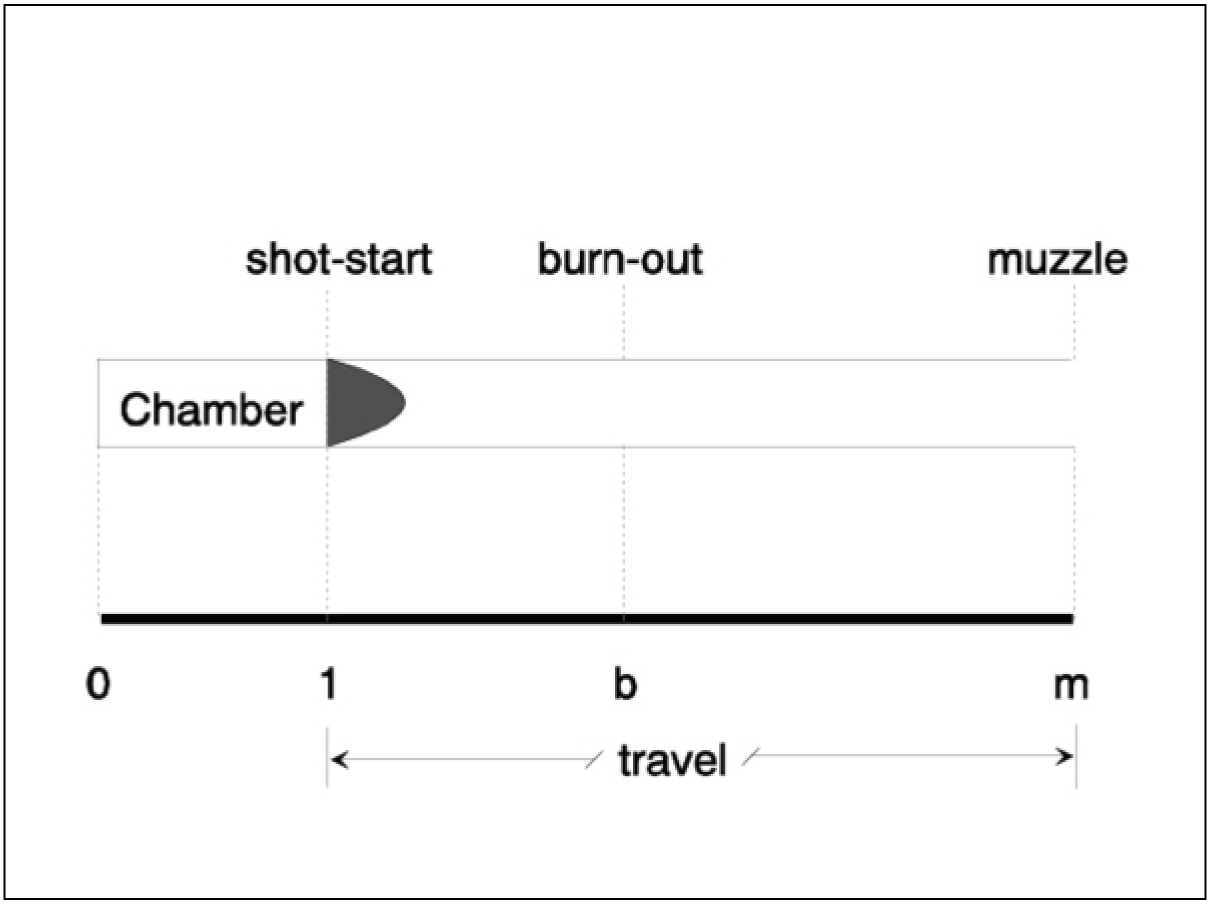

Figure 9. Coordinates for the idealized gun.

6.2 Description of the Problem

The process is adiabatic, so for each step Q = 0. The breech pressure, also called the chamber pressure, is given and is determined by known operating parameters (design parameters) for the gun. A Lagrange gradient is assumed. The method of Lagrange is often used and is dealt with elsewhere (*1*). The use of the Lagrange gradient allows a partitioning to be made of the total energy, in the form of work, done by the system. The system here is the propellant, which hereafter is used to refer to the total matter of any phase or composition that comprised the original unburnt propellant. The system, however, does not include any bulk KE of the propellant. Consequently, the internal energy of the system, U, means the thermal energy and CE of the system but not any bulk KE, $KE_{Propellant}$. Strictly speaking, the total energy E of the system is

$$E_{system} = U + KE_{Propellant}.$$

Note that in keeping with the Lagrange assumptions, any condensed phase of propellant is considered to be uniformly dispersed throughout the system volume.

