Survey of Near-Infrared Emitters

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Chemistry Division

November 10, 1997

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REPORT DOCUMENTATION PAGE

1. AGENCY USE ONLY (Leave Blank) | 2. REPORT DATE | 3. REPORT TYPE AND DATES COVERED
--- | --- | ---
 |
November 10, 1997 | |

4. TITLE AND SUBTITLE
Survey of Near-Infrared Emitters

5. FUNDING NUMBERS

6. AUTHOR(S)
Nancy L. Garland, H.H. Nelson, and A.P. Baronavski

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
Naval Research Laboratory
Washington, DC 20375-5320

8. PERFORMING ORGANIZATION REPORT NUMBER
NRL/MR/6111-97-8105

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
Office of Naval Research
800 N. Quincy Street
Arlington, VA 22217-5660

10. SPONSORING/MONITORING AGENCY REPORT NUMBER

11. SUPPLEMENTARY NOTES

12a. DISTRIBUTION/AVAILABILITY STATEMENT
Approved for public release; distribution unlimited.

12b. DISTRIBUTION CODE

13. ABSTRACT (Maximum 200 words)
This is a summary of chemical systems which emit radiation primarily in the near-infrared region. The requirements for the chemical system include: emission in the 700-900 nm region, little or no emission in the 400-700 nm region, narrow spectral features (to allow selective filtering), high spectral irradiance, and relative ease of chemical generation.

14. SUBJECT TERMS
Near-infrared emission
Chemiluminescence

15. NUMBER OF PAGES
13

16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT
UNCLASSIFIED

18. SECURITY CLASSIFICATION OF THIS PAGE
UNCLASSIFIED

19. SECURITY CLASSIFICATION OF ABSTRACT
UNCLASSIFIED

20. LIMITATION OF ABSTRACT
UL

NSN 7540-01-280-5500

Standard Form 298 (Rev. 2-89) Prescribed by ANSI Std 239-18
298-102
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SURVEY OF NEAR-INFRARED EMITTERS

INTRODUCTION

There is a Navy need for a small, inexpensive, expendable source of near-infrared radiation. Code 6111 has been tasked with surveying the literature, making recommendations for initial studies, and performing these initial studies to either specify the production of these sources or procure them outright. Due to the amount of radiation required, approximately 50 µW cm⁻² at a distance of 100-1000 m, and the limited space for power supplies or batteries, it is desirable to store most of the energy as chemical energy. Thus, highly exoergic reactions which produce a significant amount of excited state products are sought. The reactions being considered are similar to those employed in chemical laser systems, however the source need not lase, and consequently, some of the requirements are not as stringent. For example, in chemical laser systems, a population inversion is necessary whereas in the current application only a large fraction of the energy need be converted to IR radiation and so a population inversion is not required. Similarly, since it is desired that the source emit over 4π steradians, no laser cavity is needed. However, many of the systems considered have been studied for possible chemical laser applications leading to a fairly comprehensive database to draw upon.

This report is a summary of chemical systems which Code 6111 has screened for possible application to this problem. Specific requirements include: strong emission in the 700-900 nm region, little or no emission in the 400-700 nm region, narrow spectral features to allow for possible selective filtering, high spectral irradiance (on the order of 5 x 10⁻⁵ - 5 x 10⁻⁴ W/sr); and relative ease of chemical generation. Code 6111 has reviewed the available literature and identified a number of potential candidates as infrared emitters. These are summarized in Table 1 and briefly discussed below.

