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#### ELECTRICAL CONDUCTIVITY OF INERT GASES—SEED COMBINATION IN SHOCK TUBES

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#### ELECTRICAL CONDUCTIVITY OF INERT GASES—SEED COMBINATION IN SHOCK TUBES\*

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#### ABSTRACT

The effect of seeding shock waves with alkali metal smoke is studied. The pressure, density, temperature ratio, electron density, and conductivity were calculated with a digital computer and tabulated for various combinations of gas and seeding material. A hydrogen-driven shock tube was constructed to measure the properties of the seeded gas behind the shock wave. The conductivity of argon seeded with cesium was measured with a conductivity probe and compared with theoretical calculations.

#### 1. INTRODUCTION

The interaction of ionized gas flow with a magnetic field has been known for some time and considerable effort has been made to investigate and utilize this phenomenon. The effect of magneto gas dynamics has been applied to power generation, nuclear fusion, and space propulsion. The magnitude of all such interactions depends on the electrical conductivity of the gas.

One method of studying this interaction is to use the high temperature and hence ionized gas behind a shock wave in a shock tube. However, all gases of engineering importance have comparatively high ionization potentials and hence only small conductivities can be obtained easily. One way to increase the interaction is to increase the conductivity by an appropriate rise in the shock Mach number. This, however, reduces the interaction time and creates experimental problems. An alternative way is to increase the conductivity by introducing small quantities of easily ionizable material into the gas. The alkali metals or alkali earth groups have low ionization potentials and are, therefore, suitable as seeding material. The seeding materials are easily ionized in the hot gas behind the shock wave and contribute a large number of electrons, thus increasing the conductivity.

In the present report, the effect of seeding shock waves in the inert gases argon and helium with alkali metals potassium and cesium was investigated. The inert gases were chosen to avoid the complexity of the dissociation problem.

#### 2. SEEDED SHOCK WAVES

The gas properties behind a moving normal shock wave in various gases have been calculated in a number of previous investigations (Glass 1958). For a monatomic gas which is seeded with small quantities of alkali metals, the analysis is quite similar. As a simplification, the ratio of specific heats,  $\gamma$ , is taken to be 5/3 for both the gas and the alkaline metal vapor. This is justified since the excitation energy is only a small portion of the total enthalpy in the present range of work.

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We define the degree of seeding by

(1) 
$$c = \frac{n_{\rm s} + n_{\rm s}^+}{n_{\rm g} + n_{\rm g}^+} = \frac{\rho_{\rm s}}{\rho_{\rm g}} \cdot \frac{M_{\rm g}}{M_{\rm s}},$$

where n is the number density, M is the molecular weight, g denotes the gas and s the seed material. Assuming an equilibrium state (2) behind and a quiescent state (1) in front of the shock, the properties of regions (1) and (2) are related by the conservation of mass, momentum, energy, the Saha equation, and the equation of state:

(2) 
$$P_{21} = \frac{p_2}{p_1} = 1 + \gamma M_a^2 \left(1 + \frac{cM_s}{M_g}\right) \left(1 - \frac{\rho_{g_1}}{\rho_{g_2}}\right),$$

(3) 
$$H_2 + \frac{1}{2}(w - u_2)^2 = H_1 + \frac{1}{2}w^2$$
,

where

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(4) 
$$H_2 = \frac{5}{2} \left(1 + \alpha_g\right) \frac{R}{M_g} T_2 + \alpha_g \frac{q_g}{M_g} + \alpha_g \cdot \frac{q_g}{M_g} \cdot c + \frac{5}{2} \left(1 + \alpha_g\right) \frac{R \cdot c}{M_g} T_2$$

(5) 
$$\frac{p_2}{kT_2} = n_e + n_s^+ + n_s + n_g^+ + n_g,$$

(6) 
$$(\rho_{g} + \rho_{s})_{2} = (n_{g} + n_{g}) \frac{M_{g}}{N} + (n_{s} + n_{s}^{+}) \frac{M_{s}}{N},$$

(7) 
$$n_{e} = n_{s}^{+} + n_{g}^{+},$$
  
(8)  $\frac{n_{e}n_{s}^{+}}{n_{\pi}} = \frac{(2\pi m)^{3/2}}{h^{3}} (kT_{2})^{3/2} \frac{Z_{1g}Z_{e}}{Z_{\pi}} \cdot e^{-q_{g}/kT_{2}}$ 

