Volume Production of High Negative Hydrogen Ion Density in Low-Voltage Cesium-Hydrogen Discharge

F.G.Baksht[1], V.G.Ivanov[1], S.M.Shkol'nik[1], M.Bacal[2]

[1] *A.F.Ioffe Physical-Technical Institute, Russian Academy of Sciences, St. Petersburg, 194024 Russia,*
[2] *Laboratoire de Physique et Technologie des Plasmas, Ecole Polytechnique, UMR 7648 du CNRS, 91128 Paleseau, France*

Abstract

The work is dedicated to the theoretical and experimental investigation of the intensive H volume-plasma generation in cesium-hydrogen discharge. A low-voltage (LV) mode of the discharge, a LV arc with a heated cathode, is studied. Theory of the discharge was created. It was shown theoretically that very high H^- concentration (N_{H^-} ~ 10^{13} cm^{-3}) may be achieved in the discharge plasma. A comparison between the discharge theory and the experiments was fulfilled. The main plasma parameters were measured by probe method. The H^- concentration in plasma was determined from the experimentally measured absorption of laser radiation due to the photodetachment of electrons from H^- ions. The existence of a very high H^- concentration (~ 10^{13} cm^{-3}) in plasma was proved experimentally. Theoretical modeling shows that, with a high electron emission from the cathode (10 A/cm^2), the maximum H^- density is located in plasma near the anode. It allows extraction of narrow and bright H^- beam through a small opening in the anode.

1. Introduction

H^- ions are used in accelerators, fusion research, plasma technology etc. The volume-plasma (VP) sources are widely used for H^- generation, H^- ions being created in plasma due to dissociative attachment (DA) of electrons to rovibrationally excited H_2 molecules in the ground electronic state $X^1\Sigma_g^+(v)$ [1,2]. At present, in H^- VP sources, $X^1\Sigma_g^+(v)$ molecules are created, to a marked degree, because of radiative deexcitation of the singlet electronically excited states $B^1\Sigma_u^+$, $C^1\Pi_u^{\pm}$ etc. [3]. The excitation cross-sections of these states by electrons have noticeable values if the electron energy E > 40 eV [4]. Therefore, comparatively high voltage discharges, where 50 eV ≤ E ≤ 150 eV, are used for H^- generation. It leads to diminution of H_2 concentration and undesirable increase of H atom concentration due to H_2 dissociation by fast electrons. Because fast electrons destroy H^- ions [5], the VP H^- sources consist usually of two chambers [6]. In the first chamber, where excited molecules are generated, the beams of fast electrons exist. In the second chamber, where H^- ions are created by DA, the fast electrons are absent. Therefore the H^- source represents a complicated device (e.g. see [7,8]) with complex plasma composition because several kinds of positive hydrogen ions and electronically excited H_2 molecules penetrate in the volume, where DA occurs.

The present communication is dedicated to H generation in low-voltage cesium-hydrogen discharge. Such VP H^- source was proposed in [9], where it was shown that, in this kind of the discharge, a high concentration of vibrationally excited molecules and heated thermal electrons, needed for DA, may be obtained by a simple method in a single discharge volume.

2. The main peculiarities of LV cesium-hydrogen discharge.

In the beginning, a theory of LV cesium-hydrogen discharge was created [9-13]. A typical potential distribution in the discharge consists of a potential well for thermal electrons. The well is separated from the electrodes by very narrow near-electrode Langmuir layers, which dimensions $L_0 \sim 10^{-3}$ L [14], where L is an interelectrode gap. A cathode voltage drop φ_1 is limited by the condition $\varphi_1 < E_d/e$, where $E_d \cong 8.8$ eV is a threshold of H_2 direct dissociation by electron impact from the ground electronic and vibrational state $X^1\Sigma_g^+(v=0)$. At such φ_1 values, the processes of H_2 direct dissociation are almost eliminated in plasma as well as the processes of H_2 or H stepped ionization via electronically excited states. It leads to a comparatively small contamination of the discharge by atomic hydrogen and therefore improves significantly the vibrational distribution function (VDF) of H_2 molecules, because the rates of v-t relaxation between H_2 and H [15] exceed the corresponding rates for pure H_2 [16] up to three orders of magnitude. It also prevents a formation of atomic or molecular hydrogen ions. Therefore the discharge plasma contains only Cs^+ ions, electrons and H^- ions, the thermal electron concentration n_e being very large ($n_e \sim (10^{13} - 10^{14})$ cm^{-3}). n_e exceeds a concentration $n_e^{(1)}$ of cathode beam electrons up to two or three orders of magnitude. Therefore electron-vibration kinetics depends only on thermal plasma electrons.

