Measurement of breakdown potentials and Townsend ionization coefficients for the Penning mixtures of neon and xenon

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The Townsend primary (α) and secondary (γ) coefficients and Paschen curves for neon, xenon, and their mixtures are reported. The primary coefficient was determined by measuring the variation in the luminous flux in a self-sustained Townsend discharge between two parallel-plate nickel electrodes spaced 21.5 mm apart. The values of the reduced primary coefficient, α/p_0, for Ne and Xe are about 10–20% lower than published values obtained by the classical method of Townsend. The α/p_0 values obtained for Ne agree with previous work using the same luminous-flux method. The ionization efficiency function η ($\eta = \alpha/E$, where E is the electric field) has a maximum value for Ne and 0.01% Xe. Also, the value of η for this gas mixture is larger than that for Ne + 0.1% Ar, especially at small values of E/p_0 (1–10 $\mathrm{V\,cm^{-1}\,Torr^{-1}}$).

INTRODUCTION

In 1937, Kruithof and Penning[1] were the first to measure the Townsend primary (α) and secondary (γ) ionization coefficients for various mixtures of neon and argon. Since then the Ne-Ar system has been extensively studied by several other authors,[2-5] but surprisingly enough, the Townsend coefficients for mixtures of neon with other rare gases such as xenon and krypton have not been reported. This paper presents the Townsend coefficients and Paschen curves for Ne, Xe, and mixtures of these two gases in a self-sustained Townsend discharge maintained between two parallel-plate electrodes. Results of similar measurements for mixtures of neon and krypton will be published later.

The theory behind the luminous-flux method used in the present investigation has been discussed in detail by Corrigan and Von Engel[6] and de Hoog *et al.*[7-10] However, a short review of the measurement technique may be useful to illustrate the difference between these data and those obtained by classical method of Townsend.[1-3,5]

The electron density in a Townsend discharge—a discharge in which space-charge distortion of the electric field is negligible—between plane parallel electrodes increases with the distance x from the cathode according to

$$N(x) = N(0)e^{\alpha x}, \tag{1}$$

where x is measured in the direction of the electric field and $N(0)$ is the electron density at an arbitrary plane $x = 0$ perpendicular to the axis of the discharge. Relation (1) is valid when the electrons are in equilibrium with the electric field and ionization and excitation by multiple collisions are absent.

A swarm of electrons moving through a gaseous medium excite and ionize gas atoms by collision. Since less energy is required to excite than ionize an atom, a relatively large numer of atoms in excited states will be produced owing to collisions with electrons in the swarm. These excited atoms emit photons reverting to a lower excited state. Under certain conditions, viz., assuming that the excitation temperature is constant, the radiation $\varphi(x)$ emanating from an elementary volume in the discharge at the position x is proportional to the electron density at that position, i.e.,

$$\varphi(x) = \varphi(0)e^{\alpha x}. \tag{2}$$

Hence, the variation in the electron density can be determined by measuring the radiation emanating from an elementary slab of the discharge as a function of x. If the radiation $\varphi(x)$ is detected by a photomultiplier tube, the signal $R(x)$ will be proportional to $\varphi(x)$. Hence, α may be deduced from a measurement of $R(x)$ as a function of the distance x of the lamina from the cathode.

In 1958 Corrigan and Von Engel[6] utilized this method to determine the excitation and dissociation coefficients in a non-self-sustained Townsend discharge in hydrogen. De Hoog *et al.*[7-10] utilized the same technique to measure the ionization coefficients for Ne, Ar, and mixtures of Ne and Ar in both non-self-sustained and self-sustained Townsend discharges. De Hoog *et al.* have shown that this luminous-flux method is a very useful technique for determining α directly and is not influenced by secondary effects.

