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Generation and Properties of High Altitude Chemical Plasma Clouds

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GENERATION AND PROPERTIES OF HIGH ALTITUDE CHEMICAL PLASMA CLOUDS

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Abstract

Physical or chemical techniques can be used to form localized regions of high electron density in the upper atmosphere. One method is described in detail wherein a mixture of cesium nitrate, aluminum powder, and a high explosive is detonated in the 90 to 120 km altitude region of the upper atmosphere. The partially ionized reaction products expand to form a cloud from which radio waves can be reflected or scattered. In night releases the free electrons are generated in the explosion by thermal ionization; in day releases additional photoionization of cesium occurs by absorption of solar ultraviolet. The reflected radio signal duration and intensity are dependent on altitude, time of day, and wind shears. In addition to thermal ionization and photoionization, major factors in determining the usefulness of chemical plasma clouds for radio wave propagation are their size, structure. and orientation with respect to the transmission path.

Introduction

In this paper a special application of "ions in flames" is described, i.e., the study of partially-ionized reaction products released into the upper atmosphere from vertical probe rockets. One objective of these experiments has been to create localized high electron densities (so-called electron clouds) for study of over-horizon propagation of radio-frequency signals.

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Over-horizon radio-frequency propagation can be achieved in several ways: 1) Reflection and scattering can occur from the natural E and F layers of the ionosphere, which are the normal modes of shortwave broadcasting. The transmitted electromagnetic waves are "bounced" back and forth between the ionized layers of the atmosphere and the ground until they reach the receiver. The reflection capability of these layers is dependent, among other things, on the electron density of the reflecting region which averages about 105 electrons/cm3 at the 100-km altitude region called the E-layer and 106 electrons/cm3 at the 250-km altitude region called the F-layer.

The maximum radio frequency reflected at oblique incidence from a horizontal plasma sheet is given by

$$f = 0.009 \, n^{1/2} sec \, \emptyset$$
 (1)

where the frequency f is given in megacycles, n is the number of electrons per cm³ in the plasma sheet, and \emptyset is the angle of incidence measured from the vertical. The maximum attainable useful incidence angles from the vertical (in the F-layer) are about 70 to 75°, permitting reflection of frequencies in the HF range, i.e., 3 to 30 Mc.

- 2) Reflection of higher frequencies than those reflected by the normal E and F layers is possible from naturally occurring passive reflectors of higher electron content than the ionosphere, such as meteor trails, formed by ionization of ambient air heated by meteor passage in the 90 to 110 km region. These trails attain a radar cross-section of 10³ to 10⁴ m² for frequencies between 30 and 100 Mc. The major disadvantage of meteor trails as reflectors is, of course, their unpredictable occurrence. Reflection is obtained for a few seconds per trail, at a rate of one trail per minute, permitting over-the-horizon propagation for several minutes per hour.
- 3) Reflection can be achieved by solid-surfaced passive reflectors, such as satellites, orbiting balloons, and needles. Satellites and balloons are reflectors for wavelengths smaller than their radii and their small reflecting area requires powerful transmitters and receivers. Needles are even more selective in the transmittable frequencies. Active transponders such as the Telstar and Relay satellites receive at a preselected frequency which is amplified and retransmitted.
- 4) Artificial gaseous plasmas are generated most effectively in a relatively narrow altitude region extending between 90 and 120 km. Below this region the high collision frequencies increase the rate of electron loss mechanisms; above this

altitude the increased diffusion rates cause a rapid dissipation of released gaseous material. Within the proper altitude region, however, the following methods are being investigated presently for plasma generation: a) striking of an electric arc from rocket-borne battery cells; b) cyclotron resonance heating of a region in the ionosphere by ground-based or rocket-borne high powered transmitters of the proper frequency; c) utilization of solar and chemical energy to photoionize and thermally ionize chemicals dispersed at the proper altitude. It is this last type of plasma generation which will be discussed in more detail with regard to ionization efficiency and propagation characteristics.