Two modes of the discharge were considered theoretically: a discharge in a dense plasma [9-11] and discharge in a rare plasma [12-13]. In the dense plasma, free paths of charged and neutral particles are significantly less than the gap L and the energy relaxation length $L_\varepsilon = (D_0\tau_\varepsilon)^{1/2}$ of cathode electrons is also smaller than L. Here D_0 and τ_ε are a diffusion coefficient and a time of fast (beam) electron energy relaxation due to pair Coulomb collisions. In the discharge considered, where $n_e/N_{H2} \geq 10^{-3}$ and $\varphi_1 < E_d/e$, this relaxation occurs due to collisions between fast and thermal electrons [14,17]. In this kind of the discharge the electron energy distribution function (EEDF) is very close to Maxwellian one, and the plasma is described by the set of hydrodynamic equations (continuity equations, equations of motion and energy equations for electrons and heavy particles) and corresponding boundary conditions [11,14]. The mechanism of thermal electron heating is Coulomb pair collisions between fast and thermal electrons in near-electrode plasma region. In rare plasma, free paths of the thermal particles in the majority of cases are larger than the gap L. Also, $L_\varepsilon > L$, i.e. the length of fast electron maxwellization is greater than the gap. In this case, the EEDF is non Maxwellian, and thermal electrons cannot be heating by pair Coulomb collisions between them and beam electrons. In this case, the main mechanism of thermal electron heating is a collisional damping of Langmuir waves excited in plasma by means of plasma-beam instability. Because of this mechanism, a significant part of initial beam energy is transferred to thermal electrons [14,18].

In this report, a comparatively high H_2 pressures ($P_{H2} \geq 1$ Torr) are considered, and the plasma is dense enough. Because of high n_e and P_{H2} values, VDF of H_2 molecules is mainly created due to the volume-plasma processes, the vibrational deexcitation on the electrode surface being almost inessential. Therefore the VDF does not depend on unknown probabilities of the vibrational deactivation of molecules on cesium-coated surfaces. It allows to compare experimental and theoretical data without fitting parameters.

Experimental investigation of LV cesium-hydrogen discharge was performed in [14,19-24]. In the initial experimental works (e.g. see [14,19-21]) the probe method of plasma diagnostics was developed and local parameters (n_e , T_e , φ) were measured. It was shown experimentally that all the plasma parameters, which are needed for intensive H generation, were really obtained in the discharge plasma. In particular, just in the first experimental works (e.g. see [14,19-21]), it was shown that high electron concentration n_e and optimum electron

temperature T_e for H^- generation by means of DA were really achieved in the discharge plasma.

