AD-F300212

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ADA126129

TECHNICAL REPORT ARBRL-TR-02479

MUZZLE FLASH AND ALKALI SALT INHIBITION
FROM AN ELEMENTARY KINETIC
POINT OF VIEW

Joseph M. Heimerl

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US ARMY ARMAMENT RESEARCH AND DEVELOPMENT COMMAND
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ABERDEEN PROVING GROUND, MARYLAND

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REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM
1. REPORT NUMBER TECHNICAL REPORT ARBRL-TR-02479	2. GOVT ACCESSION NO. AD-A126 129	3. RECIPIENT'S CATALOG NUMBER
4. TITLE *(and Subtitle)* MUZZLE FLASH AND ALKALI SALT INHIBITION FROM AN ELEMENTARY KINETIC POINT OF VIEW		5. TYPE OF REPORT & PERIOD COVERED
		6. PERFORMING ORG. REPORT NUMBER
7. AUTHOR(s) J.M. HEIMERL		8. CONTRACT OR GRANT NUMBER(s)
9. PERFORMING ORGANIZATION NAME AND ADDRESS U.S. Army Ballistic Research Laboratory ATTN: DRDAR-BLI Aberdeen Proving Ground, MD 21005		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS 1L161102AH43
11. CONTROLLING OFFICE NAME AND ADDRESS US Army Armament Research & Development Command US Army Ballistic Research Laboratory (DRDAR-BL) Aberdeen Proving Ground, MD 21005		12. REPORT DATE March 1983
		13. NUMBER OF PAGES 36
14. MONITORING AGENCY NAME & ADDRESS*(If different from Controlling Office)*		15. SECURITY CLASS. *(of this report)* UNCLASSIFIED
		15a. DECLASSIFICATION/DOWNGRADING SCHEDULE

16. DISTRIBUTION STATEMENT *(of this Report)*

Approved for public release; distribution unlimited

17. DISTRIBUTION STATEMENT *(of the abstract entered in Block 20, If different from Report)*

19. KEY WORDS *(Continue on reverse side if necessary and identify by block number)*

Muzzle Flash	Flash Suppression
Flame Inhibition	Elementary Chemistry
Muzzle Blast	Alkali Salts
Potassium Salts	Flash Prediction

20. ABSTRACT *(Continue on reverse side If necessary and identify by block number)*
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     This report reviews the current state of knowledge concerning the gas kinetic mechanisms involved in the suppression of muzzle flash by alkali-salts. Elementary kinetic mechanisms are discussed. A potentially important apparent discrepancy between two laboratory measurements of flame inhibition is critically reviewed. Suggestions and recommendations for future work are given.

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I. INTRODUCTION

It is the purpose of this report to review what is presently known about the gas kinetic mechanism involved in the suppression of muzzle flash by alkali-salts. The next section defines the three important regions of flash associated with a gun firing. The third section touches upon the practical importance of a solution to muzzle flash. The fourth section discusses the means of suppression, mechanical and chemical. The fifth section, comprising the bulk of this report, details kinetic concepts, measurements and their interpretations. The sixth section outlines recommendations.

II. DEFINITIONS

Drum-camera recordings of muzzle effluent reveal the existence of three luminous regions that are separated in time and space.[1] The first region, located at the muzzle, is of small spatial extent and low luminosity. The radiation from this region is commonly called the primary flash. Farther from the muzzle is a more extensive region of greater luminosity. It is separated from the primary flash by a dark zone and is called the intermediate flash. Still farther from the muzzle and adjacent to the intermediate flash is an extensive and very luminous region called the secondary flash. Secondary flash is known to occur downstream from the location of the normal inner shock (the mach disk) at pressures around one atmosphere and temperatures comparable to muzzle exit temperatures.[2] When it occurs, the secondary flash is much more extensive and much brighter than the other two regions and is commonly called the "muzzle flash." In this paper we shall use the term muzzle flash to refer to the secondary flash region.

The sources of these regions are thought to be understood[1,3] even if, as we shall see below, a detailed understanding is lacking. The primary flash is caused by radiation from the high temperature effluent, while shock heating is the source of the intermediate flash. The secondary flash is caused by the reignition of the fuel-rich, shock-heated effluent and the oxygen from the entrained air.

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[1] G. Klingenberg and H. Mach, "Investigation of Combustion Phenomena Associated with the Flow of Hot Propellant Gases - 1 Spectroscopic Temperature Measurements Inside the Muzzle Flash of a Rifle," Combustion and Flame, Vol. 27, pp. 163-176, 1976.

[2] G. Klingenberg, "Investigation of Combustion Phenomena Associated with the Flow of Hot Propellant Gases. III: Experimental Survey of the Formation and Decay of Muzzle Flow Fields and of Pressure Measurements," Combustion and Flame, Vol. 29, pp. 289-309, 1977.

[3] Engineering Design Handbook: Spectral Characteristics of Muzzle Flash, AMCP-706-255 (June 1967).

Strong support for the source of secondary flash has been obtained by Klingenberg.[1] By firing rounds that were known to produce muzzle flash into surrounding atmospheres of nitrogen and oxygen, he found that, relative to firing into air, the muzzle flash was completely suppressed with nitrogen and was sensibly enhanced with oxygen. He also found that both the primary flash and the intermediate flash were unaffected by the changes in surrounding atmospheres. In much earlier work, Ladenburg[4] reported that, when surrounding atmospheres of nitrogen or carbon monoxide were used, the muzzle flash was completely eliminated, but the intensity of both the primary and intermediate flashes remained unchanged.

III. IMPORTANCE

The importance of suppressing the muzzle flash lies in three areas. In the field the brilliant flash is easily observed and so reveals gun positions. The muzzle flash can also cause temporary loss of vision of the gun crew, especially at night. Since the amount of energy released in muzzle flash is comparable to that released inside the gun.[3] the concomitant muzzle blast of present day high performance weapons can lead to overpressures that exceed those due to the primary blast. For large caliber gun systems these combined overpressures can be so great[5] as to cause physical damage to the gun system and nonauditory physiological damage to a nearby gun crew. Keller[6] has reported a strong correlation between the brightness of a flash and the intensity of the corresponding blast overpressure. Thus, the suppression of secondary flash is expected to lead to the suppression of the associated blast.