Chemistry of Electron Clouds

The first successful electron cloud was formed in 1956 by release of 8 kg (275 moles) of nitric oxide at 95-km altitude in sunlight.3 Nitric oxide, having an ionization potential of 9.25 ev, is photoionized readily by the solar Lyman -Q , and a region of increased electron density was detected by radar echoes at the proper altitude for about 10 min. following release. In subsequent experiments in itric oxide was replaced by potassium and cesium, the ionization potentials of which are 4.32 and 3.86 ev, respectively. The nitrate salt of the desired alkali metal was mixed with aluminum powder in a ratio of 2.66 moles Al to 1 mole MNO3, the total weight being about 25 kg. The mixture was packed into a canister equipped with a timer-initiated igniter, set to ignite the mixture when the rocket attained an altitude of about 100 km. Upon ignition the mixture reacted, bursting the canister and expanding into a fireball composed mainly of alkali vapor, nitrogen, and condensed Al₂0₃. At the high explosion temperature some of the alkali vapor was ionized, thus augmenting the solar photoionization with initial thermal ionization. In a series of day and night-time releases distinct radar echoes were obtained both on backscatter and on forward scatter circuits.5 The nighttime effect was of shorter duration than the daytime effect, due to recombination and attachment processes, without photoionization. Ground tests indicated that the alkali nitrate-aluminum reaction is deflagrative, i.e., on heating, the mixture reacts without detonation. In a closed vessel the reaction proceeds with pressure build-up until the vessel ruptures which scatters and extinguishes the unreacted portion of the mixture. Since the static burst pressure of the canister used was about 250 - 500 atm, the reaction proceeded until this pressure was attained. The void volume in the canister was such that only 3% of the above composition reacted before vessel rupture, which resulted in a low chemical yield. The estimated flame temperature of the reaction

products at 500 atm is 4900° K, at which condition potassium is about 1% ionized. However, substantial recombination, by as much as three orders of magnitude, can occur during the expansion process.

More recent experiments by Rosenberg and co-workers. 7,3 improved the chemical yield to near 100% in detonation of 18kg mixtures formed by adding a high explosive, such as RDX or TNT, to the cesium nitrate and aluminum. In such mixtures the equilibrium electron yield is relatively low due to the high pressure of the reaction zone and the presence of electron attaching species such as OH. Furthermore, it is agreed generally that aluminum does not react fully in the detonation zone, that is, considerable reaction occurs during expansion of the explosion products. Therefore, it is extremely difficult to estimate the number of electrons generated and decayed in the process of detonation and expansion of the above mixture. Radar returns obtained at a number of frequencies from high altitude releases immediately after the initial expansion is at present the best way to assess the thermal ionization yield of such reactions. Such assessments place the degree of ionization of the gaseous products after expansion in the order of 10⁻⁴. Since cesium is 6% of the gaseous products, the degree of ionization of cesium is about 1.6×10^{-3} .

The major reaction products from the RDX-CsN0₃-A1 explosion are CO, H₂, N₂, and Cs vapor, and condensed A1₂0₃. Altogether about 430 moles, or 2.5 x 10²⁶ gaseous molecules, are liberated from the 18-kg payload. These gases expand to form a cloud of 100 - 1000 m diam, depending on altitude. Optical observations show that the clouds are initially spherical, and calculations may be based on cloud models where it is assumed that the clouds have a "square-weil" density distribution, or, alternately, a Gaussian density distribution, with the tails of the Gaussian being overlapped by the ambient which was pushed aside by the expanding gases (Fig. 1). After completion of the (rapid) expansion, the total number density is assumed as a first approximation to be constant throughout the region and equal to the ambient number density. The radius of a spherical "square-weil" density cloud is given by

$$r = (3N/4\pi n_a)^{1/3}$$
 (2a)

where N is the total number of gaseous molecules released and na is the ambient number density at the release altitude. For a Gaussian cloud, the equivalent parameter is the Gaussian half width, i.e., the radial distance from the center where the density falls to 1/e of the center point density.

