GENERAL EXPERIMENTAL TECHNIQUES

A Microwave Discharge Source Operating at Pressures of Several Atmospheres

V. M. Akimov^a, G. Witte^b, L. I. Kolesnikova^a, L. Yu. Rusin^a, and J. P. Toennies^c

 ^a Institute of Energy Problems of Chemical Physics, Russian Academy of Sciences, Leninskii pr. 38, korp. 2, Moscow, 119334 Russia
^b Ruhr-Universität Bochum, Universitätsstraße 150, NBCF03/296, Bochum, D-44780 Germany
^c Max-Planck-Institut für Dynamik und Selbstorganisation, Bunsenstrasse 10, Gottingen, D-37073 Germany

Received July 11, 2008

Abstract—The design of a microwave source in which a discharge is initiated by an electromagnetic surface wave at 2.45 GHz is described. A stable discharge was supported at a gas pressure p_0 exceeding the atmospheric pressure in He, N₂, and in H₂–Ar, H₂–He, and O₂–He mixtures in a 2-mm inner diameter quartz tube with a 0.15-mm diameter nozzle at a 50- to 115-W microwave power. A degree of dissociation of up to 80% was reached for pure H₂ at $p_0 = 6$ Torr and a 6% mixture of H₂ and He at $p_0 = 50$ Torr. When p_0 increases to 19 Torr for H₂ and to 300 Torr for the mixture, the hydrogen-atom beam intensity, in spite of a decrease in the degree of dissociation, increases due to narrowing of the beam particle velocity distribution.

PACS numbers: 52.50.Dg, 07.77.Gx, 39.10.+j

DOI: 10.1134/S0020441209030166

The set of experimental methods used in studies with molecular beams includes a wide circle of instruments intended for generating various beams of various atoms and molecules in a very wide range of kinetic energies. One of the most urgent problems of this important component of the molecular-beam method is production of beams of free atoms and radicals possessing a high chemical activity, for example, hydrogen and oxygen atoms.

The most general requirement imposed on beam sources is to reach a maximal intensity with as narrow as possible velocity distribution of beam particles and the possibility of changing their energies. To one or another extent, this requirement can be satisfied using a supersonic beam with production of atoms in the high-frequency or microwave discharge plasma [1–3]. It is evident that the higher the gas pressure in the discharge region and the higher the efficient degree of dissociation, the more efficient these methods are. The latter value is determined by the ratio of the dissociation and atomic-recombination rates in the discharge region and the gas expansion from the nozzle.

Paper [3] presents an atomic oxygen beam source with a degree of O_2 dissociation of ~70% at a 350-Torr pressure for a 5- to 10% mixture of oxygen and argon, when the power in the discharge varies from 140 to 195 W. The generation of hydrogen atomic beams has a substantial problem, and the working pressures in the discharge at sufficiently high dissociation degrees are, as a rule, no more than 1 Torr [4–8].

To initiate a stable discharge for generating an atomic particle beam at increased pressures is more dif-

ficult than to do it at low pressures. These difficulties are, in particular, related to the problem of matching the impedances of the plasma and HF or microwave source when the gas pressure and, hence, the plasma density increases. In turn, the plasma characteristics substantially depend on the gas composition, the reachable plasma temperature, and another conditions. To increase the efficiency of plasma sources of beams, it is necessary to support the discharge directly near the nozzle to decrease the degree of atomic recombination before the gas-dynamic beam expansion occurs. In this case, the temperature of the discharge-tube walls near the nozzle should be in the optimal range, taking into account the kinetics of reactions in the plasma and the risk of melting of the nozzle orifice.

This work describes the results obtained in the process of designing the source for generating and supporting stable microwave discharges in various gases and their mixtures with noble gases in quartz tubes with a 2-mm inner diameter and a 0.15-mm-diameter nozzle. The design is based on a surfatron [9, 10], in which the discharge is initiated by a surface electromagnetic wave. The advantage of such devices is that it is possible to produce a stable plasma in them at pressures of up to several atmospheres, since, in this case, drawbacks related to problems of sustaining an HF or microwave discharge at elevated pressures in resonance sources are eliminated. Characteristics of the discharge initiated by the surface wave were intensively studied for plasma of Ar [11]; He [12]; O₂ [13, 14]; N₂ [15]; He, Ne, and Ar [16]; H₂ [17, 18]; and O₂–N₂ [19]; and Ar–N₂ mixtures [20, 21].