INITIAL CANDIDATES (700-900 nm)

BaF

The chemical reaction of alkaline earth metals (Ca, Sr, Ba) with halogenated species produces ground and electronically excited metal halides. Emission from excited calcium and strontium halides lies < 700 nm (Kierzkowski et al. 1996a and 1996b) while emission from excited BaI and BaBr lies > 900 nm (Bradford et al. 1975) excluding these species from further consideration. BaF* and BaCl* emit between 700-900 nm and are ideal initial candidates for the present project. Bradford et al. (1975) report that the electronically excited barium halides are generated almost exclusively in the A and B states, with <6% of the excited molecules produced in higher electronic states. Since the internuclear separations for all electronic states of a given BaX molecule are almost equal, the emission band consists of strong Δv=0 vibrational sequences. The narrow spectral bandwidth is important since it allows the use of spectral filtering. Figure 1 is an example of the chemiluminescence spectrum produced following reaction of Ba + F₂.

Absolute photon yields for the reactions of Ba with halogenated species were obtained by integrating emission band intensities and comparing to those of a calibrated standard. For the reaction Ba + F₂, Eckstrom et al. (1974) report a total photon yield of 1.2 x 10⁻² at 1.4 torr total pressure while Bradford,
et al. (1975) report that the yield of BaF decreases from a value of $6.6 \times 10^{-3}$ at 1 torr to a value of $1.4 \times 10^{-3}$ at 16 torr. Bradford et al. also report that the peak rate of production for this reaction at 5 torr total pressure is $1 \times 10^{13}$ photon/nm/s/Ba atom for the B-X emission and $\sim 1 \times 10^{14}$ photon/nm/s/Ba atom for the A-X transition. The reaction of Ba with NF$_3$ generates much more intense emission than the reaction of Ba + F$_2$ as Eckstrom et al. report a peak photon yield of 0.5. Bradford et al. (1975) observed BaF chemiluminescence following reaction of Ba with SF$_6$ to produce BaF$^*$ but they report no photon yields. Thus the reaction of Ba with NF$_3$ appears to be a promising candidate to generate an infrared flash emitter.

**BaCl**

The reaction of Ba with Cl$_2$ produces BaCl in the A- and B-states (as well as the X state) (Eckstrom et al. 1974; Bradford et al. 1975). Figure 2 is an example of a chemiluminescence spectrum.
obtained following the reaction of Ba + Cl₂. The A-X and B-X bands of BaCl overlap and cannot be separated in the emission spectra so all reported photon yields correspond to a combination of the two bands. Bradford et al. monitored emission extending to 1100 nm and reported a photon yield of 0.04 at 1 torr which decreases to 0.006 at 16 torr total pressure. The yield of 0.006 reported by Eckstrom et al. (1974) at 1.3 torr must be a lower limit since their experiment was insensitive to emission at wavelengths greater than 900 nm (where most of the lines in the A-X system lie).

For the reaction of Ba + NOCl, Obenauf et al. (1973) reported that BaCl was generated in the X and C states but little to no BaCl produced in the A state but Eckstrom et al. report a photon yield of 4.8 x 10⁻⁴ for C-X emission and a yield of 0.0025 for A-X + B-X emission; again, the yield for the latter bands must be regarded as a lower limit. Since this reaction generates low photon yields and may create visible emission (C-X band at 510 nm), it will not be useful in generating an infrared emitter.

Ar

Argon excited in a dc arc lamp emits radiation from 200-1400 nm with 40-50% of the total energy output between 700-900 nm. Figure 3 is an illustration of the emission from such a lamp filled with 6 atm Ar. Two intense lines at 810 nm and 840 nm are identified as 4p[3/2]₁ → 4s[3/2]₁ and 4p[5/2]₂ → 4s[3/2]₁, respectively, with transition probabilities of ~2 x 10⁶ s⁻¹ (Alonso-Medina 1996). The maximum total output power of a commercial lamp (Vortek) is 150 KW. Argon is a promising candidate since it possesses intense radiation in the near-infrared region with little emission in the visible. The spectral width of the emission lines is narrow and the transition probabilities are high.