(9) 
$$\frac{n_{\rm e}n_{\rm s}^{+}}{n_{\rm s}} = \frac{(2\pi m)^{3/2}}{h^{3}} (kT_2)^{3/2} \frac{Z_{\rm is}Z_{\rm e}}{Z_{\rm s}} e^{-q_{\rm s}/kT_2}.$$

The above equations have been solved simultaneously for  $P_{21}$ ,  $\rho_{21}$ ,  $T_{21}$ , and  $n_e$  on a computer for s = c.  $M_e/M_g = 0.0002,0.002$ , and 0.02. The pressure, density, and temperature differ only very slightly between the seeded and the unseeded case. The electron density  $n_e$ , however, is increased appreciably and is plotted in Fig. 1.

#### 3. ELECTRICAL CONDUCTIVITY BEHIND SHOCK WAVES

The electrical conductivity of shock-heated argon has been measured experimentally and computed theoretically by Lin *et al.* (1955). There the reciprocal of the conductivities due to close encounters (between electron and neutrals) and distant encounters (between electron and ions) was added to give the reciprocal of the total conductivity. The average collision cross section was used instead of correctly using the total collision frequency

$$\nu_{\text{total}}(v) = \nu_{el}(v) + \nu_m(v)$$

and electron-electron collisions are neglected. To obtain a better approximation, the analysis of Shkarofsky (1961) was used. In this reference, the

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FIG. 1. Electron density of shock waves in seeded argon and helium.

collision frequencies between electrons and neutrals were assumed to vary as a simple power r of the electron velocity. The tensor conductivity including all interactions is given by

(10) 
$$\sigma = \begin{pmatrix} b+c & j(b-c) & 0\\ -j(b-c) & b+c & 0\\ 0 & 0 & d \end{pmatrix},$$

where

$$b = \frac{n_e e^2}{2m} \frac{1}{(\langle \nu_m \rangle + \langle \nu_{el} \rangle)g + j(\omega - \omega_b)h},$$
  

$$c = \frac{n_e e^2}{2m} \frac{1}{(\langle \nu_m \rangle + \langle \nu_{el} \rangle)g + j(\omega + \omega_b)h},$$
  

$$d = \frac{n_e e^2}{m} \frac{1}{(\langle \nu_m \rangle + \langle \nu_{el} \rangle)g + j\omega h},$$
  

$$\langle \nu_m \rangle = -\frac{4\pi}{3n_e} \int \frac{\partial f_0^0}{\partial v} v^3 \nu_m dv.$$

 $f_{0}^{0} =$  Maxwellian distribution function of electrons and g and h are functions of  $\omega_{b}/(\langle \nu_{m} \rangle + \langle \nu_{el} \rangle)$ ,  $\langle \nu_{m} \rangle / \langle \nu_{el} \rangle$ , and Z.

$$\langle \boldsymbol{\nu}_{e1} \rangle = \frac{4(2\pi)^{1/2} n_e \ln \Lambda}{3m^{1/2} (kT)^{3/2}} \left( \frac{Z_e^2}{4\pi\epsilon_0} \right)^2,$$

$$\Lambda = \frac{3}{2Z_e^3} \frac{(4\pi\epsilon_0 kT)^{3/2}}{(\pi n_e)^{1/2}}.$$

Z = charge number = 1 in the present case.

The values of g and h were computed and plotted as functions of  $(\omega \pm \omega_b)/(\langle \nu_m \rangle + \langle \nu_{e1} \rangle)$  and  $\langle \nu_m \rangle/\langle \nu_{e1} \rangle$  in Shkarofsky (1961) for values of r = -3, -2, -1, 0, 1, 2, 3. A review of the literature on momentum transfer cross sections gives the approximate dependence of the collision frequencies between neutrals and electrons on the electron energy (Frost 1961). The collision frequencies for the various seed and gas materials used in this report are given below. For the case of argon, the expression is derived from a curve of the momentum transfer cross section obtained by Frost and Phelps.