The simple relation (1), and hence (2), is valid for pure gases when the electric field between the electrodes in homogeneous and the electrons in the gap are in equilibrium with the field. The growth of the radiation emitted from an elementary volume between cathode and anode is representative of the growth of the electron avalanche provided the fol-

lowing conditions are fulfilled: (i) The natural lifetime of the excited states under observation is small compared to the diffusion time of the excited atoms. This condition is readily met by most of the excited states. (ii) The imprisonment or self-absorption of the radiation is negligible; i.e., resonance radiations should not be used. (iii) The excitation of the state under observation is mainly by direct excitation of the ground-state atoms by electron impact; i.e., the pumping of the excited state by the cascade effect (population of lower nonresonant states by higher-energy states) is negligible compared to direct excitation of a ground-state atom by electron impact. In a Townsend discharge the charge particle density is quite low. Consequently, the population of excited states by electron-ion recombination processes will be small. Buurnsen *et al.*[10] have shown that in neon the cascading effect and the redistribution of excited states have no significant influence on the value of α measured with the luminous-flux method.

The definition of α is valid for Penning mixtures if the excited state acts as an intermediate state for the Penning ionization only in such a way that this stepwise ionization occurs at the same spot where the excitation took place. Gas atoms excited in the metastable and resonance states contribute toward ionization by Penning effect. The coupling of the resonance and metastable states together with diffusion of resonance radiation cause displacement of the intermediate state before Penning ionization takes place.[11,12] This displacement of the intermediate state influences the ionization mechanism and the ionization rate. Consequently, relation (1) is no longer strictly valid in Penning mixtures because the growth in the electron density is strongly influenced by the resonance radiation.[5,13] Nevertheless, the increase in the electron density with distance from the cathode can be described by an *apparent* ionization coefficient α defined by (1).[13]

The breakdown condition for a discharge gap between two parallel-plate electrodes is given by the condition[14]

$$\gamma(e^{\eta(V_b-V_0)}-1)=1\,, \tag{3}$$

where V_b is the breakdown voltage and V_0 is a constant characteristic of the gaseous medium. V_0 takes into account the fact that the electrons released from the cathode must be accelerated by the field before they are in equilibrium with the applied E/p_0. The ionization function η ($\eta=\alpha/E$, where E is the electric field) is defined as the numer of electrons produced in the gaseous medium by a single electron passing through a potential of 1 V. The Townsend secondary coefficient, γ, represents the number of electrons liberated from the cathode per positive ion arriving there. It incorporates the emission of electrons from the cathode under the action of radiation, metastable atoms, and positive ions. Relation (3) was used to determine the value of γ for various gases. The ionization efficiency function η was determined by the luminous-flux method discussed earlier, and the breakdown voltage for the gap was taken as the potential difference between the electrodes of the Townsend discharge.

EXPERIMENTAL APPARATUS

The discharge tube was made of two parallel planar electrodes of 82-mm diameter spaced 21.5 mm apart and housed in a Pyrex cylindrical tube of 120-mm i.d. The electrodes were parallel to within 0.2%. They were made of 20-mil-thick high-purity nickel (99.99% purity) and had a Rogowskii profile.[15,16] The electrodes were first machine polished with rouge and then electropolished. The nickel electrodes were vacuum fired at 900 °C for about 25 min before the final assembly.

Figure 1 is a schematic diagram of the experimental tube and the electrical connections. The

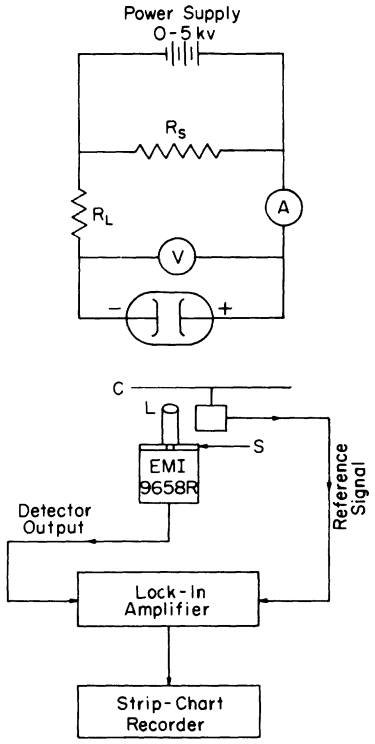

FIG. 1. Schematic diagram of electrical circuit and optical system for measuring luminous flux: S, 0.5-mm slit; L, lens; C, mechanical chopper.