The discharge structure of the surfatron consists of two coaxially placed metallic cylinders forming a section of the coaxial line short-circuited on one end by a plunger and having an circular gap on the other end. The microwave field, penetrating through this gap, can excite a surface wave creating plasma in a gas-filled dielectric tube. The power is transmitted to the discharge structure through a coupler ensuring the capacitive coupling with the inner cylinder of the discharge structure. The efficiency of the microwave energy transmission from the generator and transmitting line into the plasma in the surfatron depends on three characteristics, namely, distance c between the coupler and surface of the inner coaxial cylinder; distance L between the plunger and end of the discharge structure; and annular gap value a. These distances (gaps) can be optimized based on measurements of the influence from each factor on the reflected power. The optimized dependencies of the surfatron, i.e., the reflected-power variation as a function of positions of the above parameters, given in [22], show that, for argon, an increase in the gas pressure from 0.15 Torr to atmospheric requires almost no changes in tuning component positions to reach the minimal reflected power. However, the given data on the discharge in helium indicate a significant sensitivity of settings and a shift of the optimal tuning point, when low pressures change to higher ones. Since this difference in positions of tuning components is several tenths of millimeter, while designing our discharge source, it was expedient to take measures to substantially increase the accuracy of tuning and setting measuring indicators.

DESIGN OF THE PLASMA-GENERATING DEVICE

Figure 1a shows the design of the source we developed, and Fig. 1b shows one version of the discharge tube plant. The surfatron consists of three basic units, namely, microwave-energy inputting section 1, housing 2, and tuning section 3. Inputting section 1 is a short $50-\Omega$ coaxial line formed by conductor 4 and screen 5. On one side, the line terminates with microwave connector 6, and on the other, with coupling plate 7. The entire section can be moved upward and downward by a screw-nut pair and a worm gear. One turn of the tuning flywheel fixed at the end of the worm corresponds to a 1/34-mm shift.

Eighty-mm-long housing 2 with a 64×64 -mm section and a 32-mm inner diameter cylinder is made of brass. The housing is cooled by water that circulates along 12 5-mm-diameter channels drilled concentrically along the axis. The surface of the inner cylinder is polished to improve the contact between it and plunger 8 moving during tuning. For higher reliability, the contacts between the plunger, housing, and tuning tube 9 are made as divided beryllium bronze springy lobes.

Tuning section 3 contains three identical worm gears with a 1 : 108 transmission ratio for moving the

Maximal attainable pressures (in atm) of the gas in the plasma-sustaining zone for various gases and mixtures at different microwave power levels

Gas or mixture	50 W	70 W	115 W
Helium	1.2	2.5	3.2
Nitrogen	0.4	0.75	1.5
Hydrogen	0.06	0.15	0.3
6% H_2 in Ar	1	2.5	3.5
10% H_2 in Ar	0.7	1.5	2.5
3% H ₂ in He	0.5	1.2	1.75
6% H ₂ in He	0.4	1	1.5
10% O ₂ in He	0.7	1.1	2.5

plunger, tuning bush, and discharge tube I. They shift at 1/54, 1/72, and 1/72 mm, respectively, per turn of the drive flywheel placed at the worm end. The second end of the worm, as for the motion of the microwave-inputting section, was connected to five-digit turn counter (not shown in the figure). The worm wheels rotated on balls IO placed in grooves, which were made on wheels and clamp parts adjoined to them.

Two types of discharge tubes can be used in this design. One of them (tube *I* shown in Fig. 1a) is a quartz tube with a 2-mm inner and 4-mm outer diameter and a 0.15-mm-diameter nozzle. Discharge tube *II* (Fig. 1b) is soldered of three 4×1 -, 8×1 -, and 12×1 -mm-size concentrically placed quartz tubes. There are holes in the back part of the discharge tube for the cooling-air inlet and outlet. The inner tube, in which the discharge was initiated, had a 0.15-mm-diameter nozzle at the end of its front part.

