



## **The Role of Nitrogen in Gun Tube Wear and Erosion**

**by Paul J. Conroy, Charles S. Leveritt,  
James K. Hirvonen, and John D. Demaree**

**ARL-TR-3795**

**May 2006**

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Weapons and Materials Research Directorate, ARL**

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<b>14. ABSTRACT</b> The U.S. Army Research Laboratory has recently made discoveries in the area of interior ballistic propellant combustion product interactions with a gun-tube bore. These discoveries were based on two hypotheses. The first was that the products could and are “dynamically” nitriding the bore of the gun, thus creating a nitride coating which inhibits the dissociation of CO and subsequently reduces the amount of carbon uptake. The second was that the combustion products richer in nitrogen have a lower temperature upon expansion in our experimental fixture as well as in a gun barrel. This is due to Joule and/or Joule-Thomson cooling effects. Both of these hypotheses were proven and validated through numerical and experimental methods. Higher nitrogen containing combustion products have definitively been shown to have relatively lower erosivity with respect to those having lower nitrogen content. These results have revolutionized the gun propellant development community in that for the first time, there is guidance for erosivity control through propellant chemical constituent formulation.				
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## 1. Introduction

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Gun tube wear and erosion has been a limiting factor in gun performance since their invention. Frederick Abel described gun tube erosion in the nineteenth century when he developed cordite. Since that time, erosion has had a cyclic history between performance requirements, resulting erosion issues, and subsequent palliatives. Historically, palliatives have been additives to the propelling charge to create a protective surface coating, a cool boundary layer, or a heat flux impediment. To date, this is the preferred method of mitigating erosion given an existing weapon-charge system.

For over 10 years, the U.S. Army Research Laboratory (ARL) has been performing basic research in the area of gun tube wear and erosion, after a hiatus of about 10 years from the early 1980s through the early 1990s. Propellant chemistry and the interaction of the combustion products with the gun-tube wall has been the primary driver of the research. Since WWII, it has been noted (Smith, 1954) that propellants containing nitramine are inherently more erosive than conventional propellants such as M30 (a triple-based propellant having a similar adiabatic flame temperature). This claim was later verified by Ward et al. (1981, 1982). The reason for the high erosive behavior of this propellant was unknown. Surface chemical analysis of gun surfaces by Benet Laboratory (Kamdar and Venables, 1984) and ARL modeling (Conroy et al., 2001) assisted in developing an explanation for this behavior.

The primary finding of this previous work was that the carbon monoxide in the combustion product gas dissociated on the surface with a very small energy barrier. Subsequently, the free hydrogen scavenged the oxygen from the surface into water and the carbon diffused into the steel surface, resulting in a surface carburization (Conroy et al., 2001). This transforms the steel into a cast iron, which reduces the surface melt temperature from 1723 K down to 1423 K. RDX-based propellants have significantly higher amounts of carbon monoxide than the triple-base propellant M30, which supports the hypothesis of CO dissociation and subsequent carburization.

The current report describes work involving advanced propellant formulations having relatively high concentrations of nitrogen in the product species.

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## 2. Discovery of Dynamic Nitriding

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Propellant erosivity characterizations were conducted in a blowout chamber fabricated from the breech and chamber of a 37-mm gun shortened and threaded to accept a retaining end cap for experimental samples. A photograph is shown in figure 1 and sectional drawing is shown in figure 2. This facility in various forms has been in use since WWII (Weigand, 1945).