He 
$$\nu_m/N = 3.14 \ u^{1/2}$$
  
K  $\nu_m/N = 160$   
Cs  $\nu_m/N = 160$   
A  $Q = 9.612 \times 10^{-17} \ u^{1.35}(0.3u \times 10 \ eV)$   
 $N =$  number of neutral atoms.

In the seeded gas behind the shock, therefore, the collision frequency between electron and neutrals is

(11) 
$$\nu_m = A + BvC.$$

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Since the results presented in Shkarofsky are based on the power law relation  $\nu_m = \text{constant} \times v^r$  (r = -3, -2, -1, 0, 1, 2, 3), it is necessary to approximate the collision frequency by a similar expression in order to use the tabulated results in g and h. Assuming that the relation is

(12) 
$$\nu_m = K v^r,$$

the values of the constants K and r can be obtained in terms of A, B, C, and  $v_0$ 

if we take only small variations of v from the center point  $v_0$ .  $v_0$  is then the electron velocity corresponding to the temperature at which we want to compute the conductivity. Equating equations (11) and (12), and assuming  $v = v_0(1 + \xi)$  where  $\xi \ll 1$ ,  $v_m$  is approximated by the equation:

 $\nu_m = (A + Bv_0C)\left(\frac{v}{v_0}\right)BCv_0C/(A + Bv_0C),$ 

and

$$\langle \nu_m \rangle = -\frac{4\pi}{3n_e} \int_0^\infty \frac{\partial f_0^0}{\partial v} v^3 \nu_m \, \mathrm{d}v$$

$$= \frac{k\Gamma(2.5 + \frac{1}{2}r)}{\Gamma(2.5)} \left(\frac{2kT}{m}\right)^{r/2}.$$

Using the above equations,  $\langle \nu_{e1} \rangle$  and  $\langle \nu_m \rangle$  were calculated for each Mach number and gas-seed combination on a digital computer. The value of  $v_0$  depends on the equilibrium temperature and is calculated as follows. The d-c. conductivity without magnetic field and considering neutral scattering is given by

$$\sigma = -\frac{4\pi e^2}{3m} \int_0^\infty \frac{1}{\nu_m} v^3 \frac{\partial f_0}{\partial v} dv.$$

To find the power law for  $\nu_m$  that would best suit the gas and seed mixture, the center point  $v_0$  is taken to be at the maximum of the integrand  $(v^3(\partial f_0/\partial v))$ . Differentiating the integrand and setting it equal to zero, and assuming that  $f_0$  is Maxwellian,

$$4v^{3}\mathrm{e}^{-mv^{2}/2kT}-\frac{m}{kT}v^{5}\,\mathrm{e}^{-mv^{2}/2kT}=0,$$

from which  $v_0^2 = 4kT/m$ . The values of g were interpolated from the results in (2) and  $\sigma$  is calculated by the following equation:

$$\sigma = \frac{n_{\rm e}c^2}{m} \frac{1}{g(\langle v_m \rangle + \langle v_{\rm el} \rangle)} \,.$$

The effect of a large magnetic field can be incorporated without difficulty. The values of g and h can be interpolated as above and the tensor conductivity calculated according to equation (10).

#### 4. EXPERIMENTAL APPARATUS

The shock tube used in this investigation consists of a cylindrical driven section about 30 feet long and a driver section about 6 feet long. Both sections have inside diameters of  $2\frac{1}{2}$  inches. The test section is a cylindrical pyrex pipe about 2 feet long with inside diameter of  $1\frac{1}{2}$  inches. Ahead of the pyrex pipe is a 4-foot,  $1\frac{1}{2}$ -inch inside diameter steel section which can be inserted into the driven section of the shock tube. This has the effect of discarding the turbulent boundary layer induced behind the shock, together with any displarage fragments or impurities that might have been swept from the shock tabe well. The insert prevents the reflected shock wave from disturbing the flow into the

test section. A dump tank is connected to the end of the test section to relieve the pressure and prevent the formation of a reflected shock. A metal plunger activated by compressed air was designed to break the diaphragm. Several types of metal diaphragms were tested and copper diaphragms up to 0.032 inch thick were found to be best since they petal nicely and no fragments were broken off. The experimental setup is shown in Fig. 2.