Cup 11 was connected to the front of the surfatron housing. To match the source to the vacuum system, the cup was inserted into the vacuum chamber with a sealing ring gasket around the outer cylinder. The entire assembly was fixed on a movable table moving along the axis to optimize the nozzle-skimmer distance, when the unit was further used as an atomic source. When discharge tube II was used, adjusting Teflon bushes 12 were replaced with bushes 13 (Fig. 1b) and the tube-back-end-sealing and cooling-air-supplying system was also replaced. In this case, the front part of the surfatron was directly connected to the vacuum chamber by flange 14 with a seal. In this case, to seal the inputting section, tuning tube, and plunger, viton ring gaskets were used during their movements. The discharge can be observed through window 15 in adapter 16, through which the gas was injected, and electrode 17 for initiating the discharge. The distance between this electrode and the discharge zone was 30 mm.

The discharge source was powered by a JJy4-11 microwave generator at 2.45 GHz ($P_{max} = 200$ W). The powering circuit contained a circulator for protecting the generator magnetron at high reflected power levels,



Fig. 1. (a) Design of the discharge microwave source and (b) one version of the discharge tube design and its installation: (1) microwave energy-inputting section, (2) housing, (3) tuning section, (4) inner conductor, (5) screen, (6) microwave connector, (7) coupling plate, (8) plunger, (9) tuning tube, (10) ball, (11) cup, (12) Teflon bush, (13) Teflon bush, (14) flange, (15) window, (16) adapter, (17) initiating electrode, and (I, II) discharge tubes.

a bidirectional coupler, and a matched absorbing load (50 Ω , 50 W). In addition, identical direct- and reflected-power meters including calorimetric converters and measuring wattmeter modules were used.

RESULTS

One of basic purposes for studying the operation of the discharge source was to determine the possibility of generating stable and reproducible plasma in a quartz tube in various gases and mixtures at pressures in the discharge varying from several Torr to several atmospheres. For this purpose, microwave plasma was excited in mixtures of 6% H₂ in Ar, 10% H₂ in Ar, 3% H₂ in He, 6% H₂ in He, 10% O₂ in He, and in pure gases (Ar, He, N₂, and H₂). Compressed air was used as a cooler of the discharge tube.



Fig. 2. Dependence of the reflected microwave power on the position of the coupling component (gap size c) for the discharge in He at $p_0 = 1$ atm (1, a = 5.25 mm and L = 26.1 mm) and 2 atm (2, a = 4.82 mm and L = 26.08 mm). The power in the discharge is 75 W.

Plasma is easily formed in Ar, where discharge can be maintained up to 5-atm pressures and even higher at a 20-W applied power. Most of the tests were performed with mixtures of noble gases and hydrogen, bearing in mind the further use of this unit as a source of hydrogen atoms.

As was already noted, positions of three tuning elements can be optimized, based on measured actions of each on the reflected power value. For this purpose, each of these elements was tuned at fixed positions of two others. Figures 2-4 show examples of dependences of the reflected power on the positions of the tuning elements for the discharge in helium at pressures $p_0 = 1$ and 2 atm and a 75-W power in the discharge. As follows from these dependencies, the optimal positions of the coupler (c) and plunger (L) do not vary when the gas pressure increases from 1 to 2 atm. In this case, gap a changes by 0.43 mm and the tuning range of the plunger and tube becomes narrower. When the gas is changed, the nature and behavior of tuning curves remains unchanged. However, the positions of the plunger, coupler, and tuning bush change. For example, for a 6% mixture of H_2 in Ar, distances L = 25.82 mm, a = 5.12 mm, and c = 1.9 mm at $p_0 = 1$ atm and at the same power as for helium plasma. For $p_0 = 1$ atm, the minimal reflected power corresponded to L = 26.1 mm, a = 5.25 mm, and c = 3.5 mm and, for $p_0 = 2$ atm, L =26.08 mm, a = 4.82 mm, and c = 3.5 mm.

Discharge was initiated for all studied gases and mixtures, excluding Ar, at pressures of several Torr and powers of 50 W and higher by the electrode inserted along the axis of the discharge tube. As the pressure Reflected power, % 18 ° 16 01 14 ο 12 0 10 8 6 4 2 0 <u></u> 25.6 25.8 26.2 26.4 26.8 26.0 26.6 L, mm

Fig. 3. Dependence of the reflected power on the position of the plunger (gap size *L*) for a discharge in He at $p_0 = 1$ atm (*1*, *a* = 5.25 mm and *c* = 3.5 mm) and 2 atm (2, *a* = 4.82 mm and *c* = 3.5 mm). The power in the discharge is 75 W.

increased, the positions of the tuning elements were determined more exactly.