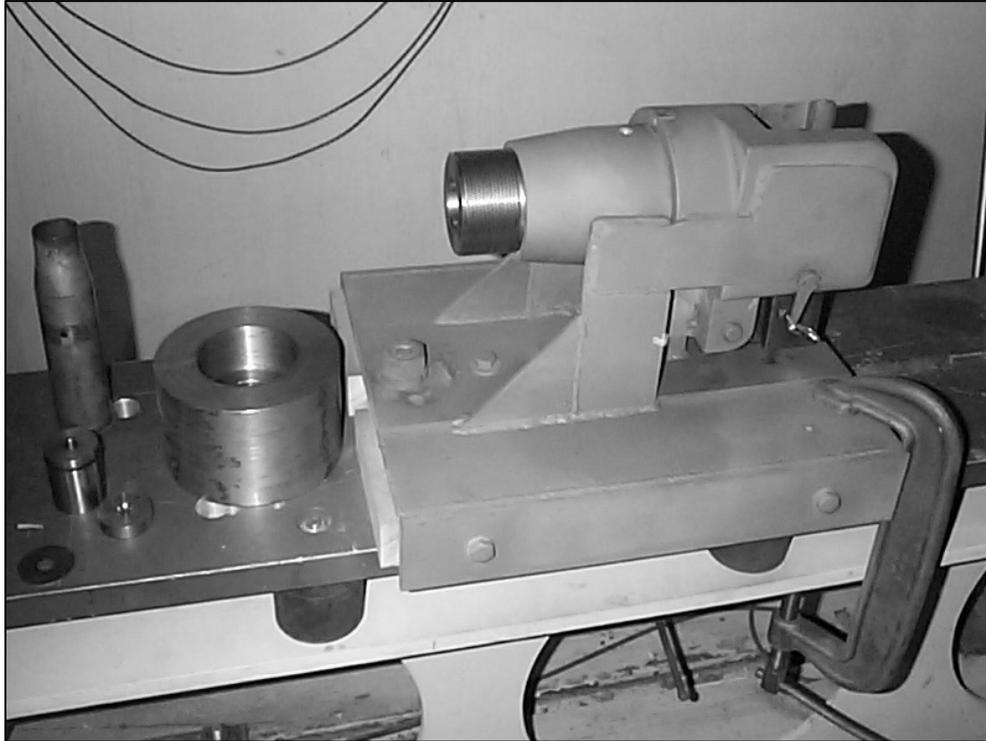


Figure 1. Photograph of an ARL 37-mm erosion fixture.

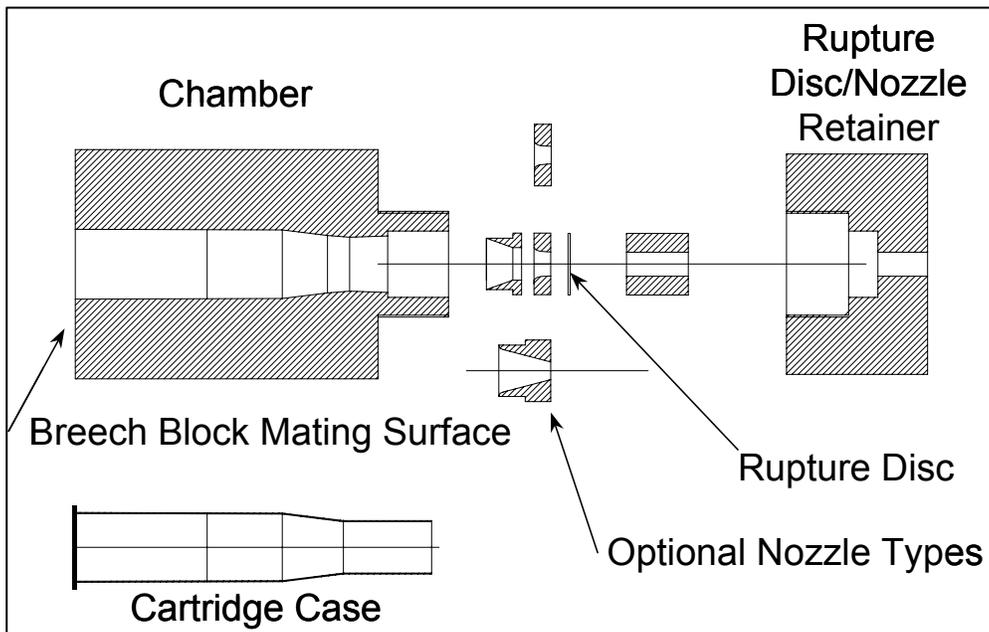


Figure 2. Drawing of an ARL 37-mm erosion fixture.

Figure 3 shows a detail of an ARL erosion nozzle made typically of ASTM A723M-02 alloy (2006). The entrance to the nozzle is opposite to the throat. The nozzle profile was developed by continuously firing through a sample with a straight hole until the mass loss became consistent from one firing to the next.

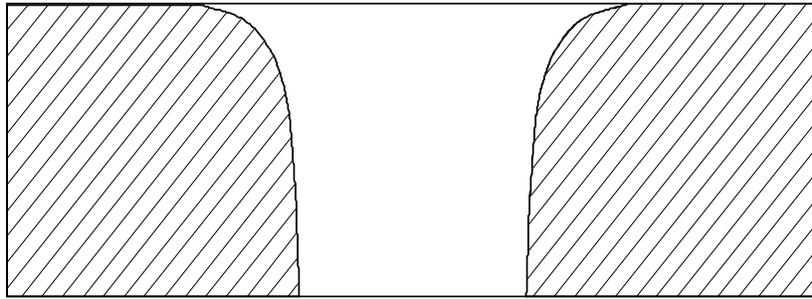


Figure 3. ARL erosion fixture nozzle.