With driver pressure up to 1000 p.s.i. hydrogen and channel pressure from 2.5 to 20 mm Hg, shock Mach numbers from 6 to 10.5 were obtained in the test section. Nitrogen was used to flush out the hydrogen after each run.

The conductivity of the hot gas behind the shock was measured with the conductivity probe, the theory of which is described in detail by Savic (1962), and only a brief description is given here. The probe consists of a 6-inch-long pyrex tube which fits over the test section, and has six coats of platinum paint baked on the inside to eliminate the capacitive coupling effect of any substance put inside the tube. Two turns of copper wire were wound on the middle of the tube, constituting part of the inductance of a tuned circuit. This inductance changes slightly when the hot conducting gas behind the shock passes through the test section, and appears as a frequency change in the tuned circuit. It is then fed into a discriminator circuit which transforms it into a d-c. voltage. Mercury and sulphuric acid were used for its calibration since there are no substances within the right range of conductivity except for fused salts and semiconductors. For this reason, considerable difficulty arises in obtaining a reliable calibration and the accuracy of the probe is limited to about 15%.

#### 5. SEEDING OF THE SHOCK TUBE

There are two different methods of seeding the shock tube. The first one is to inject the vapor of the alkali metals into the gas. This is the ideal way of seeding since a genuine gas-seed mixture can be obtained. However, this necessitates heating the shock tube (at least the driven section) up to a temperature where the vapor pressure of the alkali metal is sufficiently high. An oven would have to be constructed to enclose the whole shock tube and a predetermined amount of alkali metal introduced inside the tube. This raises problems involving measurements of the shock properties, especially optical measurements and material problems, since alkali metal vapor at this temperature is very corrosive.

An alternative method of seeding the shock tube is by introducing a cloud of small particles of alkali compound into the shock tube. This can be done by blowing the gas through a finely divided powder of the seeding material (Basu 1960) or bubbling it through the molten material and allowing the vapor to cool. However, the above methods do not produce a cloud of fine uniform-size particles. A slight improvement is obtained by heating the cesium chloride to about 1000° C and letting a jet of cool argon blow onto the vapor. The compound sublimes readily at this temperature and above the molten cesium chloride there is formed a region of high concentration of small crystalline particles. The argon jet blowing on the surface then carries off these particles, which are suspended in the form of smoke. A sample of this smoke was deposited on a small carbon-coated grid and examined under an electron



FIG. 2. Experimental setup.

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microscope. It was seen to consist of minute particles having linear dimensions of about 500 Å, together with large particles which are aggregates of a few tens or even hundreds of the basic crystals. A few plates with various magnification are shown in Fig. 5.

The shock tube and the seeding apparatus are evacuated initially. The CsCl in a quartz container is then heated up to about 1000° C. A jet of argon is directed onto the surface of the molten compound and a predetermined amount of the smoke is let off from the top of the quartz container into a cylindrical chamber which is separated from the shock tube by a butterfly valve. A sample bottle with two electrodes is connected to the cylindrical chamber for measuring the amount of seeding. After the required amount of smoke has been fed into the chamber, the butterfly valve is flipped open and the smoke fills the shock tube up to the desired pressure. The shock tube is then fired immediately to avoid settling of the smoke particles on the shock-tube walls.

The degree of seeding is measured by adding water to the sampling bottle and measuring the conductivity of the solute between the electrodes.

The disadvantages of seeding with solid smoke particles of a compound are:

1. The seeding material has to be vaporized and dissociated before yielding electrons. However, in the present range of work, the energy required to vaporize and dissociate the alkaline compound is but a few percent of the total enthalpy of the hot gas, so that the effect on the final conductivity is very slight.

2. The affinity of the nonmetallic radical on electrons leads to a reduction of the electron density. However, an estimate shows that the reduction of electron density by the nonmetallic radical is only appreciable at temperatures below 5000° K.