potential applied to the discharge tube was obtained by passing a current (~2 mA) through the resistor R_s from a voltage-stabilized power supply (0.01% regulation). The load resistor R_L (~10^9) was used to limit the discharge current. The discharge current was monitored with a Keithley model 411 micro-micro ammeter (A). The potential across the discharge tube was measured using a Sensitive Research electrostatic voltmeter model ESD of input impedance 10^{15} having an accuracy of 1%.

The gas-handling system was of the Alpert type[17] and used Granville-Phillips bakeable valves and a capacitance manometer. Following a bakeout at 350 °C for over 48 h, the system attained a pressure of ~10^9 Torr with a rate of rise in the contamination pressure on the order of 10^{-10} Torr/min. The gas pressure was measured by using the capacitance manometer as a null indicator with an accuracy of 1%. The gas samples used were of high purity, obtained from General Electric in 2-liter Pyrex flasks. The various gas mixtures were obtained by mixing xenon and cataphoretically cleaned neon in measured volumes. Xenon contained about 0.005% Kr and 0.002% N_2; the other impurities were less than 0.001%.

The spatial variation in the luminous flux emitted by the discharge between the two parallel planar electrodes was measured with a photomultiplier tube (EMI 9658R) having an extended S-20 response. A 500-μm slit of 20-mm height mounted in front of the photocathode was imaged at the axis of the discharge tube with the aid of a spherical lens. The photomultiplier tube, the sampling slit, and the lens built into one single unit mounted on a microscope mechanical stage was moved parallel to the discharge tube axis. The light from the discharge was periodically interrupted by a mechanical chopper, detected by a lock-in amplifier (PAR 128) and displayed on a strip-chart recorder.

For the determination of α, the discharge tube was filled at a relatively high pressure (~100 Torr) and a discharge was maintained by passing a current of 10^{-6} A for 12 h or more. Various values of E/p_0 were obtained by pumping out the gas from the discharge tube. Thus, α/p_0 values were obtained at various E/p_0 values for the same gas.

RESULTS AND DISCUSSION

The variation of detector output $R(x)$ as a function of position x in the electrode gap for various values of the discharge current is shown in Fig. 2. The position $x = 0$ was arbitrarily chosen near the cathode. It clearly shows that for currents below 10^{-7} A, the detector output is proportional to the electron density in the discharge. At higher discharge currents, the space-charge distortion of

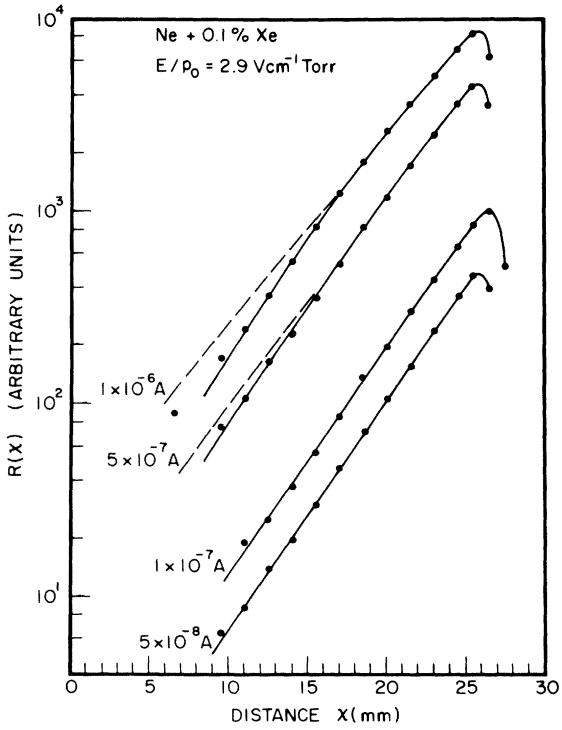

FIG. 2. Detector output $R(x)$ as a function of position x in the electrode gap for various discharge currents.