Selected propellants that have recently been evaluated for erosivity are listed in table 1. As the propellants were evaluated, all results supported the proposed carburization mechanism for erosion, until the testing of the Thiokol Gun Development propellant TGD-002. While TGD-002 has an adiabatic flame temperature similar to that of M8, the CO/CO<sub>2</sub> ratio is about ten times higher than that of M8. Considering only the carburization mechanism, one would predict that TGD-002 erosion would be much higher than that of M8. What was found was that the TGD-002 erosion was actually lower than that of M8, as shown in table 1. This effect was also noted in Indian Head propellant IHGP-300, a 70% RDX system with a nitrocellulose and diazidonitrazapentane (DANPE) binder. As shown in table 1, the mass loss from IHGP-300 was less than half of that of the M8, despite having a CO/CO<sub>2</sub> ratio eight times that of the M8.

Table 1. Selected propellant properties and mass loss.

Propellant	Chamber			Nozzle			Wear (mg)
	Temp	F (J/g)	Temp	CO/CO <sub>2</sub>	N <sub>2</sub> /CO	N <sub>2</sub>	
JA2	3450	1152	2569	2.53	0.35	0.126	90
PAP-8165	3419	1284	2444	8.29	0.78	0.271	62
TGD-9/2/9	3410	1291	2375	19.55	0.77	0.300	80
009+002	3410	1291	2375	19.55	0.77	0.300	104
TGD-019	3268	1296	2298	15.59	0.86	0.294	40
TGD-036	3191	1280	2238	18.26	0.84	0.292	57
TGD-035	3181	1273	2226	24.40	0.81	0.295	66
M43	3100	1186	2201	7.78	0.66	0.237	27
M30	3035	1080	2190	3.10	1.08	0.280	21
TGD-021	2795	1164	1995	22.27	0.91	0.305	5
TGD-009	2577	1072	1898	20.26	0.67	0.257	21
M8 (Ward et al., 1982)	3768	1168	2922	1.44	0.49	0.146	241
JA-2/ RPD351	3743	1172	2892	1.50	0.48	0.143	251
TGD-002	3718	1354	2643	14.88	0.84	0.324	193
IHGP-300	3725	1333	2730	4.13	0.89	0.284	114

It was hypothesized that high nitrogen propellants might “dynamically” nitride the inside bore surface of the erosion nozzle during firing. This nitriding could occur over milliseconds, whereas typical gaseous nitriding can take hours or days. We investigated this possibility by measuring the nitrogen content on the surface of test nozzles after firing, using resonant nuclear reaction analysis at the ARL ion accelerator facility shown in figure 4. An energetic beam of protons was directed onto the surface, inducing the  $^{15}\text{N}(p,\gamma)$  reaction, and the characteristic gamma rays were detected using a scintillation detector near the analysis chamber (figure 5). Since this reaction occurs only at a particular ion energy, the beam energy was increased to query the nitrogen concentration at increasing depths into the sample; the gamma yield could then be calibrated to give a depth profile of nitrogen in the gun steel. Figure 6 shows atomic nitrogen content vs. energy (depth) at various locations on the inner surface of an ARL erosion nozzle, which had high nitrogen propellant products exhausted through it. The resultant concentrations are as high as 9 atomic percent. This places the nitriding at  $\text{Fe}_4\text{N}$  levels.