IV. SUPPRESSION BY MECHANICAL/CHEMICAL MEANS

1. Mechanical

The mechanical attachments to the muzzles of small caliber guns have proved effective; but, for large caliber guns such mechanical devices would increase cost and weight to the gun system, and the devices themselves would be susceptible to damage during handling.[3] As an illustration let us assume that dimensions scale linearly with caliber. (This would be valid for inviscid, nonreacting flows and similar muzzle exit conditions.) Then let us scale up the mechanical suppressor of the M-16 rifle to a device suitable for an 8-inch gun. We find about three tons of material, about 2 meters in length, would have to be attached to the end of the 5-meter-long gun barrel. Placing tons of material at the end of a long moment-arm would require expensive redesign of the gun and gun carriage. Thus, the mechanical method is not an acceptable solution for large caliber gun systems.

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[4]R. Ladenburg, "Report on Muzzle Flash," BRL Report No. 426, Nov. 1943 (AD896671).

[5]See, for example, G. Soo Hoo, "Gun Blast Experiments With An 8"/51 Gun," NSWC/DL Tech. Note TN-T-1/75, Feb. 1975.

[6]G.E. Keller, "The Effect of Propellant Composition on Secondary Muzzle Blast Overpressure," BRL Memorandum Report- in final review.

2. Chemical

The suppression of muzzle flash by chemical means leads to two quite distinct approaches. Since muzzle flash requires, among other things, a fuel-rich effluent, operating with a liquid propellant that is stoichiometrically balanced would preclude muzzle flash events. Though such a propellant system may someday be realized, at the present time liquid propellants have not been fielded and are at the stage of a research and development project.

The other chemical approach has been the addition of suppressant compounds to the solid propellants. Historically, these have been the alkali metal salts, the most common being potassium sulfate, K_2SO_4, and potassium nitrate, KNO_3.

One might think that simple addition of alkali salt would suffice. However, all such additions lead to a degradation in ballistic performance. To compensate for this degradation, the amount of propellant is increased. This iterative cycle of suppressant addition/propellant compensation encounters two limitations. First, the volume of the breech of a weapon is limited, and for a given charge there simply may not be sufficient volume to insert the proper amount of suppressant material and additional charge. Second, Carfagno's[7] empirical correlations for propellants composed of H-C-N-O atoms show that alkali salt concentrations above about 2% are of dubious value in suppressing flash. So the simple addition of more salt turns out not to be a viable solution to suppress muzzle flash in the more recent, high-performance, large-caliber gun systems.

This leads us to ask the question: "What is the chemical mechanism by which muzzle flash occurs?" In the answer to this question lies the potential for tailoring both charge and suppressant to achieve a desired ballistic performance.

V. ELEMENTARY CHEMISTRY OF FLASH SUPPRESSION

1. Homogeneous Gas Phase Process

In our discussion of the elementary chemistry, we adopt the point of view that downstream of the mach disk the suppression mechanism interferes with a homogeneous gas phase ignition process and is itself a homogeneous gas phase process. Though this cannot be proved, there are several factors that indicate this is a reasonable postulate. First, the empirical correlation[7] (noted above) for large caliber guns is indicative of a chemical rather than a physical process; otherwise, the more salt added to the system, the greater the suppression would be expected to be.

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[7] S.P. Carfagno, "Handbook on Gun Flash," The Franklin Institute, Nov. 1961, (AD 327051).

In the laboratory, Rosser et al [8] have added small amounts (~ 1-3% by weight) of sodium and potassium salts to methane-air flames and observed reductions in the flame speed of factors of five or so. Similar amounts of $NaHCO_3$ when added to an ammonia-oxygen-nitrogen flame affected the measured flame speed by only about 20%. In addition, they observed that the inhibiting effect of Na_2CO_3 upon a methane-air flame could be reversed by addition of 0.86% methyl chloride, CH_3Cl. (By itself CH_3Cl is a known inhibitor of hydrogen-air flames.[9]) They also calculated that under flame conditions appreciable vaporization and dissociation of the alkali salts should take place.

Iya et al [10] used the rise in temperature of a flat, premixed, methane-air flame as a measure of inhibition. They added sodium bicarbonate, $NaHCO_3$, and sodium tartrate, $Na_2C_4H_4O_6 \cdot 2H_2O$ at selected particle sizes to the flame and found a one-to-one correlation between the observed atomic sodium concentration from vaporized salts and the observed temperature rise. This correlation was independent of particle size and composition and was much better than a correlation based on surface area presented to the flame.

Taken together these experiments strongly suggest the combustion process is inhibited by a homogeneous chemical process. Birchall[11] added alkali oxalates and alkali ferrocyanides to turbulent diffusion flames of town gas and air ($CO/H_2/air$) and reached the same conclusion.

The case is by no means as clear-cut as the above discussion might indicate. For a fuller discussion of the types of inhibition mechanisms, the reader is referred to reviews on the subject.[12-14] We also note that the

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[8]W.A. Rosser, Jr., S.H. Inami and H. Wise, "The Effect of Metal Salts on Premixed Hydorcarbon-Air Flames," *Combustion and Flame*, Vol. 7, pp. 107-119, 1963.

[9]D.R. Miller, R.L. Evers and G.B. Skinner, "Effects of Various Inhibitors on Hydrogen-Air Systems," *Combustion and Flame*, Vol. 7, pp. 137-142, 1963.

[10]K.S. Iya, S. Wollowitz and W.E. Kaskan, "The Mechanism of Flame Inhibition by Sodium Salts," The 15th Symposium (International) on Combustion, pp. 329-336, 1975.

[11]J.D. Birchall, "On The Mechanism of Flame Inhibition by Alkali Metal Salts," *Combustion and Flame*, Vol. 14, pp. 85-95, 1970.

[12]R.M. Fristrom, "Combustion Suppression," *Fire Res. Abs. and Rev.*, Vol. 9, pp. 125-152, 1967.

[13]J.W. Hastie, "Molecular Basis of Flame Inhibition," *J. Res. NBS A. Phy. and Chem.*, Vol. 77A, pp. 733-754, 1973.