While being tuned, discharge tubes of both types (I and II) demonstrated the same characteristics. However, when the surfatron is used as a source of atoms, a certain preference should be given to tube I due to better cooling conditions. The drawback of this design



Fig. 4. Dependence of the reflected power on the position of the tuning tube (gap size *a*) for the discharge in He at $p_0 = 1$ atm (*1*, *c* = 3.5 mm and *L* = 26.1 mm) and 2 atm (*2*, *c* = 3.5 mm and *L* = 26.08 mm). The power in the discharge is 75 W.



Fig. 5. Efficiency of dissociation for a discharge in pure H_2 as a function of the hydrogen pressure. The power in the discharge is 15 W.

consisting in attaching the sulfatron to the movable table for changing the nozzle–skimmer distance is not important. It is necessary to thoroughly trace the discharge position with respect to the nozzle for tubes of both types. When the power in the discharge increases, it shifts to the front end of the discharge tube, resulting in the nozzle burning-through, and the discharge escapes outside. The table summarizes a report of studied gases and mixtures for inducing plasma and lists the maximal gas pressure values, which were reached at the said power for tube *I*.

The table does not contain data on plasma in argon, since, in this case, as was earlier noted, the discharge is very easily initiated and it is possible to support it at a pressure of several atmospheres even at 20 W. It can be seen from the table that the surfatron is intended to sustain stable plasma at pressures exceeding the atmospheric one for all studied gases and mixtures. The exclusion is pure hydrogen, which, as is known, is the most difficult object for initiating and sustaining plasma. However, even in this case, a high pressure was reached at a relatively low applied power.

The analysis of the obtained dependences for the optimal tuning and data allows one to conclude as follows:

(i) for studied gases and mixtures at different pressures and different applied powers, the fine tuning allows one to reach a reflected power that does not exceed 0.5-2.0%;

(ii) in these conditions in the discharge, the optimal tuning is quite critical to shifts of a corresponding element and requires their thorough installations; and

(iii) design solutions aimed at increasing the tuning accuracy ensure the required accuracy in searching for the optimal position.



Fig. 6. Efficiency of dissociation of H_2 for a discharge in a 6% H_2 -He mixture as a function of the mixture pressure. The power in the discharge is 30 W.

As an example of using the described surfatron version as a source of hydrogen atoms, Figs. 5 and 6 show dependences of the degree of dissociation of hydrogen as a function of the gas pressure in pure hydrogen and 6% mixture of H₂ in He. Discharge tube *I* with cooled compressed air transmitted through the thermostat for decreasing its temperature down to 2°C was used. The highest degree of dissociation was reached at a 15-W minimal power in the discharge for H₂ and a 30-W minimal power for the mixture almost at the zero reflected power.

The dependences illustrated in Figs. 5 and 6 and pressure dependences of the hydrogen-atom beam intensity, obtained from the time-of-flight spectrum maxima (Figs. 7 and 8), show an important regularity. In spite of the fact that the degree of dissociation decreases as the gas pressure increases, the beam intensity rises both for the discharge in pure hydrogen up to $p_0 \cong 19$ Torr and for the discharge in the mixture at $p_0 \cong$ 300 Torr and further drops insignificantly as p_0 increases in the studied pressure range. Taking into account that the beam particle velocity distribution narrows as the pressure increases, it seems optimal to use a seeded beam and, for this mixture composition, operate at a 300- to 400-Torr pressure. It is necessary to note that a high degree of dissociation of hydrogen (>70%) was also obtained in the microwave surface discharge in [23]. However, the data given in this work relate to a discharge in a flow corresponding to $p_0 \sim 0.76$ Torr with the further selection of atoms by the nozzle cooled by liquid helium. In this case, the minimal power applied into the discharge at a high degree of dissociation was 400 W.

Intensity of atom beam, arb. units



Fig. 7. Dependence of the relative beam intensity of hydrogen atoms on the hydrogen pressure for a discharge in pure H_2 . The power in the discharge is 15 W.