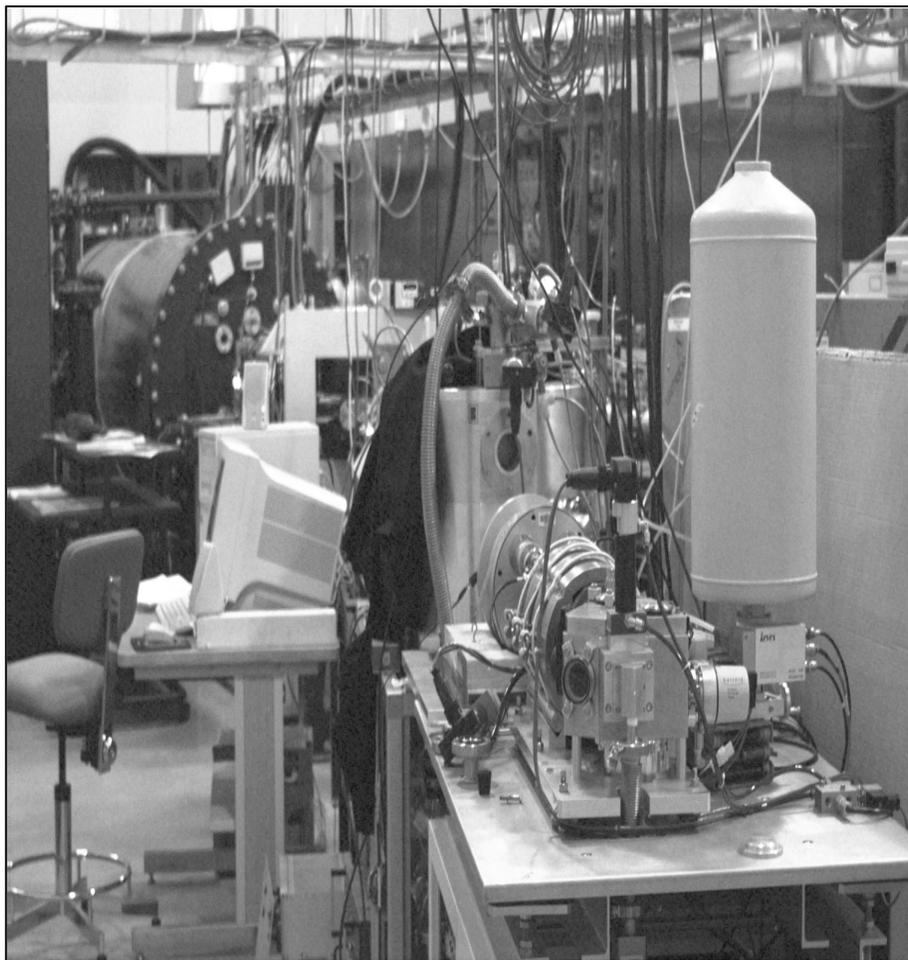


Figure 4. ARL ion accelerator facility.

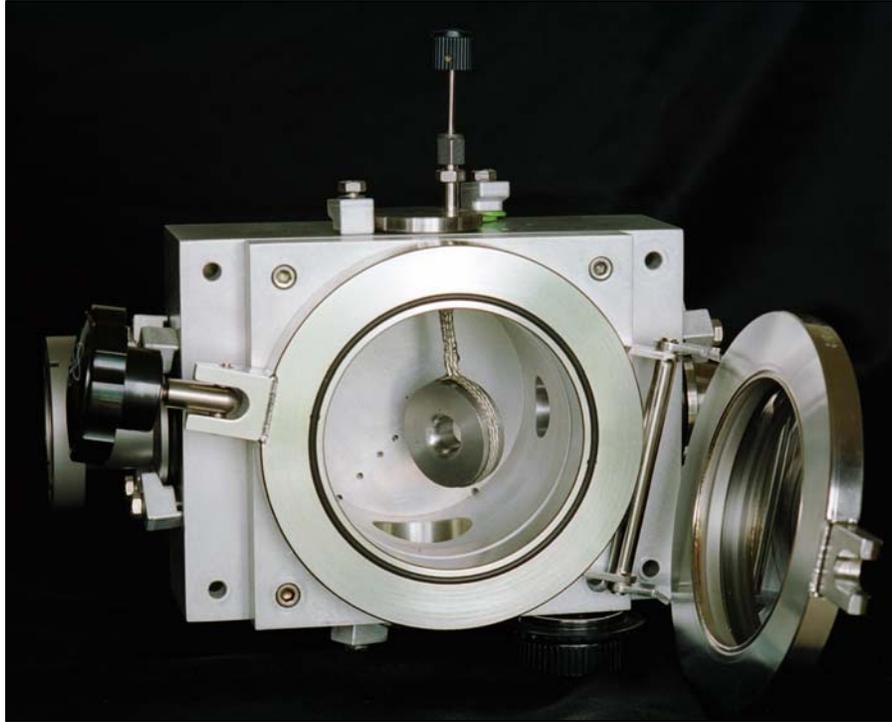


Figure 5. Nuclear reaction analysis (NRA) chamber.