[14]E.T. McHale, "Survey of Vapor Phase Chemical Agents For Combustion Suppression," *Fire Res. Abs. and Rev.*, Vol. 11, pp. 90-104, 1969.

above discussion does not rule out the participation of particulates in the flash event. Such particles may be necessary[6],[15] to sustain the event. Our point of view here is the ignition process and its suppression and not the combustion process.

2. Model Requirements

Within the above framework, then, we wish to efficiently and economically "test" propellant/suppressant combinations. To do this a computer program[16],[17] has been developed. This program requires the specification of the reaction sequence appropriate to an adequate description of muzzle flash and its suppression. To do this we must know: 1) the identity of the kinetically important chemical species, 2) the reaction network that describes how these species interact and, 3) the rate coefficients that determine how fast these species interact.

a. Species

Knowledge of molecular identities in muzzle effluent is sparse. In one experiment[1] the species Ca, CaOH, CaO and CN have been spectroscopically detected and in another[3] OH, NH, NH_3, CaOH, CuCl, CuH, CuO and CuOH. (By material substitution the copper was shown to arise from the projectiles used.[3]) However, spectroscopically active species may or may not be kinetically important. To our knowledge, there are no experiments that have systematically indentified chemical species and measured their relative abundances. Such measurements are sorely needed.

We then must estimate what species are important from equilibrium computations of the combustion of H-C-N-O type propellants. They are the expected products of combustion, N_2, CO_2 and H_2O, together with significant amounts of H_2 and CO.[18] These products are those for a rocket motor whose internal pressures are of the order of one atmosphere.

When the CEC-76 (NASA-Lewis) program is executed for M30A2 propellant at a typical muzzle pressure and temperature (i.e., 164.9 atmospheres and 1700k),

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[15] See, for example, S.R. Moore and F.J. Weinberg, "High Propagation Rates of Explosions in Large Volumes of Gaseous Mixtures," *Nature*, Vol. 290, pp. 39-40, 1981.

[16] V. Yousefian, I.W. May and J.M. Heimerl, "Modeling the *Occurrence of Muzzle Flash in Guns*," 17th JANNAF Combustion Meeting CPIA Pub. 329. Vol. II, pp. 125-140, Nov. 1980.

[17] V. Yousefian, "Muzzle Flash Onset," Contract Report ARBRL-CR-00477, Feb. 82 (AD B063573L).

[18] See, for example, E.T. McHale, "Experimental Evaluation of Chemical Agents in Suppressing Missile Plume After Burning," AIAA Paper No. 75-1233, AIAA/SAE 11th Propulsion Conference, Anaheim, CA, Sept 29 - Oct 1, 1975.

we find significant amounts (~ .001 mole fraction) $K_2CO_3(\ell)$ among the equilibrium products. When this program is run at one atmosphere, typical of rocket engines, no K_2CO_3 (ℓ) is produced. Should $K_2CO_3(\ell)$ actually form in the gun, there is the possibility that inhibition could occur via reactions on the surfaces of these droplets. Such a notion is consistent with the fact that the higher the temperature, the less effective is the alkali salt suppressor and with the independence of suppressor upon anion identity. Specifically, it could occur that KNO_3 and K_2SO_4 might have to form $K_2CO_3(\ell)$ prior to suppression. However, in what follows below, we have ignored this complication by assuming that there is not enough time for the liquid aerosol to form in significant quantities. Methods for proving or disproving this assumption have yet to be found.

b. Reactions

From the above it is reasonable to suppose that the gas phase chemistry involves all or part of the well-known $H_2/O_2/N_2$ reaction network[16,19] and reactions that convert CO to CO_2.[16,20] We also require reactions that describe the flash suppression mechanism.

The most important reaction promoting muzzle flash (or practically every fuel-rich H-C-N-O combustion process) is the chain branching reaction

$$H + O_2 \rightarrow OH + O + 71 \text{ kJ/mole.} \tag{1}$$

Once sufficient amounts of OH and O are formed, other reactions can proceed. Competition for the hydrogen atom of reaction (1) by an alternate kinetic path can lead to combustion inhibition.

c. Inhibition/Suppression Species and Reactions

As mentioned above, for real gun systems there is a paucity of data concerning alkali-salt suppression at the elementary chemistry level. In fact, there is no experimental evidence how K_2SO_4 or KNO_3 are converted into their equilibrium products. And so, for our ideas concerning alkali-salt suppression we necessarily turn our attention to the laboratory studies of flame inhibition. These experiments can indicate the controlling or dominant suppressant species and thereby permit construction of plausible suppressant mechanisms. Below we briefly review the key laboratory studies.

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[19]G. Dixon-Lewis, "Kinetic Mechanism Structure and Properties of Premixed Flames in Hydrogen-Oxygen-Nitrogen Mixtures," Phil. Trans. of Roy. Soc., London, Vol. 291, pp. 45-99, 1979.

[20]D.E. Jensen and G.A. Jones, "Reaction Rate Coefficients for Flame Calculations," Combustion and Flame, Vol. 32, pp. 1-34, 1978.

Friedman and Levy[21] employed an opposed-jet diffusion flame of CH_4 and O_2 in their experiment. Each jet flow was suitably diluted with nitrogen. They added atomic potassium to the fuel-flow up to about 6% of the fuel and did not observe any quenching of the flame. Similar amounts of CH_3Br did quench the flame and they concluded that atomic potassium is <u>not</u> the inhibiting species. They proposed that gaseous KOH is the species responsible for flame inhibition by potassium-containing salts. They supposed that it might act through reactions such as