The Gaussian half width, h, is given by

$$h = (N/n_a)^{1/3}/\pi^{1/2}$$
 (2b)

Thus, r = 1.1h, i.e., these parameters differ only by 10%, and for calculation of diffusion times it is quite immaterial which model one assigns to the density distribution. In Table I the Gaussian half widths are calculated as a function of altitude based upon ambient density values of the CIRA standard atmosphere.9

Table 1

Gaussian half-width, h, for a cloud containing
2.5 x 10²⁰ gaseous molecules as a function of altitude

Altitude (km)	n _a (particles/cm ³)	h (cm)	
80	4.03 × 10 ¹⁴	5.0 × 10 ³	
100	9.98 x 10 ¹²	1.65 × 10 ⁴	
120	5.15 × 10 ¹¹	4.5 × 10 ⁴	
140	6.55 x 10 ¹⁰	9.0 × 10 ⁴	

Based on these parameters, the optimum altitude region shall be estimated for the deposition of reasonably long-lived r.f. reflecting electron densities. The upper altitude limit is determined by rapid diffusional loss. An electron cloud becomes "invisible" to radio frequency waves of 10 Mc when the number of electrons per cubic centimeter drops below 10⁶ at its center. The diffusion time of a spherical Gaussian cloud is given by 10

$$t_u = h^2(u^2/3 - 1) / 4D_a$$
 (3)

where t_u is the time in which the center point density reaches a value 1/u of its initial value and D_a is the ambipolar diffusion coefficient at a given altitude. The time required to fall from a center point density of electrons $n_e(0) = 10^{-4} n_a$ to $n_e(t_u) = 10^{6} cm^{-3}$ where 10^{-4} is the

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assumed degree of ionization, is set forth in Table 2 for various altitudes. From Table 2 it is seen that the life time of the cloud at 140 km altitude is 40 sec and this height is probably the upper limit for reasonable life. In practice it was found that this altitude is closer to 120 km.

Table 2

Initial center point electron densities and the times necessary to reduce these to 106/cm3 by ambipolar diffusion

Altitude (km)	n _e (0)=10 ⁻⁴ n _a (electrons/cm ³)	D _a (cm²/sec)	u	t _u (sec)
80	4 × 10 ¹⁰	4.8 × 10 ³	4 × 10 ⁴	1.5 × 10 ⁶
100	1 × 10 ⁹	2.2×10^5	1×10^{3}	31000
120	5 × 10 ⁷	6.9 x 10 ⁶	50	920
140	6.5×10^6	1.3 × 10 ⁸	6.5	40

Optical observations show that clouds deposited below 100 to 110 km usually grow by eddy diffusion (Fig. 2) with turbulent diffusion rates higher by as much as two orders of magnitude than molecular diffusion rates. Therefore, the diffusion times for 80 and 100 km in Table 2 are perhaps overestimated by a factor of 10 - 100.

The lowest useful altitude is determined by losses due to recombination of electrons with positive ions and attachment of electrons to neutrals. Electron densities above 10^{9}cm^{-3} cannot be maintained for considerable time periods, because a Bates, Kingston, and McWhirter type collisional-radiative recombination 11 with an effective rate coefficient of $10^{-9} \text{cm}^{3} \text{sec}^{-1}$ will take about 10 sec to decrease the electron density from 10^{10} to 10^{9}cm^{-3} . When the electron density is in the order of 10^{8}cm^{-3} , the predominant loss mechanism at the lower altitudes is probably a three body attachment to the explosion products and to molecular exygen diffusing into the cloud by molecular and turbulent diffusion. The rate coefficient for this process is about $5 \times 10^{-30} \text{cm}^{-6} \text{sec}^{-1}$. 12 With a density of the attaching molecule and the third body of 10^{10}cm^{-3} at 80 km, the time to decrease the electron density from 10^{8} to 10^{6}cm^{-3} is about 6 sec; at 90 km $(n_{a} = 6 \times 10^{12} \text{cm}^{-3})$, the time is 250 sec. This places the

lower altitude limit at about 90 km, in agreement with experience. Thus, the 90- to 120-km altitude region is where long-lived electron clouds can be generated. In defining the optimum altitude region the effect of photoionization was not considered. However, photoionization can result in the prolongation of electron cloud life time within the optimum altitude region but will not extend this region, since the arguments about diffusion and recombination losses are not altered.