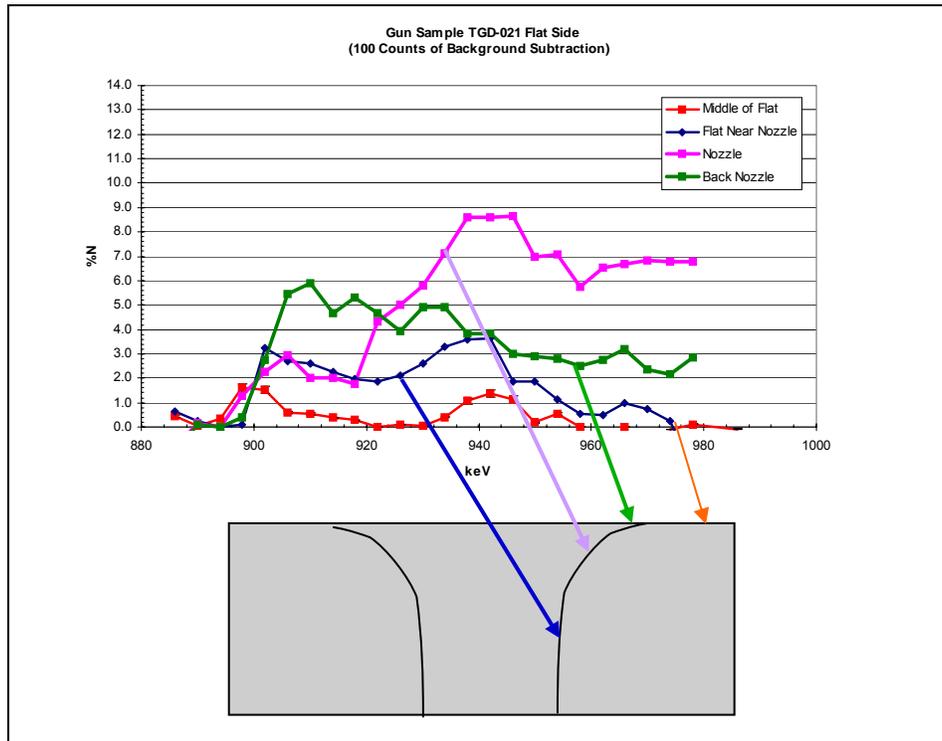


Figure 6. Depth profiles of nitrogen taken using NRA at four different positions relative to the inner exposed surface of the test nozzle.

Possible explanations for how surface nitriding can assist in erosion reduction come from surface chemistry, as well as from the binary phase diagram of iron nitride.

Literature suggests that nitrogen on a steel surface may inhibit dissociation of CO (Ponec and van Barneveld, 1979). Thus, dynamic nitriding can act to interrupt the primary driving reaction of the chemical erosion mechanism, which is the adsorption/dissociation of carbon monoxide on the surface. Also, the presence of nitrogen takes up sites in solution with iron and could inhibit the subsurface diffusion of the free carbon into the steel.

Carburization of the gun reduces the melt temperature of the surface from 1723 K to 1423 K. The binary iron-carbon phase diagram in figure 7 shows how this reduction in melt temperature occurs. The virgin 4340 or ASTM A723M-02 (2006) steel begins with about 0.4% carbon. As carbon is added to this mixture, the melt temperature continuously reduces until it stabilizes at 1423 K, which defines the boundary between cast iron and steel. Once the carbon concentration exceeds 2.1%, the material is then cast iron. Erosion calculations have shown that the surface can saturate with carbon providing the best boundary source available for diffusion (Conroy et al., 2001). This enables the formation of substrate iron carbide at the maximum rate possible. As the ballistic cycle progresses the surface temperature rises, all the while carbon is diffusing and creating  $Fe_3C$ . Once the surface temperature reaches 1423 K, the energy balance on the surface including phase transformation results in macroscopic melting. This is one of the primary mechanisms for the removal of surface and interfacial material (Conroy et al., 2001).

The iron/nitrogen phase diagram presented in figure 8 (Guillermet and Du, 1994) demonstrates how nitrogen in solution with iron can be more resistant to erosion than carbon in solution with iron under ballistic conditions. The phase transition from solid to liquid for iron nitride is shown as a constant 1683 K, between 7 and 20 atomic percent nitrogen. This presents a remarkable 260 K increase in the surface melt temperature over that of the iron carbide at 1423 K. For chrome plated tubes, this increase is obtained at the weakest point of the system, which is at the interface between the chrome and steel at the bottom of cracks in the chrome coating (Conroy et al., 2001). This represents an enormous opportunity for improvement for gun systems in terms of tube life and possibly system performance.

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### **3. Temperature Reduction Due to Expansion**

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In addition to the dynamic nitriding, nitrogen can also reduce the product gas temperature in regions of expansion due to higher intermolecular attraction forces and larger product molecular size through the Joule and Joule-Thomson effects.