BLAKE only calculates the internal energy U. The relationships that follow from the choice of the Lagrange gradient assumption are used to partition the change in internal energy, ΔU, between the KE of the combustion gases (i.e., the [propellant] system) and the projectile:

$$\Delta U = KE_{Propellant} + KE_{projectile}$$

Thus, the change in internal energy of the system results in work on the surroundings, which manifests itself as KE (velocity) of the propellant and the projectile.

6.3 How To Do the Calculation

The calculation can be done for either dynamic compositions or static compositions. The traditional approach is static,, meaning the working fluid is inert. It is actually easier both conceptually and computationally to do the dynamic calculation. Since the objective is to explore the role of the chemistry, the calculation is only done here for dynamic compositions. For dynamic compositions only three states need be calculated:

1. The initial state of the propellant.
2. The state at burnout.
3. The state at muzzle.

Each of these states is now considered.

6.3.1 State 1: Initial

For the initial state, the internal energy of the propellant must be calculated. The initial state of a propellant is usually at ambient conditions taken here to be P = 1 atm and T = 298.15 K. These are also the conditions for which the standard enthalpy of formation is tabulated. This is also the reference T and standard pressure for BLAKE's assigned energy scale; consequently, for condensed-phase propellants $U \approx H = (\Delta H_f^o)_{298.15K}$ (see Kotlar [*13*]). The internal energy of the initial state, U_0, can therefore be calculated from the tabulated $(\Delta H_f^o)_{298.15K}$, the proportions of the components, the chemical formulas, and the atomic weights. Although it is not necessary to run BLAKE to get this number, it can be conveniently done by merely specifying the COMposition and running.

6.3.2 State 2: Burnout

The energy of the state at burnout, U_b, is calculated from the internal energy of the propellant and the work done along the constant pressure path that is specified as a design parameter of the gun. Identification of this state is the key to performing the calculation. It is straightforward to write

$$U_b = U_0 - W$$

where, for the total work,

$$m_c W = P(m_c V_{0b} - V_C)$$

and, for the mass specific work,

$$W=P(V_{0b} - V_{0C})$$

where $V_{0C} = V_C/m_c$, V_C is the volume of the chamber, and m_c is the mass of the charge (the propellant).

Now, using the relationship

$$U_b = H_b - P V_{0b}$$

$$U_0 - PV_{0b} + PV_{0c} = H_b - PV_{0b}$$

$$H_b = U_0 + PV_{0c}$$

or

$$H_b = U_0 + P V_c/m_c$$

Thus, the thermodynamic state at burnout is fully specified by the operating pressure and the enthalpy at burnout as indicated, where the initial internal energy U_0, the operating pressure P, the chamber volume V_c, and the charge mass m_c are all known design parameters.

6.3.3 State 3: Muzzle

From the state at burnout, a reversible adiabatic expansion takes place to muzzle. For a reversible expansion

$$dS = \delta Q_{rev}/T$$

Since the process is adiabatic,

$$\delta Q_{rev}= 0$$

and the expansion proceeds along a constant entropy path. This is easily calculated by BLAKE using ISOline or POInt, by taking the value of the entropy at burnout, holding it constant, and going from the specific volume at burnout to the specific volume at muzzle, which can be calculated from the design parameters (i.e., at muzzle)

$$V_m = V_{Total}/m_c$$

where V_{Total}, the total volume, is the sum of the chamber and tube volume

$$V_{Total} = V_{chamber} + V_{tube}$$

6.4 A Sample Calculation

The design parameters (*7*) of the idealized gun are summarized in table 5, and the composition of the JA2 propellant used is given in table 6. A sample calculation follows for JA2, with each "run" corresponding to one of the three states needed; each calculation is illustrated using BLAKE commands.

6.4.1 RUN 1

```
FOR,XNC1306,-1.6541E8,C,6000,H,7395,O,10209,N,2605
FOR,NGX,-8.8600E4,C,3,H,5,O,9,N,3
FOR,DEGDNX,-99400,C,4,H,8,N,2,O,7
FOR,AKAR2X,-25500.,C,14,H,14,N,2,O,1
FOR,CX,0.,C,1
FOR,ALCX,-6.6420E4,C,2,H,6,O,1
COM,XNC1306,59.5,NGX,14.9,DEGDNX,24.8,AKAR2X,.7,CX,.05,ALCX,.05
STOP
```

Table 5. Design parameters of the idealized gun.