![Argon emission from a dc arc lamp](image)

**Fig. 3** Argon emission from a dc arc lamp

BaOH

BaOH is included as a potential infrared emitter since the behavior of alkaline earth monohydroxides is expected to be similar to that of the alkaline earth halides (Kinsey-Nielsen 1986). BaOH is isoelectronic with BaCl and one might expect that the oscillator strength of BaOH to be similar to that of BaCl. BaOH⁺ generated from the reaction of ground state Ba (¹S) atoms with H₂O₂ (Cheong and Parson 1994) emits a weak, broad band centered around 830 nm. If the Ba reactant is then electronically
excited to the $^3$D state, the intensity of the BaOH emission increases dramatically. Both the B-X band at ~750 nm and the A-X band at 830 nm are observed but the emission is still broad with little structure. BaOH chemiluminescence has been detected following reaction of Ba ($^1$P$_1$) with water, methanol, and dimethyl ether (de Pujo et al. 1993). In contrast to the emission by the product of the reaction of Ba ($^1$S, $^3$D) + H$_2$O$_2$, the emission from Ba ($^1$P$_1$) with water and alcohols is sharp (FWHM = 10 nm) and structured. The sharp structure is an advantage if spectral filtering is desired but the one complication with this chemical system is the need for electronically excited Ba atom reactants. The reaction of ground state Ba ($^1$S) atoms with water and methanol generates ground state products (Davis et al. 1993).

HNO

The reaction of H with NO generates HNO* in the $^3$A$^*$ state and predominantly in the (000) level (Yoshimura et al. 1984). The $^3$A$^*$ (000) state has a radiative lifetime of ~23 $\mu$s (Obi et al. 1977) and emits to the X$^1$A$^*$ (000) state at 760 nm; the FWHM of the band is ~20 nm. The spectral characteristics (wavelength, spectral bandwidth) of the HNO* emission are attractive. Unfortunately, the rate constant of the reaction which produces HNO* (H + NO + M $\rightarrow$ HNO* + M) is $\leq$ 2 $\times$ 10$^{11}$ mol$^{-2}$ s$^{-1}$ (5.5 $\times$ 10$^{-32}$ cm$^6$ molecule$^{-2}$ s$^{-1}$) (Oka et al. 1977), much less than gas kinetic. The slow rate of HNO* production coupled with the small oscillator strength (Williams 1975) (100 times less than that of BaCl) and long radiative lifetime (Obi et al. 1979) prevent HNO* from becoming an initial candidate.

OH

The reaction of H with O$_3$ generates vibrationally excited OH radicals which emit in the visible, near-infrared and infrared regions. The Meinel bands in the earth’s nightglow arise from this reaction and the major bands in the nightglow include the (6,2), (5,1), and (9,4) bands at 840, 800, and 780 nm, respectively (Meinel 1950). The rate of the reaction of H + O$_3$ is rapid; the temperature dependence of the rate constant has been experimentally determined to be $k(T) = (1.33 \pm 0.32) \times 10^{-10}$ exp$[-(898 \pm 116)/RT]$ cm$^3$ molecule$^{-1}$ s$^{-1}$ (Lee et al. 1978). OH* is formed predominantly in high vibrational levels up to the energetic limit ($v = 8$ and 9) (Murphy 1971). At atmospheric pressure, the vibrationally excited OH molecules undergo collisional deactivation and emission occurs from low vibrational levels ($v = 1$-3) (McDade and Llewellyn 1987).

OH* produced by the reaction of H + O$_3$ is not considered as an initial candidate for a near-infrared emitter for two reasons. First, the radiative lifetime of the emitting states is long (3-50 ms) (Mies 1974) so the irradiance will be low. Second, the vibrational transition probabilities are highest for the $\Delta v = -1$ and -2 bands (Mies 1974) and these bands occur at $\lambda >$ 900 nm outside the spectral region of interest.