$$KOH + H \rightarrow H_2O + K \qquad - 138 \text{ kJ/mole} \qquad (2)$$

or

$$KOH + OH \rightarrow H_2O + KO \qquad \sim + 22 \text{ kJ/mole}. \qquad (3)$$

McHale[22] studied afterburning suppression using solid rocket propellant' formulated with known amounts of selected potassium salts. For K_2SO_4, $KHCO_2$ and $K_2C_2O_4$, he observed the intensity of the 4.09 μ CO_2 bands to decrease by factor of about 15 relative to the unsuppressed case. Computation of the equilibrium species distribution showed that either K or KOH or both are the important species containing potassium. Relying on the results of Friedman and Levy, he then concluded that KOH and not K is the important suppressant species. This correlation between importance of KOH and observed suppression is strengthened by further observations. Namely, McHale added KBF_4 to the rocket propellant and found that the intensity of the CO_2 bands remained essentially the same as in the case with no KBF_4 addition. In this case the equilibium computations showed that <u>neither</u> KOH nor K is the important potassium-containing species.
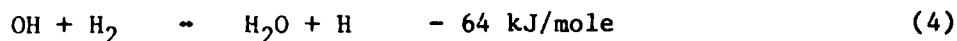
Cohen and Decker[23] performed shock-tube experiments to determine the effect of KOH, K_2SO_4 and KNO_3 on inhibiting H_2/O_2 explosions. Inhibition and suppression were observed in the experiments involving KOH (presumed in the gas phase), but not in the experiments involving K_2SO_4 or KNO_3, both in the form of particulates. These experiments: 1) experimentally confirm McHales's inference about the importance of KOH and 2) indicate that KNO_3 or K_2SO_4, routinely employed as flash suppressors, must first be physically or chemically converted before being effective. These results are consistant with a gas phase suppressant mechanism.

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[21] R. Friedman and J.B. Levy, "Inhibition of Opposed-Jet Methane-Air Diffusion Flames. The Effects of Alkali Metal Vapours and Organic Halides," *Combustion and Flame*, Vol. 7, pp. 195-201, 1963.

[22] E.T. McHale, "Flame Inhibition by Potassium Compounds," *Combustion and Flame*, Vol. 24, pp. 277-279, 1975.

[23] A. Cohen and L. Decker, "Chemical Mechanism For Secondary Flash Suppression," 18th Symposium (International) on Combustion, The Combustion Institute, pp. 225-231, 1981.

We now can construct the following qualitative picture. When oxygen is entrained into the fuel-rich effluent the chain-branching reaction (1) initiates a reaction sequence and secondary flash can occur. When an alkali salt is present reaction (2) competes with (1) for the radical species, atomic hydrogen. If there is sufficient KOH, then the end product, H_2O, is formed by reaction (2) and the reaction sequence leading to flash is bypassed. Since the fast reaction
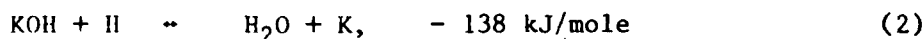
$$OH + H_2 \rightarrow H_2O + H \quad -64 \text{ kJ/mole} \quad (4)$$

tends to keep the H/OH ratio constant, the suppressant mechanism may also involve reaction (3).

d. Inhibition/Suppression Rate Coefficients

The rate coefficients of the suppressant reactions usually have been estimated.[20] In a carefully worded paper Jensen, Jones and Mace[24] reported the addition of potassium, in the form of potassium dipivaloylmethane, to a fuel-rich premixed $H_2/O_2/N_2$. They monitored the temperature and the hydrogen atom profile in the recombination region above the flame and observed a decrease in hydrogen atom concentration with increasing amount of suppressant. By assuming the two-step suppressant mechanism

$$K + OH + M \rightarrow KOH + M \quad -362 \text{ kJ/mole} \quad (5)$$

and

$$KOH + H \rightarrow H_2O + K, \quad -138 \text{ kJ/mole} \quad (2)$$

and by fitting the observations, they were able to deduce expressions for these rate coefficients.

Because the net result of reactions (5) and (2) is $OH + H \rightarrow H_2O$ this suppressant mechanism looks attractive. However, there appears to be a contradiction between the experimental results of Friedman and Levy[21] and the assumed suppressant mechanism of Jensen et al. In this mechanism, atomic potassium is obviously important and it should make no difference how the atomic potassium is added, provided only that the two experiments are comparable. In other words, if this mechanism is correct, why was there no flame quenching observed by Friedman and Levy?

e. Comparison of Experiments

Given the experimental observables of Jensen et al[24] and Friedman and Levy[21] we see that either the assumed mechanism (reactions 5 and 2) is incorrect or the experiments are not comparable. Are premixed flame and diffusion flame experiments ever comparable?

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[24] D.E. Jensen, G.A. Jones and A.C.H. Mace, "Flame Inhibition by Potassium," J. Chem. Soc. Faraday Trans. I, Vol. 75, pp. 2377-2385, 1979.

Simmons and Wolfhard studied extinction limits for methyl bromide, CH_3Br, added to several hydrocarbon-fuel/air premixed[25] and diffusion flames.[26] They found that the amount of CH_3Br required to extinguish the premixed flame was of the same order of magnitude as that for the diffusion flame, provided the CH_3Br was added to the air-feed. When added to the fuel-feed they found that much more CH_3Br had to be added to extinguish the flame. Gaydon and Wolfhard[27] discuss the comparison of diffusion and premixed flames in a general fashion. They indicate that the kinetics in one may be different from the kinetics in the other. We conclude that only under favorable circumstances will a premixed and diffusion flame give comparable results. Below, we shall adopt the attitude that the two experiments are comparable; we shall find out what assumptions are necessary to support this point of view and then critize the assumptions.

There are several categories that we shall examine to determine whether these two experiments are, in fact, comparable. These are:

(1) Flame type, i.e., methane-air vs. H2/O2/N2,

(2) Amount and method of potassium addition,

(3) Temperature, and,

(4) Amount of time for chemical reactions to occur.

To examine differences in flame type we require a knowledge of the (H + OH) radical pool for both experiments. Jensen et al provides values for both [H] and [OH], with [H]/[OH] $\simeq$ 5 to 10. (Their method depends on a superequilibrium hydrogen atom concentration.) Friedman and Levy do not provide this information. To provide a crude estimate we note that Hahn and Wendt[28] have computed the one-dimensional H and OH concentration profiles for a $CH_4/N_2/O_2$ opposed jet diffusion flame seeded with 10^{-3} moles of NH_3. They reported (H + OH) maximum radical concentrations comparable to Jensen et al but with [H]/[OH] $\simeq$ 0.25. We have assumed that the NH_3 does not affect the radical pool. In addition, since the experimental flow rates are not given, we have assumed that the model parameters are similar to the actual experiment. The necessity of these assumptions is a measure of the crudeness

[25] R.F. Simmons and H.G. Wolfhard, "The Influence of Methyl Bromide On Flames. Part 1 - Premixed Flames," Trans. Farad. Soc., Vol. 51, pp. 1211-1217, 1955.