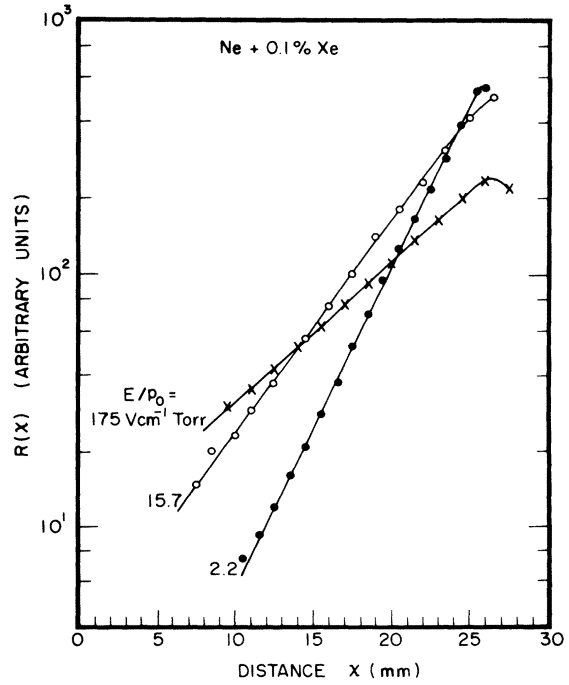

FIG. 3. Detector response $R(x)$ as a function of x for various values of E/p_0.

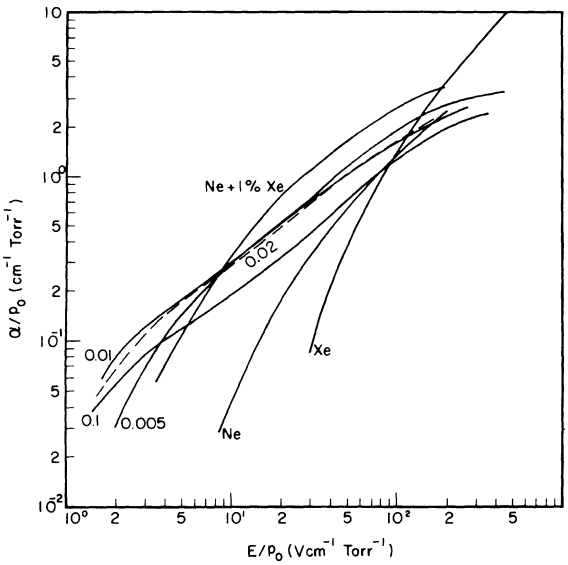

FIG. 4. α/p_0 vs E/p_0 for Ne, Xe, and Ne-Xe mixtures.

the electric field in the gap becomes important and the simple theory is no longer valid. On the basis of these observations it was decided to limit the discharge current to 5×10^{-8} A or less, which corresponds to a current density of 10^{-9} A/cm^2. At such low currents the space-charge distortion of the electric field in the electrode gap is negligible; hence the electric field E is equal to the potential difference between the electrodes divided by the electrode separation. The potential differences across the gap at these low currents is taken as the breakdown potential.

The variation in the detector output $R(x)$ as a function of x in the Ne+0.1% Xe mixture for various values of E/p_0 is shown in Fig. 3. It shows that the luminous flux emitted from an elementary slab of the discharge increases exponentially with distance from the cathode obeying relation (2). The reduced ionization coefficients α/p_0 obtained from similar data for various Ne-Xe gas mixtures

TABLE I. Ionization efficiency function η (V^{-1}) vs E/p_0(V cm^{-1} Torr^{-1}) in Ne-Xe mixtures.