3. H^- volume-plasma concentration in LV cesium-hydrogen discharge.

In the recent works [22-24] the H^- concentration, N_{H^-}, in plasma was measured experimentally. The diode with parallel-sided electrodes and interelectrode distance 3 mm was used in the experiment. The face ends of the cylinders 12 mm in diameter were used as the electrodes. A platinum foil, which was welded to the face end of the cathode cylinder, was used as an emitter in cesium vapor. A cylindrical probe was inserted into the gap through a hole in the center of the anode. The probe axis was parallel to electrode surfaces. The probe diameter and length were 0.1 mm and 2mm respectively. The probe was placed in the center of the gap. N_{H^-} was determined from the experimentally measured absorption of laser radiation due to the photodetachment of electrons from H^- ions. A semiconductor laser of a continuos operation and a power $P \cong 0.1$ W was used. A spectral width of the radiation was approximately $\Delta\lambda \cong 2$ nm, and the maximum of the intensity was at $\lambda_0 = 816$ nm ($h\nu_0 = 1.52$ eV). Concentration of the excited Cs atoms were measured experimentally and calculated by theoretical methods. It was shown that real absorption of laser radiation, which was observed in the experiment, is larger than photoabsorption by excited Cs atoms by two or three orders of magnitude. The laser wavelength corresponds to the flat maximum of H^- photoionization cross-section ($\sigma_0 \cong 4\cdot10^{-17}$ cm^2). As a result, the values of electron concentration n_e, temperature T_e and potential φ were determined by probe method in the center of the gap, and H^- concentration was measured by the absorption of laser radiation in the near-cathode plasma. Simultaneously the distributions of all plasma parameters in the gap, including the H_2 VDF and densities of excited Cs atoms, were determined by the selfconsisted discharge theory. The following processes were taken into account in calculations of VDF: e-v exchange, which was calculated according to the method of [25] (the cross section $\acute{o}_{01}$ of excitation 0f the v = 0 v = 1 transition by electron impact was borrowed from [26]) ; v-v and v-t exchange between H_2 molecules [27] (the corresponding rate constants were borrowed from [16, 28]); v-t exchange between H_2 molecules and H atoms, which was taken into account according [15, 29] and the processes of DA, which were considered according to [2, 25]. The several other processes were also taken into account as it was done in [30]. The experimental electron temperature in the center of the gap was taken as an initial value for theoretical calculations. Because, in experimental conditions, a Cs concentration N_{Cs} in the gap is significantly less than concentration N_{Cs} (T_{Cs}) just above the surface of liquid cesium, the theoretical parameter $N_{Cs}^{(0)}$, which is equal to total Cs concentration averaged over the discharge gap, was found from the additional condition, namely by equating the theoretical and experimental Cs^+ concentration in the gap center. The maximum deviation of experimental N_{H^-} values from corresponding points of the theoretical $N_{H^-}(x)$ curves, which was obtained in the present experiments, was less or equal to 40 % . H^- concentrations up to $N_{H^-} \cong (0.5\text{-}0.6)\cdot10^{13}$ cm^{-3} were obtained experimentally near cathode at cathode electron current density $j_s \cong (3\text{-}4)$ A/cm^2 and hydrogen pressure $P_{H2} \cong (1 - 2)$ Torr. According to the last theoretical calculations, the H^- concentration up to $N_{H^-} \sim 10^{13}$ cm^{-3} may be obtained in plasma near anode. These results were obtained for comparatively high cathode emission current density $j_s = 10$ A/cm^2. Such j_s value cannot be obtained in stable mode of operation from the platinum cathode at optimum Cs pressure $P_{Cs} \cong 10^{-2}$ Torr, which was used in the present experiments, but may be easily obtained from LaB_6 flat cathode.

Acknowledgments

This works was done with the support of the Scientific Program of the St. Petersburg Center of Russian Academy of Sciences. One of us (M. Bacal) gratefully acknowledges the support of the European Community (Contract HPRI-CT-2001-50021).