Projectile Mass	m_p = 11.4 kg
Charge Mass	m_c = 10.1 kg
Tube Diameter	d = 120 m
Travel	ℓ = 4.75 m
Chamber Volume	V_C = 9.75 liter = 9750 cm^3
Breech Pressure	P_{max} = 574 MPa
Calculated:	
Space Mean Pressure	P = 515 MPa = 5083 atm.
Specific Volume at Muzzle	V_m = 6.2843 cm^3/g

Table 6. JA2 propellant composition.

Name	**Weight (%)**	**Mole (%)**	**$(\Delta H_f^o)_{298.15K}$ (cal/mol)**	**Formula**
XNC130	59.500	0.106	−165,410	$C_6 H_{7.35} O_{10.209} N_{2.605}$
NGX	14.900	32.705	−8.8600	$C_3 H_5 O_9 N_3$
DEGDNX	24.800	63.031	−99,400	$C_4 H_8 N_2 O_7$
AKAR2X	0.700	1.542	−25,500	$C_{14} H_{14} N_2 O_1$
CS	0.050	2.075	0	C_1
ALCX	0.050	0.541	−66,420	$C_2 H_6 O_1$

BLAKE reports:

```
THE HEAT OF FORMATION IS −537.67 CAL/GRAM = −2.6799E+05 CAL/MOLE.
```

Therefore,

$$U_0 \approx H_0 = -537.67 \text{ cal/g}$$

the internal energy of the initial state.

6.4.2 RUNs 2 and 3

Using

$$H_b = U_0 + P\ V_c/m_c,$$

calculate the value of the enthalpy at burnout,

$$H_b = -418.787 \text{ cal/g},$$

as the target for the next BLAKE run. Once this burnout state is found by BLAKE (POInt calculation next), the reversible expansion step can be entered immediately, starting with the volume and entropy at burnout, holding the entropy constant, and going to the volume at muzzle (the ISOline calculation next). Therefore, the following are statements of the CONPRESS gun in the command language of BLAKE:

```
POI,P,5085.17,H,−418.787
ISO, S,  ,V,  ,1, 6.2843
```

BLAKE gives as output:

P (atm)	V (cm^3/g)	T (K)	H (cal/g)	E (cal/g)	S (cal/K/g)	RHO (g/cm^3)	CV (cal/K)
(1) .508377E+04	2.7859	2884.	−418.84	−761.83	2.088	0.359	0.360
(2) .145015E+04	6.2843	2333.	−735.60	−956.29	2.088	0.159	0.348

BLAKE reports the state at burnout, which is not needed for the overall energy (line 1), and the energy of the state at muzzle (line 2)

$$U_m = -956.29 \text{ cal/g},$$

from which the KE and velocity of the projectile can be calculated since the internal energy of the initial state, U_0, is also known:

$$\Delta U = U_m - U_0 = -418.62 \text{ cal/g} = -1751.51 \text{ J/g}$$

where

$$-\Delta U = -\text{Work} = KE_{\text{projectile}} + KE_{\text{Propellant}}$$

The overall process and the thermodynamic states reported by BLAKE are summarized in figure 10. The Lagrange formulas are now used to partition the energy given up by the system, ΔU, between the KE of the propellant and KE of the projectile giving