% Xe / E/p_0 (Vcm^{-1} Torr^{-1})	0	0.005	0.01	0.02	0.1	1.0	100
				100η (V^{-1})			
1.5		2.57		3.13			
1.8		2.64	3.75	3.28			
2.0		2.68	3.90	3.38	1.58		
2.5		2.72	4.00	3.54	1.96		
3.0		2.70	3.93	3.63	2.30		
3.5		2.67	3.86	3.56	2.54	1.57	
4.0		2.65	3.75	3.50	2.70	1.79	
4.5		2.52	3.65	3.44	2.82	2.00	
5		2.48	3.52	3.38	2.90	2.14	
6		2.25	3.38	3.25	2.91	2.50	
7		2.07	3.21	3.14	2.93	2.79	
8		2.00	3.18	3.04	2.94	2.99	
9		1.94	3.06	2.94	2.89	3.11	
10	0.420	1.88	2.97	2.85	2.90	3.17	
12	0.550	1.79	2.86	2.71	2.83	3.46	
15	0.693	1.70	2.73	2.55	2.73	3.63	
18	0.806	1.64	2.64	2.44	2.64	3.69	
20	0.885	1.63	2.58	2.40	2.58	3.73	
25	1.04	1.58	2.48	2.30	2.44	3.70	
30	1.12	1.53	2.40	2.23	2.33	3.50	
35	1.17	1.50	2.33	2.17	2.23	3.37	0.386
40	1.23	1.48	2.28	2.11	2.14	3.30	0.513
45	1.24	1.46	2.22	2.06	2.07	3.22	0.633
50	1.28	1.43	2.20	1.99	2.00	3.14	0.730
60	1.35	1.41	2.17	1.90	1.92	3.03	0.904
70	1.38	1.38	2.07	1.81	1.81	2.91	1.05
80	1.38	1.34	2.00	1.76	1.71	2.81	1.18
90	1.39	1.30	1.94	1.71	1.67	2.72	1.29
100	1.38	1.26	1.88	1.65	1.60	2.63	1.40
120	1.37	1.20	1.78	1.50	1.48	2.38	1.54
150	1.31	1.11	1.60	1.33	1.33	2.11	1.70
200	1.25	0.975	1.38		1.18	1.75	1.90
250		0.860	1.20		1.03		1.98

are plotted as a function of E/p_0 in Fig. 4. Also plotted are the values of α/p_0 for neon and xenon. There is a fairly good agreement between the present values of α/p_0 for neon and that of Buurnsen *et al.*[10] obtained by using the present luminous-flux method. However, values of α/p_0 determined here are lower than those of Kruithof and Penning[1] as well as Chanin and Rork.[18] The present values of α/p_0 for xenon are also approximately 20% lower than those of Kruithof[19] determined by the classical method. The possible reasons for the discrepancy between values determined by authors utilizing the classical method of Townsend and the luminous-flux method have been discussed in detail by Buurnsen *et al.*[4]

Table I lists the η values for the various mixtures of Ne and Xe together with those for pure gases for comparison. Figure 5 shows a plot of η vs E/p_0 for neon containing 0.01 and 0.1% of xenon and argon.[1] For the Ne-Xe system, η has the largest value for Ne admixed with 0.01% Xe. In comparison, 0.1% Ar produces the largest value of η for the Ne-Ar system. Also, in Ne+0.01% Xe, the maximum value of η (0.0040 V^{-1}) is larger than that (0.003 71 V^{-1}) for Ne+0.1% Ar. The neon metastable state $1s_5$ has an energy of 16.58 eV and the ionization potentials of Ar and Xe are 15.76 and 12.13 eV, respectively. Because of the larger energy difference in the Ne-Xe system compared to Ne-Ar, the concept of resonance energy exchange would imply it to be the weaker Penning mixture of the two systems. The present study casts some doubt on this concept. However, a direct measurement of the Penning reaction cross section, perhaps using the optical absorption technique of Mitchel and Zemansky,[20] and Phelps *et al.*[21,22] will be of much help in explaining this observation.