[26] R.F. Simmons and H.G. Wolfhard, "The Influence of Methyl Bromide on Flames. Part 2 - Diffusion Flames," Trans. Farad. Soc., Vol. 52, pp. 53-59, 1956.

[27] A.G. Gaydon and H.G. Wolfhard, Flames, Their Structure, Radiation and Temperature, Chapman Hall, 3rd ED., Revised 1970, pp. 150-151.

[28] W.A. Hahn and J.O.L. Wendt, "Studies of Pollutant Formation in Opposed Jet Diffusion Flames," Final Report, Fundamental Combustion Research Program (U.S. Environmental Protection Agency), Nov. 1979; Fig. 41, p. 95.

of our estimate, and the most favorable statement that can be made is that the two experiments _may_ have similar amounts o the H + OH radical pool.

Jensen et al added $3X10^{14}$ to $3X10^{15}$ potassium atoms/cm^3 and Friedman and Levy added about $2X10^{17}$ potassium atoms/cm^3 as suppressant. A larger amount is seen to have been added to the experiment in which no inhibition was observed. In this comparison we assume in the former that one molecule of potassium dipivaloylmethane produces one potassium atom and in the latter that the atomic potassium reaches the flame zone without reacting.

By using N_2 as a diluent Jensen et al are able to adjust the temperature in the recombination zone from 1800 to 2200K. For a given $H_2/O_2/N_2$ mixture this temperature is observed to be constant. On the other hand, we compute adiabatic flames temperatures of 2285K ($[K]/[CH_4]$ = 0.035) and 1742K ($[K]/[CH_4]$ = 0.061) for the experimental parameters given by Friedman and Levy (their Table 1). This is an estimate of the maximum temperature attained between the jets. There is a continuous temperature distribution from one jet to the other.[29] Note that the greater addition of potassium corresponds to the lower (maximum) temperature.

Finally, the time available for reactions to occur in the experiment of Jensen et al is about one millisecond. Using the experimental parameters given by Friedman and Levy together with the results of Wendt and coworkers,[28,30,31] we estimate that the time available for reactions in the experiment of Friedman and Levy lies in the range 4 to 10 milliseconds. By these estimates, Friedman and Levy's experiment has at least as long a time available for reactions to occur as the experiment of Jensen et al.

To summarize, we find the supporting evidence to be rather flimsy in the area of radical pool size and composition, and the effect of a continuous temperature distribution for the opposed-jet experiment is not readily accounted for. The experiments are sufficiently different and sufficiently complex that results are mutually contradictory or both are correct in different operating regimes. There is sufficient circumstantial evidence, however, to cast doubt on the popular use of reactions (5) and (2) as elementary suppressant mechanism appropriate to alkai-salts.

We can ask whether the use of these reactions makes any difference in the computer program designed to predict muzzle flash.

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[29] _See, for example, Ref. 27, p. 419._

[30] _W.A. Hahn, J.O.L. Wendt and T.J. Tyson, "Analysis of the Flat Laminar Opposed Jet Diffusion Flame with Finite Rate Detailed Chemical Kinetics," Combust. Sci. and Tech., Vol. 27, pp. 1-17, 1981._

[31] _W.A. Hahn and J.O.L. Wendt, "NO$_x$ Formation in Flat, Laminar, Opposed Jet Methane Diffusion Flames," 18th Symposium (International) on Combustion, The Combustion Institute, pp. 121-131, 1981._

f. Application in the Model

We can assume for the moment that reactions (5) and (2) properly describe the suppression mechanism and ask what the results are from the model. The model itself has been described in detail[17] and the kinetic network employed is shown in Table 1. The first 13 reactions involving the species O, O_2, H, OH, H_2, H_2O, CO, CO_2, K and KOH were the only ones initially considered. The model results for three different types of propellants are shown in Table 2. As can be seen the predictions are quite good. (The onset of muzzle flash was determined from the output of the computer code by a steep rise in temperature; see Figure 1.)

Two difficulties were encounted, however. The first was the realization that the inhibitor action must be operating at temperature below 1000K. (See Figure 1 and compare the curve M30A1 with that for M30A2.) Below about 1100K the three-body reaction forming HO_2, reaction 14 in Table 1, effectively competes and dominates the two-body reaction involving the same reactants, reaction 6. Thus, HO_2 chemistry had to be added to the reaction network. Second, in performing sensitivity tests on the rate coefficients, it was found that the coefficient for reaction 1 in Table 1 could be set to zero with no change in the code predictions! The obvious interpretation is that this reaction is unnecessary in the description of muzzle flash suppression.

If just the HO_2 chemistry reactions 14-18 were added, then the predictions changed and no flash was predicted for any of the three propellants. From the above sensitivity test we knew that the entire suppressant action was from reaction 2. Kaskan[32] had shown in the laboratory that other reactions besides this one were important. He suggested the formation of KO_2 by a three-body reaction analogous to reaction 14 and so the reactions 19-22 of Table 1 were postulated and their rate coefficients estimated. The computer code with the entire set of reactions then was able to reproduce the observed flash/no-flash conditions for the three propellants. Later,[33] using this same full set of kinetics, four of five other cases were accurately predicted. The two cases of eight not predicted accurately were both borderline flash cases in the field. Further validation tests to probe the limits of the accuracy of the code were not done.

For purposes of this paper, then, we conclude that reaction 1 of Table 1 does not appear to be important, reaction 2 can not be the only suppressant reaction and other reactions involving alkali-containing molecules are sufficient to describe flash suppression. However, there remains the possibility that these predictions based upon the chemical network of Table 1, are fortuitous. Neither KOH nor KO_2 have been detected in the laboratory or in the field and the question still remains: What are the measured alkali-containing species in an inhibited flame? Are KO_2, KO, etc., observed? Are

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[32]W.E. Kaskan, "The Reaction of Alkali Atoms in Lean Flames," *Tenth Symposium (International) on Combustion*, pp. 41-46, The Combustion Institute, 1965.