CONCLUSIONS

The test results of the designed microwave discharge source, in which the discharge is initiated by a surface electromagnetic wave, show that it is possible to sustain plasma for various gases and mixtures at pressures of up to several atmospheres at powers of no more than 115 W. An almost zero level of the reflected power is due to the fine adjustment of tuning elements. The preliminary tests of the unit as a source of hydrogen atoms showed a high degree of dissociation (80%)for both pure hydrogen (at p_0 of up to 6 Torr) and a mixture of H_2 with helium (at p_0 of up to 50 Torr) at a low power inputted to the discharge (15 W for pure H_2 and 30 W for a 6% mixture of H_2 in He). In spite of a decrease in the efficiency of dissociation with a further increase in p_0 , the effective beam intensity increases in the both cases owing to a change in the ratios of atomic velocities in the beam, granting good possibilities for generating supersonic atomic hydrogen beams.

ACKNOWLEDGMENTS

We are grateful to E. Pick for assistance in performing tests and Deutsche Forschungsgemeinschaft (DFG) for supporting this work.

REFERENCES

- 1. Miller, D.R. and Patch, D.F., *Rev. Sci. Instrum.*, 1969, vol. 54, p. 1566.
- Gorry, P.A. and Grice, R., J. Phys. E: Sci. Instrum, 1979, vol. 12, p. 857.
- 3. Sibener, S.J., Buss, R.J., Ng C.Yiu, and Lee Yu.T., *Rev. Sci. Instrum.*, 1980, vol. 51, p. 167.
- 4. Wilsch, H., J. Chem. Phys., 1972, vol. 56, no. 3, p. 1412.
- 5. Silvera, I.F. and Walraven, J.T.M., *Phys. Lett.*, 1979, vol. 74A, p. 193.

Intensity of atom beam, arb. units



Fig. 8. Dependence of the relative beam intensity of hydrogen atoms on the mixture pressure for the discharge in a 6% H₂–He mixture. The power in the discharge is 30 W.

- 6. Hershcovitch, A., Kronou, A., and Niinikoski, T.O., *Rev. Sci. Instrum.*, 1987, vol. 58, no. 4, p. 547.
- Ding, A., Karlau, J., and Weise, J., *Rev. Sci. Instrum.*, 1977, vol. 48, no. 8, p. 1002.
- 8. Sleven, J. and Stirling, W., *Rev. Sci. Instrum.*, 1981, vol. 52, no. 12, p. 1780.
- Moison, M., Zakrzewski, Z., and Pantel, R., J. Phys. D: Appl. Phys., 1979, vol. 12, p. 219.
- 10. Moison, M. and Zakrzewski, Z., J. Phys. D: Appl. Phys., 1991, vol. 24, p. 1025.
- 11. Boisse-Laporte, C., Granier, A., Dervisevic, E., et al., *J. Phys. D: Appl. Phys.*, 1987, vol. 20, p. 197.
- 12. Daviaud, S., Boisse-Laporte, C., Leprince, P., and Marec, J., J. Phys. D: Appl. Phys., 1989, vol. 22, p. 770.
- Granier, A., Pasquiers, S., Boisse-Laporte, C., et al., J. Phys. D: Appl. Phys., 1989, vol. 22, p. 1487.
- 14. Granier, A., Chereau, D., Henda, K., et al., *J. Appl. Phys.*, 1994, vol. 75, p. 64.
- 15. Chave, C., Boisse-Laporte, C., Marec, J., and Leprince, P., *Mater. Sci. Eng. A*, 1991, vol. 140, p. 494.
- 16. Ricard, A., Barbeau, C., Besner, A., et al., *Can. J. Phys.*, 1988, vol. 66, p. 740.
- 17. Rousseau, A., Granier, A., Gousset, G., and Leprince, P., J. Phys. D: Appl. Phys., 1994, vol. 27, p. 1412.
- 18. Tomasini, L., Rousseau, A., Gousset, G., and Leprince, P., J. Phys. D: Appl. Phys., 1996, vol. 29, p. 1006.
- Granier, A., *Microwave Discharges: Fundamentals and Applications*, Ferreira, C.M., Ed., New York: Plenum, 1993, p. 491.
- 20. Ricard, A., Besner, A., Hubert, J., and Moison, M., J.Phys. B: At. Mol. Opt. Phys., 1988, vol. 21, p. L579.
- Callede, G., Deschamps, J., Godart, J.L., and Ricard, A., J. Phys. D: Appl. Phys., 1991, vol. 24, p. 909.
- 22. Selby, M. and Hieftje, G.M., *Spectrochim. Acta B*, 1987, vol. 42, p. 285.
- 23. Koch, N. and Steffens, E., *Rev. Sci. Instrum.*, 1998, vol. 70, p. 1631.