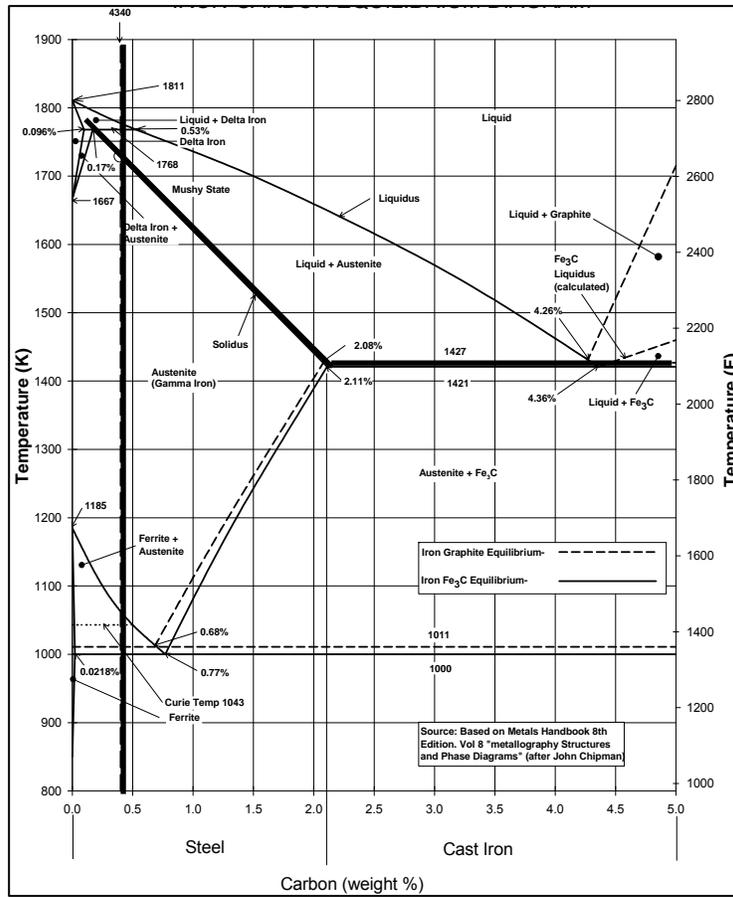


Figure 7. Iron/carbon phase diagram (Metals Handbook, 1973).

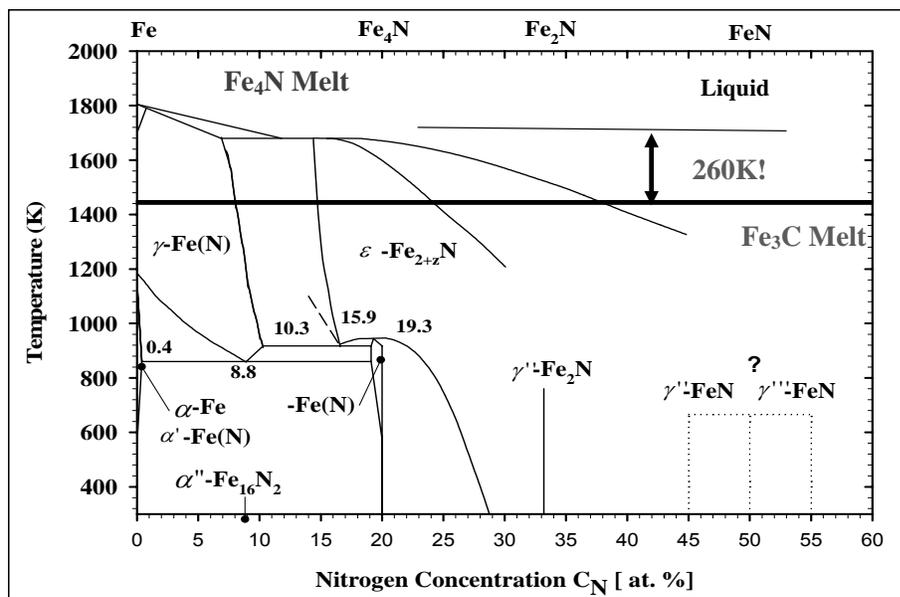


Figure 8. Iron/nitrogen phase diagram (Guillermet and Du, 1994).

Two processes have been considered for their effect on temperature reduction: isenthalpic expansion (throttling) and free expansion of gases (Sears and Salinger, 1975; Van Wylen and Sonntag, 1985). These processes relate in some respects to the expanding gases found in a gun during the interior ballistic cycle. The following analysis is an attempt to physically describe why the gases cool upon expansion as well as the effect that mixture composition might have upon the expanding combustion products.