$$KE_{\text{projectile}} = 13.66 \text{ MJ}$$

and

$$\text{velocity}_{\text{projectile}} = 1548 \text{ m/s}$$

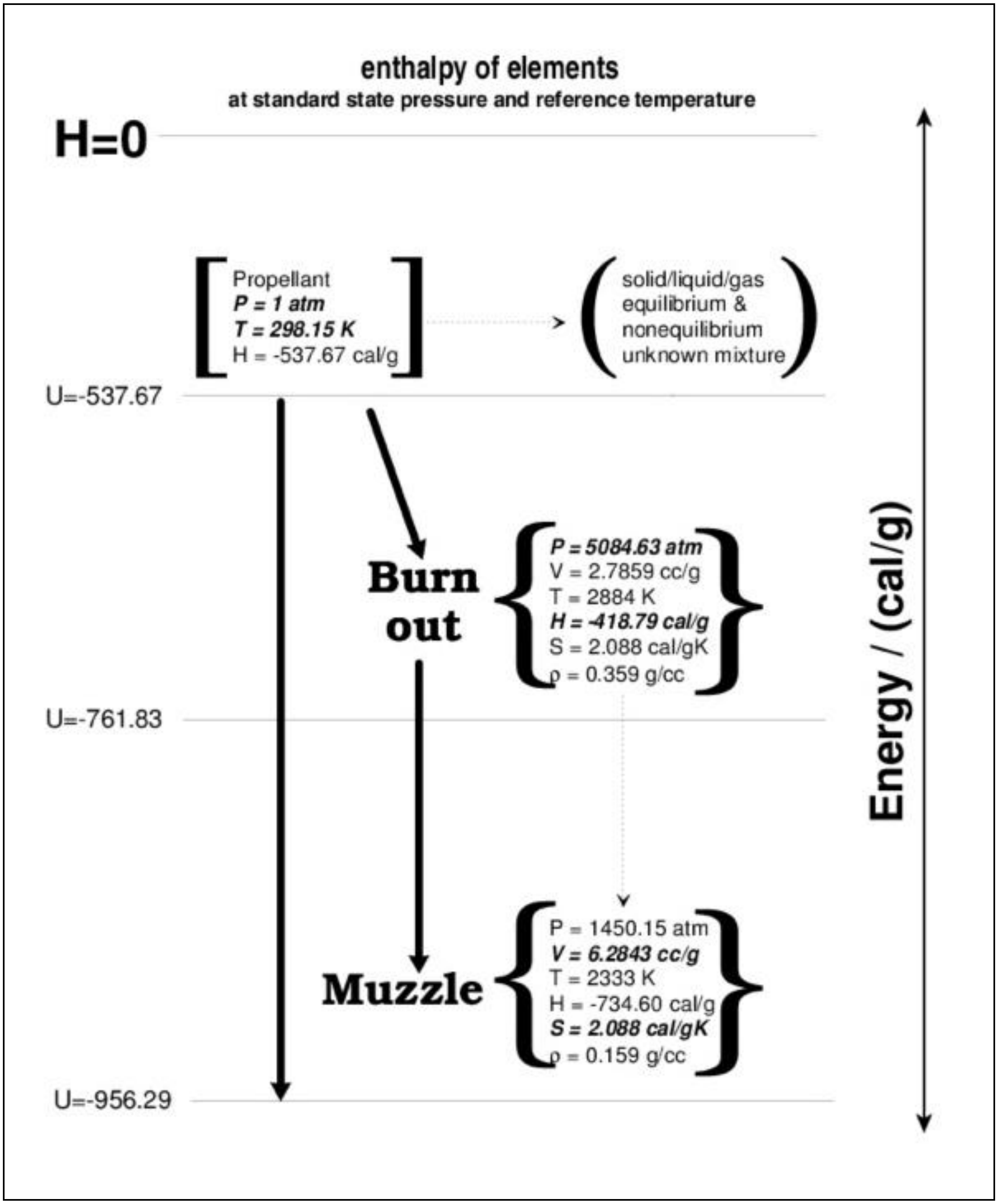

Figure 10. States involved in the idealized gun. (Specified state variables appear in bold print.)

For comparison, the same gun system using CONPRESS and thermodynamics estimated at l d = 0.20 g/cm^3 (using BLAKE’s GUN command) gives projectile KE of 14.38 MJ and projectile velocity of 1588 m/s. Thus, for this example, CONPRESS results are 5.3% and 2.6% higher for the total projectile KE and velocity, respectively, than for the full thermochemistry BLAKE code level results.

7. Conclusions

- The traditional method of calculating the CE (a misnomer) gives a biased estimate of propellant energetics.
- A properly designed general thermodynamic code allows detailed interrogation of propellant energetics (calculates a process).
- The nature of the IB versus the thermodynamic energy scale shows the deficiency of the traditional IB scale when treating reacting systems.
- The energy anomaly encountered in the ETC problem is explained as the (chemical) energy of a composition change.
- A general thermodynamic equilibrium code (e.g., BLAKE) can be used to perform an “exact” calculation of the energetics of an idealized, but often used, model gun, the constant breech pressure gun.

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List of Symbols, Abbreviations, and Acronyms

b or η	Noble-Abel equation of state covolume
CBP	constant breech pressure
CE	chemical energy
C_P	heat capacity at constant pressure
C_V	heat capacity at constant volume
EOS	equation of state
ETC	electrothermochemical
H	enthalpy
I	impetus
IB	interior ballistic
KE	kinetic energy
ld	loading
mc	charge mass
mp	projectile mass
n	number of moles
P	pressure
T	temperature
T_{ref}	= 0; reference temperature
U	internal energy
V	volume
W_{gas}	weight (mass) of gaseous combustion products
W_{Total}	weight (mass) of gaseous and condensed phase combustion products
ΔU	change in U
ΔH	change in H

ΔH^0_f	standard state enthalpy (heat) of formation
{}	dynamic composition
[ ]	static composition
γ	ratio of heat capacities

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