NaO

The reactions of Na + O$_3$ and Na + N$_2$O produce NaO chemiluminescence in the region 650-1000 nm. The B$^2$Π - X$^2$Π emission band appears as a single peak centered around 800 nm with a FWHM of 200 nm. Most laboratory studies of NaO chemiluminescence have explored the reaction of Na with N$_2$O even though the rate constant for the reaction of Na with O$_3$ (Plane and Rajasekhar 1989) is almost three orders of magnitude higher than that for reaction with N$_2$O (Ager et al. 1986). The oscillator strength of the B-X system is unknown but Woodward et al. (1989) point out that it must be considerably less than that of Na dimer. The lifetime of NaO is not known, but the radiative lifetime of the analogous LiO is > 600 ns (Pugh et al. 1996). Luiqiu et al. (1976) point out that the dominant emission from the Na + N$_2$O system arises from Na atoms with weak emission from Na$_2$ and the total photon yield for that reaction is < 0.1% at a total pressure of 2 torr. Since the photon yield of NaO* is very small, the emission is broad
covering the entire 700-900 nm region, and the lifetime is probably >600 ns, NaO does not appear to be a promising candidate.

BiF

The reaction of Bi vapor with F₂ produces chemiluminescence from BiF⁺ (Devore et al. 1991). Three emission bands are identified in the chemiluminescence spectrum: the BO⁺-XO⁺ band near 380 nm, the AO⁺-XO⁺ band near 440 nm, and "red bands" : 610-760 nm. Under multiple collision conditions, ≤ 2.5 torr, the emission from the AO⁺ clearly dominates by a factor > 20. Since one of the requirements for the infrared flashbulb is little or no emission in the visible region, BiF is not a likely initial candidate. No other information about the origin of the red bands is available.

SeF

SeF⁺ in the A²Σ⁺ state is generated by the reaction of Se (or H₂Se) with F₂. SeF⁺ emits "weak" chemiluminescence throughout the visible region from 380-890 nm (Thorpe et al. 1985). The A²Σ⁺-X²Σ⁺ system is off-diagonal and there are many emission bands diluting the strength of the transition. The radiative lifetime derived from Stern-Volmer analysis is long, 12.8 μs, again indicating that this transition is weak and precluding SeF as an initial candidate.

TeF

Chemiluminescence in the B-X (380-540 nm) and A-X (560-860 nm) systems of TeF was observed when Te or H₂Te reacted with F₂ (Thorpe et al. 1985; Newlin et al. 1981). The spectral emission from reaction of Te with F₂ is reported to be more intense than with the H₂Te/F₂ system but photon yields and rates of production are unknown. Analogous to SeF, the electronic transitions of TeF are assumed to be weak.

INITIAL CANDIDATES (600-700 nm)

BaO

Ba reacts with N₂O, O₂, and O₃ to produce BaO⁺ which emits a broad band with maximum intensity at ~630 nm and a FWHM of 100-150 nm (Eckstrom et al. 1975; Edelstein et al. 1977; Long et al. 1982). BaO⁺ chemiluminescence is discussed here since there is possible interest in a chemical system with emission in the region 600-700 nm. Experimental rate constants have been determined over the temperature range 250-850K for the reaction of Ba (¹S) with N₂O and O₂ (Nien and Plane 1991), while reaction cross sections have been obtained for all three oxidants. The formation rate constants are temperature-dependent and increase from 0.1 gas kinetic at room temperature to gas kinetic at T > 700 K.

For the reaction Ba + N₂O, the reaction cross section is 15.2 ± 0.71 Å² at 588K (Nien and Plane 1991) and the chemiluminescence cross section at P<1 mTorr is 2.23 Å² (Cox and Daghigian 1983). The fraction of product molecules in electronically excited states is estimated to be 3-18% (Lallement et al. 1993). For the reaction Ba + O₂, the reactive cross section is 23.6 ± 1.8 Å² (Nien and Plane 1991) at 1057 K and the total photon yield is 0.08 at 3 Torr total pressure with excess Ba but <0.001 with excess O₂ (Eckstrom et al. 1975). And finally, the reaction cross section for reaction of Ba with O₃ is ~70 Å² (Cox and Daghigian 1983) but the photon yield is unknown. These photon yields are similar to those found for the reactions of Ba with halogen-containing species.