[33]V. Yousefian and G.E. Keller (private communication, 1982).

TABLE 1. REACTIONS AND THEIR FORWARD RATE COEFFICIENTS

REACTION	RATE COEFFICIENT*	UNCERTAINTY
1. $K + OH + M = KOH + M$	$1.5(-27) \ T^{-1}$	2.5
2. $K + H_2O = KOH + H$	$5.0(-10) \ exp \ (-20,000/T)$	3
3. $CO + OH = CO_2 + H$	$2.8(-17) \ T^{1.3} \ exp \ (330/T)$	3
4. $CO + O_2 = CO + O$	$4.2(-12) \ exp \ (-24,000/T)$	3
5. $C + O + M = CO_2 + O$	$7(-33) \ exp \ (-2200/T)$	30
6. $H + O_2 = OH + O$	$2.4(-10) \ exp \ (-8250/T)$	1.5
7. $O + H_2 = OH + H$	$3.0(-14) \ T^{-1} \ exp \ (-4480/T)$	1.5
8. $OH + OH = H_2O + O$	$1.0(-11) \ exp \ (-550/T)$	3
9. $OH + H_2 = H_2O + H$	$1.9(-15) \ T^{1.3} \ exp \ (-1825/T)$	2
10. $O + H + M = O_2 + M$	$1.0(-29) \ T^{-1}$	30
11. $O + O + M = O_2 + M$	$3.0(-34) \ exp \ (900/T)$	10
12. $H + OH + M = H_2O + M$	$1.0(-25) \ T^{-2}$	10
13. $H + H + M = H_2 + M$	$3.0(-30) \ T^{-2}$	30
14. $H + O_2 + M = HO_2 + M$	$1.5(-32) \ exp \ (500/T)$	3
15a. $HO_2 + H = OH + OH$	$1.7(-10) \ exp \ (-500/T)$	4
15b. $HO_2 + H = H_2O + O$	$8.5(-12) \ exp \ (-500/T)$	5
15c. $HO_2 + H = H_2 + O_2$	$4.2(-11) \ exp \ (-350/T)$	3
16. $HO_2 + O = OH + O_2$	$3.5(-11)$	3
17. $HO_2 + OH = H_2O + O_2$	$3(-11)$	3
18. $HO_2 + CO = CO_2 + OH$	$2.5(-10) \ exp \ (-11,900/T)$	3
19. $K + O_2 + M \rightarrow KO_2 + M$	$1.0(-30) \ T^{-1}$	10
20. $KO_2 + H_2 = KOH + OH$	$3.0(-12) \ exp \ (-10,000/T)$	100
21. $KO_2 + OH = KOH + O_2$	$2.0(-11)$	30
22. $K + HO_2 = KO_2 + H$	$1.0(-11) \ exp \ (-1000/T)$	30

*Units of cm-particle-s, read 1.5(-27) as 1.5×10^{-27}

TABLE 2. COMPARISON OF CODE PREDICTIONS WITH
OBSERVATIONS FOR THREE PROPELLANTS

PROPELLANT	FLASH OBSERVATIONS	FLASH PREDICTIONS		
		1-13*	1-18	1-22
M30A1	Yes	Yes	No	Yes
M30A2	Usually	No	No	No
M31E1	No	No	No	No

*Reaction network used in code; numbers refer to those reactions listed in Table 1.
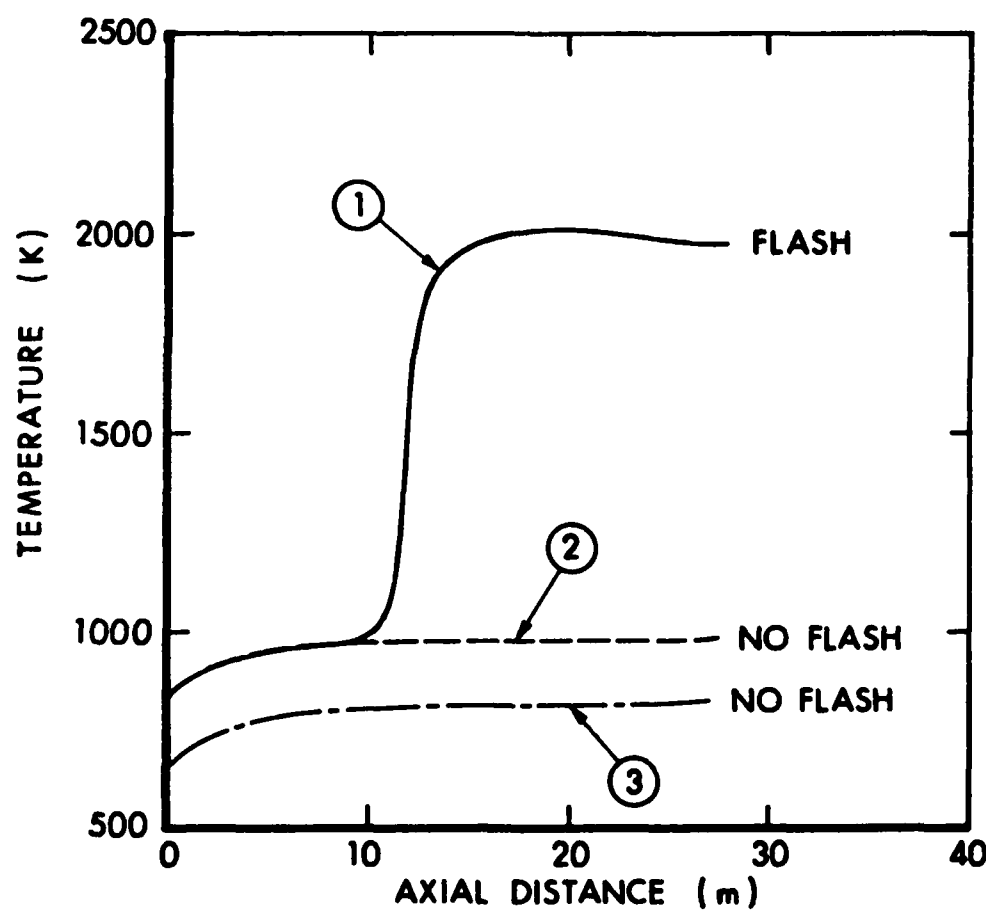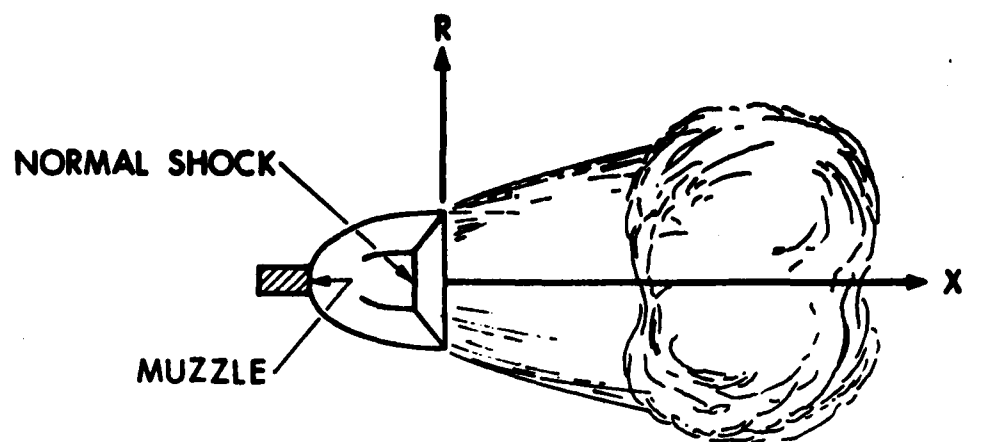
Figure 1. Computed Temperature vs. Distance for Three Propellants:
Curve 1, M30A1 (1% K_2SO_4 Suppressant); Curve 2, MA30A2
(2.7% KNO_3 Suppressant); Curve 3, M31E1 (1% K_2SO_4 Suppressant).
A schematic of flash is shown above the graph.