It is important to note that the amount of Xe required for optimum Penning effect in Ne is ten times smaller than with Ar. This is not surprising since the average electron energy and hence the production of metastable neon atoms is influenced by the concentration of the admixed gas. The electron-neutral collision cross section of Xe is much higher than that for Ar as well as Ne for electron energies larger than 1 eV.[23] The maximum value of η occurs at values of E/p_0 approximately corresponding to the electron energy for optimum production of metastable neon atoms.[14,24] Another "maximum" in η at large values of E/p_0 is due to direct ionization of gas atoms by electron impact. At large concentrations of xenon most of the electron production in the electrode gap is due to direct excitation and ionization of xenon, which reduces the average energy of electrons and results in fewer neon metastables. At very low concentrations (~10^{-3}%) of xenon there are not enough of them available for the Penning reaction,

$$Ne^m + Xe \rightarrow Ne + Xe^+ + e + KE\,. \qquad (4)$$

Consequently, the value of η is small. The present investigation shows that Ne containing 0.01% Xe produces the largest value of η for the Ne-Xe gas mixture.

Breakdown potentials (Paschen curves) between two parallel-plate nickel electrodes for various mixtures of Ne with Xe as a function of the product of the electrode separation (d) and the reduced gas pressure (p_0) are shown in Fig. 6. Also shown are the breakdown potentials for pure Ne and Xe.

At low values for p_0d, i.e., far on the left-hand side of the Paschen minimum, the discharge quite frequently occurred not between the front surfaces of the electrodes but it took a longer path (corresponding to a more favorable p_0d value) around and to the back surface of the electrodes. This was clearly evident from the measured variation in the luminous flux in the discharge tube. Under such conditions the maximum in the luminous flux

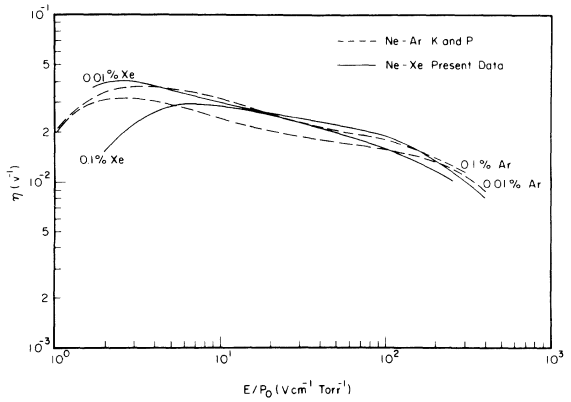

FIG. 5. Ionization efficiency function η vs E/p_0 for neon containing 0.01 and 0.1% of xenon and argon.

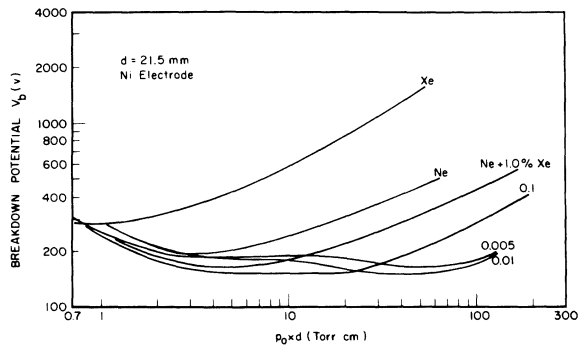

FIG. 6. Breakdown potential V_b vs p_0d for Ne, Xe, and Ne-Xe mixtures.

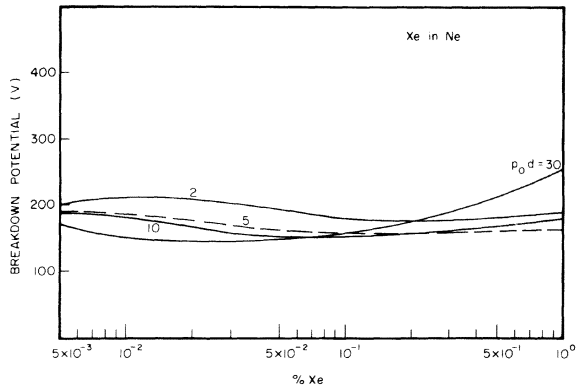

FIG. 7. Variation in the breakdown potential for Ne-Xe mixtures as a function of xenon concentration.

occurred not near the front surface of the anode, but behind it. The determination of α was restricted to those values of p_0d for which the discharge took place between the front surfaces of the electrodes. The effect of the addition of xenon to neon in lowering the breakdown potential of the gap is evident at large p_0d, values which correspond to low E/p_0 values. A plot of the variation in the breakdown potential as a function of xenon concentration obtained from Fig. 6 is shown in Fig. 7.