The identity of the emitting states is the subject of some controversy and is important since the lifetime of the emitting state affects the irradiance of the system. For the reaction of Ba with O₂, Edelstein et al.
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(1977) identify the excited state as $A^1\Sigma^+$. Whereas, Long et al. (1982) suggest that the reaction $\text{Ba} + \text{N}_2\text{O}$ initially populates the $A^1\Sigma^+$ and $A^1\Pi$ states (as well as the $X$ state) which undergo intersystem crossing to the $a^3\Sigma^+$ state which then emits. For $\text{Ba} + \text{N}_2\text{O}$, Nien and Plane (1991) report that the consensus of numerous studies is that the nascent emitting states are $A^1\Sigma^+$ and $A^1\Pi$. The difficulty in characterizing the emitting states in these reactions arises because there are four excited electronic states within ~2000 cm$^{-1}$ of each other (Pruett and Zare 1975). Two of the states $A^1\Pi$ and $a^3\Pi$ have significantly larger average internuclear distances than the other states $A^1\Sigma^+$ and $a^3\Sigma^+$ (and the $X^1\Sigma^+$ state). Emission from the $\Pi$ states should show long Franck-Condon progressions. Since, the $A$ and $A'$ states have different radiative lifetimes, 350 ns and 9 $\mu$s, respectively, it should be possible to identify the emitter under pulsed conditions. Unfortunately, two studies carried out under molecular beam conditions aimed at identifying the emitting state led to opposite conclusions. The reaction of $\text{Ba} + \text{CO}_2$ is nearly thermoneutral (Edelstein et al. 1977) and Gottscho et al. (1978) note that this reaction yields little chemiluminescence. In spite of the confusion about the identity of the emitting states, $\text{BaO}^*$ appears to be a promising candidate for an infrared emitter in the region 600-700 nm.