### 3.1 Isenthalpic Expansion

To begin with a description of isenthalpic expansion, we write the enthalpy equation as the internal energy plus the work term  $pv$  as shown:

$$h = u + pv . \quad (1)$$

The indefinite derivative of the enthalpy with no work is

$$dH = TdS + VdP . \quad (2)$$

Using the identity

$$TdS = C_p dT - T \left( \frac{\partial V}{\partial T} \right)_p dP , \quad (3)$$

one can show that for an isenthalpic process that

$$VdP = T \left( \frac{\partial V}{\partial T} \right)_p dP - C_p dT , \quad (4)$$

and subsequently,

$$V = T \left( \frac{\partial V}{\partial T} \right)_p - C_p \left( \frac{\partial T}{\partial P} \right)_h . \quad (5)$$

With some rearrangement, this reduces to the familiar expression for the Joule-Thomson coefficient

$$\mu = \left( \frac{\partial T}{\partial P} \right)_h = \frac{1}{C_p} \left( T \left( \frac{\partial V}{\partial T} \right)_p - V \right) . \quad (6)$$

If we assume an ideal gas, this expression reduces to zero, while if we assume a Van der Waals equation of state, then the Joule-Thomson relationship becomes the following:

$$\mu = \left( \frac{\partial T}{\partial P} \right)_h = - \frac{1}{C_p} \left( \frac{RTv^3b - 2av(v-b)^2}{RTv^3 - 2a(v-b)^2} \right) . \quad (7)$$

When  $\mu$  is equal to zero in this relationship the function defines the temperature inversion curve, which is the limit at which a specific species will cool if expanded when initially below the inversion temperature and heat if expanded above the inversion temperature. The relationship of the inversion temperature is

$$T_i = \frac{2a(v-b)^2}{Rv^2b} . \quad (8)$$

From table 2, the inversion temperature shows that reducing or replacing the hydrogen with nitrogen would be beneficial from a temperature reduction point of view during an isenthalpic process.

Table 2. Van der Waals constants comparisons of products.

Product Species	Van der Waals Constant a (Intermolecular Attractive Force Constant) (kJm <sup>3</sup> /kmole)	Van der Waals Constant b (Molecular size constant) (m <sup>3</sup> /kmole)	Inversion Temperature (K) (@ v = 0.4m <sup>3</sup> /kg)
H <sub>2</sub>	25	0.0266	1000.
N <sub>2</sub>	137	0.0387	7803
CO	148	0.0395	8610
CO <sub>2</sub>	365	0.0428	23032
H <sub>2</sub> O	553	0.0305	24656

Unfortunately, there will be a penalty in performance for such a propellant, because the impetus will suffer as the average molecular weight of the products is increased as equation 9 shows:

$$Impetus = \frac{RT}{MW} . \quad (9)$$

### 3.2 Free Expansion

Of course, what happens in a real gun is expansion with work. An adiabatic system with no work undergoing free expansion might be more representative of what occurs before the throat of a nozzle. In this case the internal energy is constant and the change in volume is accompanied by a change in temperature. Differentiating the internal energy with the aforementioned constraints, as well as constant mass, results in

$$dU = \left( \frac{\partial U}{\partial T} \right)_{V,N} dT + \left( \frac{\partial U}{\partial V} \right)_{T,N} dV . \quad (10)$$

From this, we note that the change in internal energy is zero so that

$$dT = - \frac{\left( \frac{\partial U}{\partial V} \right)_{T,N}}{\left( \frac{\partial U}{\partial T} \right)_{V,N}} dV = - \frac{1}{C_V} \left( \frac{\partial U}{\partial V} \right)_{T,N} dV = \left( \frac{\partial T}{\partial V} \right)_{U,N} dV , \quad (11)$$

which is the differential Joule coefficient. If we assume an ideal equation of state, this term becomes zero and the effect is lost. However, if we assume the Van der Waals equation of state

$$\left(P + \frac{an^2}{V^2}\right)(V - nb) = nRT, \quad (12)$$

then through the following identity computed through differentiation of the first and second laws

$$\left(\frac{\partial U}{\partial V}\right)_T = T\left(\frac{\partial P}{\partial T}\right)_V - P, \quad (13)$$

the following relationship can be derived describing the change in internal energy with respect to volume:

$$\left(\frac{\partial U}{\partial V}\right)_T = \frac{an^2}{V^2}. \quad (14)$$

This implies that the larger the intermolecular attractive force the larger the temperature decrease will be. Therefore, from table 1, the more hydrogen that can be replaced by nitrogen in the combustion products the lower the temperature will be in the entrance region of the nozzle. This once again implies a negative impact upon the impetus of the propellant, as equation 9 shows.

A general conclusion is that a reduction in expansion temperature due to the replacement of hydrogen with nitrogen in the products will be accompanied by a reduction in impetus for a gun propellant. While the erosion might be reduced through the Joule and/or Joule-Thomson effects, the impetus of the propellant will decrease through the overall increase in average molecular weight. Thus, there is an apparent balance between erosion and performance. However, it may be possible to increase the loading density through advanced charge designs to overcompensate for the reduced impetus, which would result in an overall performance increase while reducing/minimizing the erosion.