they important? These questions can not be answered without recourse to experimentation.

3. Some Comments

a. Control Molecules

Both groups used control molecules in their experiments. Friedman and Levy showed that bromine compounds (introduced in the same concentrations as K) were powerful flame inhibitors. We now know the basic kinetics of the HBr system[34] involves a subnetwork of Br and Br_2 reactions. Presumably, other bromine compounds produce similar networks. The kinetic network of the alkalis and their compounds is expected to be different (halogens and alkali metals are at opposite ends of the periodic table) and so the control used by Friedman and Levy simply demonstrates that their apparatus was operational and not that it could have or should have been able to detect alkali metal/compound inhibition. An experimentally simpler test of the direct addition of, say, KOH was not performed.

Jensen et al used 2,2,6,6 – tetramethyl-3, 5-heptanedione as a control and observed "no significant changes in flame temperature or composition." Here, the assumption is implicitly made that the enol form of the heptanedione, with a hydrogen atom "resonantly" bonded between two oxygen atoms, comes apart at the same temperature (flame location) and in the same manner as the potassium dipivaloylmethane. In this molecule the potassium is ionically bonded to the anion. The inhibition/suppression effectiveness of the alkali metals usually follows the order* Li < Na < K for the same anion.[13,14] Thus, a more proper control might have been lithium dipivaloylmethane rather than heptanedione. Because their method of hydrogen detection involved Li and LiOH, another method of measuring [H] would have to be employed.

b. Alternate Mechanisms

In this section we speculate on possible mechanisms that explain, at least qualitatively, the results of both the experiments of Jensen et al and Friedman and Levy. These are: The former observed the decrease of hydrogen atoms with increasing addition of potassium dipivaloylmethane (K-DPM) and the latter observed no inhibition effects due to potassium atoms. We also know that heptanedione does not affect the hydrogen atom concentration.

We can first suppose, as do Jensen et al, that

$$K\text{-}DPM \xrightarrow{M} K + R_{DPM} \cdot \tag{6}$$

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[34]G. Dixon-Lewis and R.J. Simpson, "Aspects of Flame Inhibition by Halogen Compounds," *Sixteenth Symposium (International) On Combustion*, pp. 1111-1119, The Combustion Institute, 1977.

*According to McHale[14] rubidium compounds are better inhibitors than potassium, but cesium shows a poorer inhibition property.

To be consistent with Friedman and Levy, we must assume K does not inhibit. The hydrogen atom decrease observed by Jensen et al could then be explained by

$$H + R_{DPM} \overset{M}{\rightarrow} \text{heptanedione.} \tag{7}$$

This global reaction removes hydrogen and forms heptanedione, a species that is known not to inhibit.

At the high temperatures measured in the recombination zone, 1800-2200K, the radical R_{DPM} and the heptanedione would be expected to decompose. (The actual thermochemistry, decomposition temperature and decomposition pathways are not known to this author.) We note, however, that the inhibition reactions (6) and (7) could take place in the cooler parts of the flame with hydrogen atoms transported from the recombination zone.

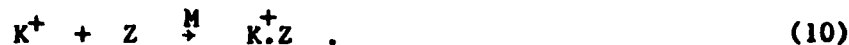Another decomposition pathway might involve ions:

$$K-DPM \overset{M}{\rightarrow} K^+ + R_{DPM}^-. \tag{8}$$

Two subsequent paths might follow; one could be:

$$R_{DPM}^- + H^+ \overset{M}{\rightarrow} \text{heptanedione.} \tag{9}$$

This requires that the H^+ -- H equilibrium be rapidly maintained and/or transported from the high temperature part of the flame to be sufficiently fast to produce the observed hydrogen atom depletion.

The other could be

$$K^+ + Z \overset{M}{\rightarrow} K^+ \cdot Z , \tag{10}$$

where Z is a polar or polarizable molecule. Hydrogen could then be depleted by the global reaction:

$$K^+ \cdot Z + H \rightarrow K^+ + \text{product.} \tag{11}$$

This sequence requires that $K^+ \cdot Z$ have a sufficiently long lifetime that it can react with H. The mechanisms that invoke ions are automatically consistent with the experimental results of Friedman and Levy.

Were any of the above sequences correct, then a comparison between the results of Friedman and Levy and Jensen et al would be moot; i.e., both would be correct.