The second ionization coefficient γ determined by substituting the measured values of η and the breakdown potential V_b in Eq. (3) is shown in Fig. 8. The value of V_0 is assumed to be the same as that for Ne-Ar mixtures.[1,19] Since η appears in the exponent, values for γ are less accurate than those for η. As expected the values of γ are dependent upon the nature of the cathode surface, the gas composition, as well as E/p_0.[1,14]

CONCLUSION

The present values of α/p_0 for neon are in good agreement with those reported by Buurnsen *et al.*[10] utilizing the same luminous method and are about 10–20% lower than those obtained by the classical Townsend method. This is also observed in the case of xenon. It appears, as pointed out by Jones and Morgan,[25] that the difference in the values of α/p_0 obtained by the two methods is of a fundamental nature. It is interesting to note, however, that de Hoog[26] and Buurnsen *et al.*[10] did not find any significant difference in the values of α/p_0 for Ne admixed with 0.1% Ar obtained by the classical Townsend and the luminous-flux methods.

This study shows that neon containing small amounts of xenon forms a very efficient Penning mixture. Neon admixed with 0.01% Xe gave the largest value for η. In addition, the maximum value of η for Ne + 0.01% Xe is larger than that for 0.1% Ar. The difference in energy between the

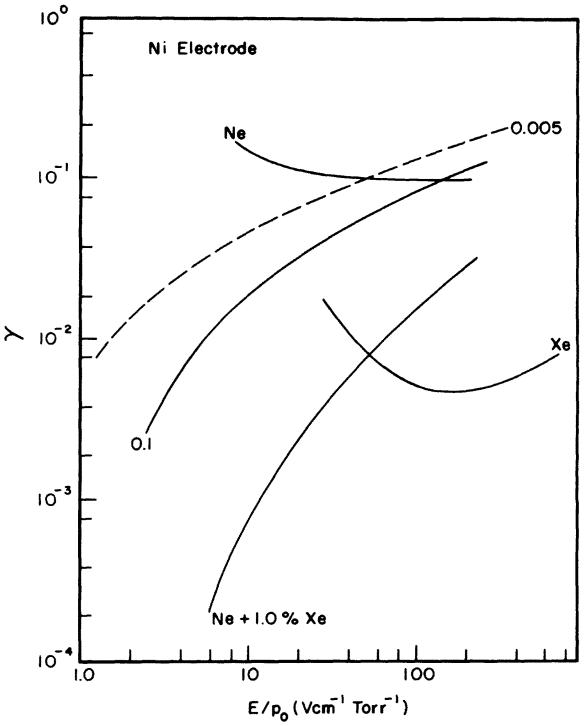

FIG. 8. γ vs E/p_0 for nickel cathode.

lower neon metastable level ($1s_5$) and the ionization potentials of Ar and Xe are, respectively, 0.82 and 4.43 eV. According to the theory of resonance energy transfer,[27] the Ne-Ar system, being very close to exact resonance ($\Delta E = 0.09$ eV), should be a better Penning mixture than Ne-Xe. The value for the ionization efficiency function η for Ne + 0.01% Xe is larger than that for Ne + 0.1% Ar at low values of E/p_0 (1 to 10 V cm^{-1} Torr^{-1}), which indicates that the concept of the resonance energy transfer may not be quite applicable to the Penning mixtures.

The variation of the second ionization coefficient γ with E/p_0 indicates that for the Ne-Xe gas mixture the secondary electron emission from the cathode surface is mainly due to the action of positive ions. Also, as expected, the value of γ depends on the nature of the cathode surface and the composition of the gaseous medium.

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