## 4. Pre-nitriding

The discovery of dynamic nitriding leads to an exciting corollary: pre-nitriding a gun tube could provide significant benefits. Nitriding has been performed for over half a century on gun tubes (Di Pietro, 1947). The M242 Bushmaster cannon has specifications stating that the bore can be either chrome plated or nitrided. Current nitriding results in a very reproducible wear and barrel life in comparison to chrome plating, which is sometimes less reproducible (Waterfield, 2003). Unfortunately, nitrided M242 barrels do not have the erosion life of the chrome barrels. We may now be able to explain this result.

Conventional nitriding produces an error function distribution of nitrogen in the surface. This distribution ranges from a hard white layer (FeN) on the surface, through all the substoichiometric nitrides of iron, to deep into the iron where there is no nitrogen present. This implies a melting temperature range from as low as 1200 K at the surface up to the optimal temperature of 1683 K where the nitrogen content ranges from 7–20 atomic percent (figure 8). Unfortunately, by the time the erosion progresses until the surface has an optimal melt temperature there is not much nitrogen left, as shown in figure 9 in the traditional nitride profile.

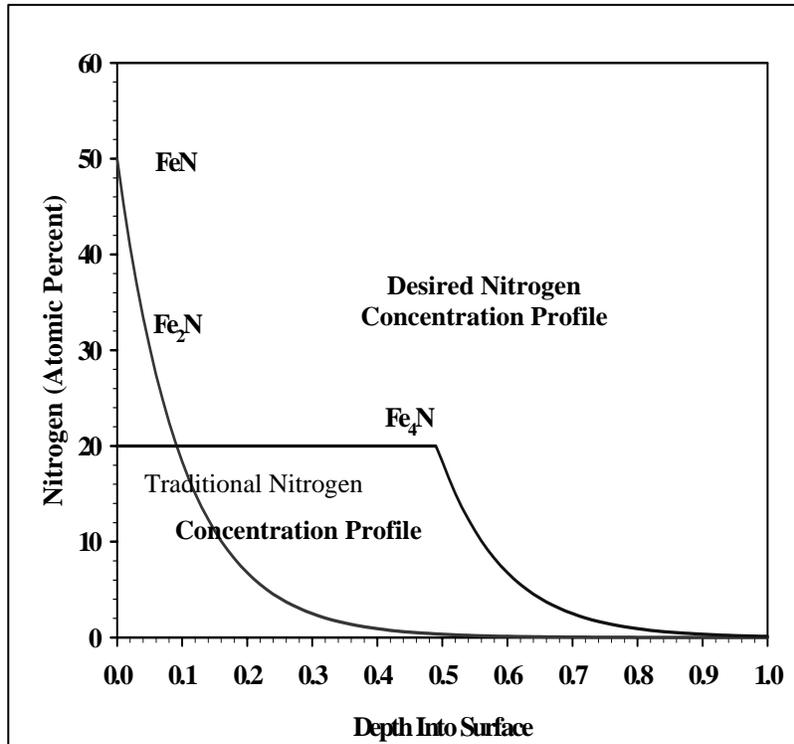


Figure 9. Traditional and preferred nitrogen profiles.

Conversely, if the nitrogen profile appears as in the desired nitrogen profile of figure 9, then the benefit is realized throughout the life of the gun tube. This profile may be reached through laser nitriding (Schaaf, 2002).

The benefit could also be realized if the surface of chromed or otherwise coated tubes were pre-nitrided to protect the interface between the coating and substrate steel from degradation when cracks occur.

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## **5. Conclusions**

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The dynamic nitriding and the temperature reduction due to propellant gas expansion have been shown to lead to remarkably reduced erosion resulting from propellant formulations having relatively high nitrogen content.

The new energetic propellant formulations that include “high nitrogen” ingredients can lead to reduced gun barrel erosion for both legacy systems and future armaments. This discovery is applicable to every gun system in the Department of Defense inventory, including mortars. Furthermore, for the first time propellant formulators have the ability and guidance to produce inherently less erosive propellants through energetic ingredient selection.

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## List of Symbols, Abbreviations, and Acronyms

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a	Van der Waals intermolecular force constant
b	Van der Waals molecular volume constant
$C_p$	specific heat at constant pressure
h	specific enthalpy
MW	molecular weight
P	pressure
R	ideal gas constant
S	entropy
T	temperature
u	specific internal energy
V	volume
v	specific volume
$\mu$	Joule Thomson coefficient

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