VI. RECOMMENDATIONS

Based on the above discussions, we recommend the following general courses of action. First, in the field, measurements of radical species associated with suppressed and unsuppressed flash need to be made. Ideally, other important parameters, such as temperature and flow velocity, would be measured simultaneously. Second, in the laboratory, a survey experiment to determine the identity of the alkali-containing molecules of an intermediate nature; e.g., KO, KO_2, etc., must be made. This experiment would probably use a mass

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spectrometer as the diagnostic tool. Low pressures would be preferred so that the flame zone could be sampled. Such an experiment could be extended to include spectroscopic identifications of molecules in flames employing alkali salt suppressants. Finally, either or both experiments discussed above could be redone. In the flat flame burner experiment, measurement of species other than hydrogen (e.g., OH, K, etc.) should be carried out. Also, several potassium salts might be tried. In the opposed jet experiment, hydrogen, H_2 should be used as the fuel, and atomic potassium, as well as KOH and other potassium containing salts, should be added to the fuel stream.

It is my present opinion that the laboratory survey experiment has the greatest potential for advancing our understanding of the alkali-salt suppressant mechanism. Such an experiment has recently been undertaken.[35]

VII. ADDENDUM

Husain and Plane[36] have recently published measured rate coefficients for the reaction

$$Na + O_2 + M \rightarrow NaO_2 + M \qquad (12)$$

where M = N_2, He and CO_2. They found $k(M=N_2) = (1.0 \pm 0.24) \times 10^{30}$ cm^6 particle^{-2}s^{-1} with N_2: He: CO_2: : 1.0: 0.6: 2.0. They did not observe any significant temperature variation over the experimental temperature range 724 < T < 844K. Since we expect the equivalent potassium reaction to have a rate coefficient of the same order of magnitude, this measurement supports the rate coefficient expression used for reaction 14 in Table 1.

Jensen and Jones[37] repeated their flame inhibition studies for sodium dipivaloylmethane. Their results for both sodium and potassium are shown in Table 3. Based upon these results Jensen[38] was able to quantitatively reinterpret Kaskan's hydroxyl observations[32] taken in a lean $H_2/O_2/N_2$ flame inhibited by potassium chloride. Jensen did this without the need to postulate the presence of any potassium compound (e.g., KO or KO_2) other than the hydroxide. (The reinterpretation of Kaskan's sodium inhibited flames was

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[35] J. Hastie, NBS, private communication, 1981.

[36] D. Husain and J.M.C. Plane, "Kinetic Investigation of the Reaction Between Na + O_2 + M by Time-Resolved Atomic Resonance Absorption Spectroscopy," J. Chem. Soc. Faraday Trans. 2., Vol. 78, pp. 163-178, 1982.

[37] D.E. Jensen and G.A. Jones, "Kinetics of Flame Inhibition by Sodium," to be published in J. Chem. Soc. Faraday Trans. I, Vol. 78, 1982.

[38] D.E. Jensen, "Alkali-Metal Compounds in Oxygen-Rich Flames (A Reinterpretation of Experimental Results)," to be published, J. Chem. Soc. Faraday Trans. I, Vol. 78, 1982.

much less satisfactory.) Since the idea of postulating the superoxide, KO_2, rests upon Kaskan's interpretation, Jensen's reinterpretation underscores the speculative nature of the suppressant mechanism used in Table 1 and points to the need to experimentally identify the potassium containing species inhibited flames.

The basic problem of assuming the reaction mechanism to be reactions (5) and (2) may not have disappeared. The fact that corresponding rate coefficients of Table 3 produce similar values implies similar effectivenes in flame inhibition for both Na and K. This seems counter to the expectation that Na is less effective than K.[13,14]
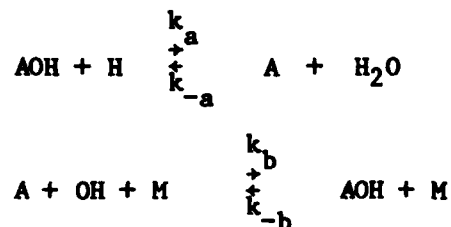
Referring to the speculative mechanism given by reactions (6) and (7), we note that if the slower, rate-controlling step were reaction (7), then the observed loss of atomic hydrogen would be about the same for sodium or potassium dipivaloylmethane addition. Thus, using reactions (5) and (2) as the assumed mechanism, as did Jensen, we should expect to find similar values of the rate coefficient. If we adopt this position, however, we necessarily imply that the quantitative fitting of Kaskan's hydroxyl results in a potassium inhibited flame are fortuitous.

Jensen[38] also estimated the bond energies of both Na-O_2 and K-O_2 as 170 ± 25 and 170 ± 30 kJ/mole, respectively. Figger et al [39] measured the K-O_2 dissociation energy as 189 ± 10 kJ/mole.

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[39]H. Figger, A. Kowalski and X.H. Zhu, "Chemiluminescent Reaction Between Alkali Dimers and Oxygen Molecules," talk given at *50 years of Reaction Dynamics*, Berlin, Oct. 1981, to be published in *J. Chem. Phys.*

Table 3.

Comparison of inferred rate coefficients for the scheme (A = Na or K):

$$AOH + H \quad \underset{k_{-a}}{\overset{k_a}{\rightleftharpoons}} \quad A + H_2O$$

$$A + OH + M \quad \underset{k_{-b}}{\overset{k_b}{\rightleftharpoons}} \quad AOH + M$$

	Na	K
k_a :	$1.8 \times 10^{-11} \exp(-1000/T)$[*]	$1.8 \times 10^{-11} \exp(-1000/T)$
k_{-a} :	$4.1 \times 10^{-10} \exp(-21{,}900/T)$	$5 \times 10^{-10} \exp(-20{,}000/T)$
k_b :	$5 \times 10^{-27} \ T^{-1}$	$1.5 \times 10^{-27} \ T^{-1}$
k_{-b} :	$2.5 \times 10^{-3} \ T^{-1} \exp(-39{,}300/T)$	$6 \times 10^{-4} T^{-1} \exp(-41{,}200/T)$

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[*]*Units are cm-particle-second; expressions are derived for the temperature range 1800-2200K.*

ACKNOWLEDGEMENTS

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