#### 6. RESULTS

The theoretical conductivities of seeded inert gases are plotted in Figs. 3 and 4. The first is a comparison between Lin's method and that of Shkarofsky. The curves tend to converge at both ends and the discrepancy is worse at the intermediate range. This is expected since Lin's analysis sums the resistivity from electron-neutral and electron-ion encounters which are accurate at the extreme ends of slightly ionized and fully ionized gases respectively. At both ends, one of the two is negligible, so that the discrepancy is slight; but at the intermediate range, the two types of encounter cannot be summed as in Lin's analysis since their velocity dependence is vastly different. For example, in the case of argon, the collision frequency is proportional to  $v^3$  for the electron-neutral encounters and to  $v^{-3}$  (or  $v^{-2}$  in Frost, taking into account the electron-electron interactions) for the electron-ion encounters.

From the theoretical curves, it can be seen that the seeded shocks have approximately constant conductivity at lower shock speeds, and the conductivity rises to meet the unseeded case at higher speeds. This is because at lower  $M_{a}$ , most of the conductivity is contributed by the seeding material, which is practically all ionized above  $M_{a} = 6$ , whereas the gas is only slightly ionized. As  $M_{a}$  increases, however, the gas becomes more and more ionized until the percentage ionization exceeds the percentage seeding, and from then on the contribution from the gas predominates.



FIG. 3. Conductivity of pure argon.

The experimental results are also plotted in Figs. 4(a) and 4(b). The amount of seeding was approximately 2% since the actual amount inside the shock tube cannot be measured. However, variation of the amount of seeding slightly above or below 2%, does not produce appreciable difference in the measured conductivity.

The measured conductivity agrees fairly well with the theoretical value for the unseeded case, whereas for the seeded shocks, the measured values are too small by approximately half. This may be due to a number of factors. Streak



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FIG. 5. Magnified pictures of deposited smoke particles.







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photographs by the method of Petschek and Byron (1957) show that while the relaxation time ranges from about 100 to 400 microseconds for the unseeded shocks, it was very small for the seeded ones (Fig. 6). This, together with the steep rise in the conductivity trace (Fig. 7), suggests that the low measured conductivity is probably not due to nonequilibrium.







(b)  $p_1 = 10 \text{ mm} \text{ Hg} (\text{SEEDED})$   $M_0 = 7.4$  $\sigma = 5.0 \text{ MHO/CM}$ 

FIG. 7. Conductivity oscillograms.

One probable cause of the low observed conductivity is the particle-size distribution. The largest aggregates have a settling rate of about 1.5 cm/sec compared with about  $3 \times 10^{-3} \text{ cm/sec}$  for the small particles, so that at the bottom of the shock tube there is a high concentration of large aggregates. Even though the estimated time required to vaporize and dissociate the small particles is of the order of microseconds, the time elapsed before the large aggregates can contribute to the conductivity might be much larger, especially when the largest ones lie in the cool boundary layer. A few shots were taken with deliberate time delay between seeding and shooting, and the observed seeded conductivity is even lower than that of the unseeded case. This shows that the additional energy required to vaporize and dissociate the rich aggregate of particles at the bottom of the tube tends to cool down the boundary layer even further.

The diffusion time required for the hot seeded gas to form a homogeneous conducting fluid, assuming ambipolar diffusion, ranges from a fraction to about 50 microseconds for particle separation of 100 and 1000 microns respectively, so that this effect is unlikely to cause lower observed conductivities. However, since the smoke particles can only be examined after being deposited on fine grid, there is no way of knowing for certain what the separation is when the particles are still suspended in the argon, especially when they tend to aggregate on settling.

#### 7. CONCLUSION

The theoretical conductivity of argon seeded with solid smoke particles was not fully attained. Because of the uncertainties of using solid particles as discussed in Section 6, it is not known which is the most important factor in the discrepancy between the theoretical and measured conductivities. The boundary layer, the solid particle distribution, and their attainment of equilibrium ionization can be investigated with optical methods such as schlieren and spectroscopy to give a clearer understanding of the ionization process behind the shock wave. However, conductivities of argon seeded with alkaline salt will be measured shortly in a plasma jet, where there are no boundary-layer and solid-particle problems since the salt is injected as a vapor. These results will then confirm whether or not the low conductivity is due to the effects mentioned above.

In view of the present experimental results, it can be concluded that even though particulate seeding in shock tubes does not give the desired conductivity, it does raise the conductivity appreciably at low Mach numbers. The main advantage is the simplicity of adding the seed material, and all the inconveniences of heating the shock tube in an oven are avoided.

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