It appears that improvement of the initial degree of ionization of the chemical reaction is not likely to increase the life time of electron clouds, because of rapid recombination losses during the early times when high electron densities are present. However, the size and structure of the cloud, which are dependent on the quantity of released material, the altitude of release, and atmospheric conditions (wind, turbulence, geomagnetic field, etc.) can affect the life time markedly, as discussed in the following section.

Propagation Characteristics of Electron Clouds

The mode of r.f. signal return from ionized regions is a complex phenomenon. The return at a given frequency can be evaluated empirically in terms of its intensity, polarization, phase delay, frequency spread and shift, and angular dependence. The scattering mechanism can only be inferred from such measurements and by construction of simplified theoretical models. For the point electron clouds three major modes of signal return seem to be operative. 13,14

- 1) Perfect or partial reflection from a wholly overdense volume. In this case the received signal cross-section is proportional to the physical area of the reflector, the electron density is given by Eq. 1, and no short period fading of the signal is obtained.
- 2) Reflective scattering from a large number of overdense irregularities. In this case the received signal cross-section is proportional to the number of irregularities and their scale and considerable snort paried fading is observed due to interference.
- 3) Booker-Gordon type scattering from underdense irregularities. ¹⁵ For this mode the scattered signal cross-section is proportional to $(\overline{\Delta n})^2$, the mean square departure of electron density from the mean, and the scale size of the (turbulent) irregularities. This mode will also result in signal fading.

Analysis of tens of electron cloud releases with regard to signal strength, duration, fading, and angular dependence indicates that for electron clouds in the 100-km altitude region all three modes are operative simultaneously, with the contribution by each mode depending on the amount of turbulence, self-induced as well as atmospheric. After tens of minutes in suntit releases and several minutes in night releases, the overdense reflectors decay and only the third mode remains significant.

Life Time of Electron Clouds

Figure 3 represents the life time of several electron clouds as a function of altituce and beam incidence angle for signals in the lower end of the VHF band.

Sunlit clouds observed with an incidence angle $\emptyset = 81.5^{\circ}$ (cf. Eq. 1) are denoted by circles. It is seen that clouds of considerable life times are obtained within the 90- to 120-km region, where signals are propagated for 100 - 3000 sec. The optimum altitude appears to be in the 100- to 110-km region. The 103-km cloud may have been the best propagation cloud because it was deposited in the optimum altitude region, and also because it was dragged out by wind shears to large horizontal dimensions, as observed by optical tracking. Therefore it seems that the size and orientation toward the transmission path are major factors in determining the life time of electron clouds as propagation links. Since wind velocities and directions may vary quite rapidly as a function of altitude. the optimum altitude for electron clouds can be expected to shift within the 100- to 110-km region, and it should not be inferred from Fig. 3 that 103 km is always the best altitude. The size of the cloud is dependent also on the amount of released gases. From recent, not yet fully analyzed experiments, it was confirmed that 4 kg charges do result in shorter lived electron clouds with smaller radio cross-section than the 18 kg charges represented in Fig. 3.

Night clouds (squares in Fig. 3) last 3 to 10 times less for a given frequency and incidence angle than sunlit clouds, depending on altitude of release. Thus, useful propagation links of several minutes duration can be maintained with night releases, where photoionization does not play a role.