CONCLUSIONS

A literature search of chemical systems which emit in the near-infrared region (700-900 nm) has generated several likely candidates. In particular, the barium halides, $\text{BaF}$ and $\text{BaCl}$ are the most promising candidates based on the ease of chemical production, the wavelength and bandwidth of the emission band, the lifetime of the emitting states, and the estimated photon yield. We will study these candidates first. Argon is also attractive since the emission from excited $\text{Ar}$ is centered in the near-infrared region, several intense lines dominate the spectrum, and these lines have strong transition probabilities. $\text{BaOH}$ is an additional candidate although the need for an electronically excited Ba reactant implies that the chemical production is not as simple as with the barium halides. $\text{BaO}$ is an attractive candidate for an emitter in the 600-700 nm region due to ease of chemical production, high photon yield, and possibly short radiative lifetime (if the emitter is the A state.)
<table>
<thead>
<tr>
<th>Species</th>
<th>Emission Wavelength (nm)</th>
<th>Band System</th>
<th>Chemical Method of Production</th>
<th>Rate of production</th>
<th>Integrated Photon Yield</th>
</tr>
</thead>
<tbody>
<tr>
<td>BaF</td>
<td>710</td>
<td>B-X</td>
<td>Ba + NF₃, F₂, SF₆</td>
<td>1 x 10^{13} photons/nm/s/Ba atom for Ba + F₂ at a total pressure of 5 torr (Bradford et al. 1975)</td>
<td>Ba + NF₃: 0.1, Ba + F₂: 0.004 (0.7&lt;P&lt;7 Torr total pressure) (Eckstrom et al. 1975)</td>
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<tr>
<td>BaF</td>
<td>810, 860</td>
<td>A-X</td>
<td>“</td>
<td>1.2 x 10^{14} photons/nm/s/Ba atom for Ba + F₂ at a total pressure of 5 torr (Bradford et al. 1975)</td>
<td>Ba + NF₃: 0.32, Ba + F₂: 0.012 (0.7&lt;P&lt;7 Torr total pressure) (Eckstrom et al. 1975)</td>
</tr>
<tr>
<td>BaCl</td>
<td>840</td>
<td>B-X</td>
<td>Ba + Cl₂</td>
<td>5 x 10^{13} photons/nm/s/Ba atom for Ba + Cl₂ at a total pressure of 3 torr (Bradford et al. 1975)</td>
<td>Ba + Cl₂: 0.04 (sum of A-X + B-X bands) (at 1 torr total pressure) (Bradford et al. 1975)</td>
</tr>
<tr>
<td>BaCl</td>
<td>910, 965</td>
<td>A-X</td>
<td>3 x 10^{14} photons/nm/s/Ba atom for Ba + Cl₂ at a total pressure of 3 torr (Bradford et al. 1975)</td>
<td>(see above)</td>
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<td>Ar</td>
<td>815, 845</td>
<td>4p-4s</td>
<td>dc arc lamp</td>
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<td>-</td>
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<tr>
<td>BaOH</td>
<td>774</td>
<td>B²Σ-X²Σ</td>
<td>Ba(¹P₁) + ROH</td>
<td>-</td>
<td>-</td>
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<tr>
<td>BaOH</td>
<td>831, 864</td>
<td>A²Π₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋₋…..</td>
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<td>Compound</td>
<td>Wavenumber (cm⁻¹)</td>
<td>Transition</td>
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<tr>
<td>TeF</td>
<td>560-860</td>
<td>$A^2Π_{3/2}^\rightarrow X^2Π_{1/2}$</td>
<td>$H_2Te, F_2$</td>
<td>$-$</td>
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<td>554-800</td>
<td>$A^2Π_{3/2}, A^2Π_{1/2}$</td>
<td>$H_2Te + F_2$</td>
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<td>BiF</td>
<td>610-760</td>
<td>“red bands”</td>
<td>Bi, F₂</td>
<td>$-$</td>
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<td>$A^2Π-X^2Π$</td>
<td>$H_2Se, F_2; Se + F_2$</td>
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<td>EuO</td>
<td>500-830</td>
<td>$Eu + O_3, N_2O^*$</td>
<td>$-$</td>
<td>Eu + N₂O: 0.2 (1-4 Torr total pressure) (Eckstrom et al. 1975)</td>
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<td>HNO</td>
<td>762</td>
<td>$A^1A^*-X^1A'$</td>
<td>$H + NO (+M)$</td>
<td>$\leq 2 \times 10^{10} l^2 mol^{-2} s^{-1}$ (Oka et al. 1977)</td>
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<tr>
<td>BaO</td>
<td>460-710</td>
<td>$A^1Σ^+ - X^1Σ^+$</td>
<td>$Ba + N_2O, O_2, O_3$</td>
<td>$k(Ba(S^1) + N_2O) = (2.99 \pm 0.14) \times 10^{-10} \exp[-(5.70 \pm 0.17) \text{kJ mol}^{-1}/\text{RT}] \text{cm}^3\text{molecule}^{-1}\text{s}^{-1}$; $k(Ba(S) + O_2) = (4.89 \pm 0.38) \times 10^{-10} \exp[-(7.06 \pm 0.23) \text{kJ mol}^{-1}/\text{RT}] \text{cm}^3\text{molecule}^{-1}\text{s}^{-1}$ (Nien and Plane 1991)</td>
<td>$Ba + N_2O: 0.25$ (4 torr total pressure) (Eckstrom et al. 1975) Ba + O₂: 0.08 (2.2 torr total pressure) (Eckstrom et al. 1975) Ba + N₂O: 0.3 (total photon yield at 1 torr total pressure) Edelstein et al. 1977)</td>
</tr>
</tbody>
</table>
REFERENCES