References

1. Bacal M. and Hamilton G.W., Phys. Rev. Lett., **42**, 1538 (1979)
2. Wadehra J.H. Phys. Rev. A, **29**, N 1, 106 (1984)
3. Hiskes J.R. Appl.Phys. Lett., **57**, 231 (1989)
4. Hiskes J.R. J. Appl.Phys., **51**, 4592 (1980)
5. Janev R.R., Langer W.D., Evans K.E. and Post D.E. *Elementary Processes in Hydrogen-Helium Plasmas.* Berlin. Springer, (1987).
6. Bacal M., Nucl. Instrum. Methods Phys. Res., B**37**/**38**, 28 (1989)
7. Kuroda T., Kaneko O., Oka Y. et. al. Proc. of the VII Int. Symposium on Production and Application of Light Negative Ions.(Brookhaven, Upton, N.Y.,USA, 201 (October 1995)
8. Tsumori K., Takanashi T., Asano S. et. al. . Proc. of the VIII Int. Symposium on Production and Application of Light Negative Ions. Villagium of Giens – Cadarache, France, 93 (1997)
9. Baksht F.G., Ivanov V.G.,Pis'ma Zh. Tekh. Fiz. **12**, N 11, 672 (1986)
10. Baksht F.G., Elizarov L.I., Ivanov V.G., Yur'ev V.G. Fiz. Plazmy, **14**, N 1, 91 (1988)
11. Baksht F.G., Elizarov L.I., Ivanov V.G.,. Fiz. Plazmy , **16**, N 7, 854 (1990)
12. Baksht F.G., Ivanov V.G., Kostin A.A. Pis'ma Zh. Tekh. Fiz., **18**, N 12, 83 (1992)
13. Baksht F.G., Ivanov V.G., Kostin A.A. Zh. Tekh. Fiz., **63**, N 9, 173
14. Baksht F.G., Djuzhev G.A., Elizarov L.I.., Ivanov V.G., Kostin A.A., Shkol'nik S.M..Plasma Sources Sci. Technol. **3**, 88 (1994)
15. Garcia E. and Lagana A. Chem . Phys. Lett., **123,** 365, (1986)
16. Kiefer J.H. J. Chem. Phys., 57, 1938, (1972)
17. Baksht F.G., Ivanov V.G. Fiz. Plasmy, **12**, 286, (1986)
    Baksht F.G., Ivanov V.G. Sov. J. Plasma Phys. , **12**, (1986)
18. Baksht F.G., Kolosov B.I., Kostin A.A., Tchuraev R.S., Yuriev V.G. Mathematical Modelling of Processes in Low-voltage Plasma-Beam Discharge. (Moscow, Energoatomizdat, 1990)
19. Baksht F.G., Dyuzhev G.A., Elizarov L.I., Ivanov V.G., Filatov A.G., Shkol'nik S.M.
    Zh. Tekh. Fiz., **62**, 148, (1992)
    Baksht F.G., Dyuzhev G.A., Elizarov L.I., Ivanov V.G., Filatov A.G., Shkol'nik S.M.
    Sov. Phys. Tech. Phys., 37, 959, (1992)
20. Baksht F.G., Dyuzhev G.A., Elizarov L.I., Ivanov V.G., Kostin A.A., Shkol'nik S.M.
    IEEE Trans. Plasma Sci. , 21, 552, (1993)
21. Baksht F.G., Ivanov V.G., Kostin A.A., Nikitin A.G., Shkol'nik S.M. Zh. Tekh. Fiz.,65, N 8, 1927 (1995)
22. Baksht F.G., Ivanov V.G., Kon'kov S.I., Shkol'nik S.M. . Zh. Tekh. Fiz., **71**, N 8,17**,** (2001)
23. Baksht F.G., Elizarov, Ivanov V.G., Kon'kov S.I., Mitrofanov N.K., Shkol'nik S.M. Fiz. Plazmy, **29**, N 2, 1 (2003)
24. Baksht F.G., Ivanov V.G., Kon'kov S.I., Shkol'nik S.M. and Bacal M. J. Phys. D: Appl. Phys., **35**, 1 (2002)
25. Skinner D.A., Bruneteau A.M., Berlemont P., Courteille C., Leroy R., Bacal M.
    Phys. Rev. E., **48**, N 3, 2112, (1993)
26. Morrison M.A., Crompton R.V., Saha B.C., PetroviæZ.L. Aust. J. Phys., **40**, N 3, 239, (1987)
27. Nonequilibrium Vibrational Kinetics (Ed. by M. Capitelli). Springer-Verlag, (1986).
28. Kiefer J.H., Lutz R.M. J. Chem. Phys., **44**, N 2, 668, (1966)
29. Garcia E., Lagana A. J. Phys. Chem., **90**, 987, (1986)
30. Baksht F.G., Elizarov, Ivanov V.G., Yuriev V.G. Fiz. Plazmy, **14**, N 1, 91 (1988)