The life times denoted by triangles in Fig. 3 were measured at a more obtuse incidence angle than those denoted by circles and are shorter by an order of magnitude. For the overdense reflection mode this is a consequence of Eq. (1), whereby a wave of a given frequency is reflected from lower electron

densities at greater incidence angles to the normal. For instance, a 30 Mc signal is reflected from a plasma sheet containing 2 x 10^5 electrons per cm³ at $\beta = 81.5^{\circ}$, whereas 1.5×10^6 electrons per cm³ are necessary for $\beta = 66.5^{\circ}$. By the same token, electron cloud life times for given incidence angle are inversely proportional to frequency.

Cloud Cross Sections

For the reflective cloud model, the radio cross-section at a qiven frequency is dependent on the effective reflection area of the overdense regions and on the scattering angle. For underdense scatter the cross-section is dependent on the mean square departure of electron density from the mean and on the scale size of turbulent eddies in the cloud. All these variables are strongly time dependent, since the cloud is in a continuous dynamic condition (expansion, diffusion, turbulence) and the electron inventory is changing by dilution, recombination, attachment and photoionization. Figure 4 gives the time function of cross-sections at about 30 Mc and $\beta = 81.5^{\circ}$ averaged over one minute intervals. The radar cross-sections display a similar altitude dependence as do overall signal durations: the 103 km cloud attained in general the largest cross-sections, at lower and higher altitudes the averaged cross-sections decrease. It is seen that radar cross-sections in the order of 10^6 - $10^8 \mathrm{m}^2$ are obtained readily for tens of minutes, but with significant fading and enhancement, due to interference and reinforcement of multipath signals.

Since the signals display a considerable short term fading. no clear-cut cross-section can be assigned to the clouds which would allow the assessment of their actual physical area (or volume) at a given time. Thus, it is difficult to use radar cross-sections from turbulent electron clouds to evaluate absolute electron densities as a function of time and space and to obtain the associated diffusion, recombination and attachment coefficients. This, of course, does not affect the possible use of artificial electron clouds as propagation links with properly designed systems. The tracking by radar of the gross movement of the clouds without knowing their intricate structure also gives important information on atmospheric winds and wind shears. Any acceptable cloud model which can be used to correlate the observed behavior of artificial electron clouds will also be of significant value in interpretation of radio frequency returns by natural events such as sporadic-E and meteor trails.

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Fig. 1 Model of density profile of released gases and ambient

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The standard cloud in the 100-km altitude region, 200 and offer burds; sunlit against dark sky background (suffight conditions); note turbulent structure; total dismeter 3 - 4 km

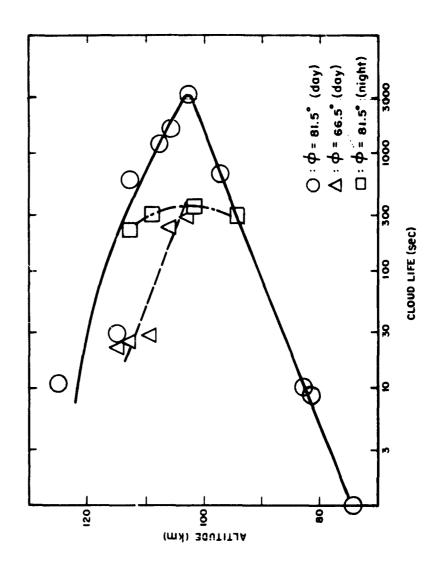


Fig. 3 Cloud life time vs altitude

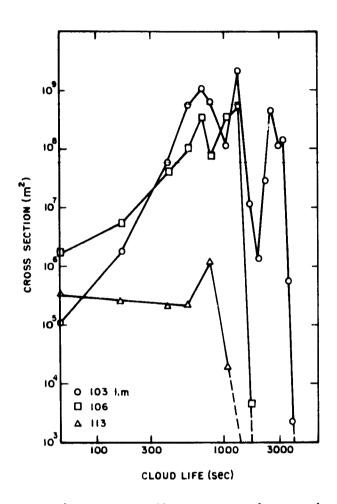


Fig. 4 Cloud radio cross-section vs time

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