

Abstract

Experimental investigations of microwave plasma sources for gas conversion
at atmospheric pressure

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Microwave plasma sources are devices that use the energy of the microwave frequency electromagnetic field to generate plasma. Microwave plasma is a non-equilibrium plasma characterized by high electron concentration and energy at a relatively low gas temperature. At the same time, its parameters (electron concentration, gas temperature) can be adjusted over a wide range by adjusting the microwave power delivered to the discharge. Microwave discharges are very stable and repetitive. Properly designed microwave plasma sources ensure nearly one hundred percent energy transfer from the electromagnetic field of microwaves into the plasma. One of the more significant practical advantages of microwave plasma is that this type of plasma can be generated in gases at atmospheric pressure and without the use of electrodes. This avoids the use of expensive vacuum systems thereby simplifying the plasma source and reducing the cost of plasma generation. It is also possible to obtain high-purity plasma free from contamination with electrode material, thus avoiding the cost of electrodes regeneration and replacement. Due to its specific features atmospheric pressure microwave plasma has already found numerous applications for several decades. It can be used for example in surface treatment, carbon nanotubes synthesis, sterilization and disinfection and biomass gasification.

One of the growing interests is to use the atmospheric pressure microwave plasma in gas conversion. The light electrons present in the gas, as a result of transferring the energy of the electromagnetic field of microwave frequency to them, collide with gas molecules, which lead to their excitation, ionization and dissociation. As a result, new electrons and ions, atoms and molecules in an excited state, and radicals are formed, respectively. In this way, highly reactive chemical species are formed, which enter into numerous reactions resulting in the formation of new plasma components. Therefore, plasma gas conversion leads to changes in the physical and chemical properties of plasma components, including the change of some plasma components into others. Because microwave plasma is a source of active chemical

species that increase the rate of chemical reactions, expensive and contamination-sensitive catalysts can be avoided in gas conversion.

According to the above, this doctoral dissertation presents the results of experimental investigations of an atmospheric pressure microwave plasma sources for gas conversion.

In the first part of the dissertation author's own research, investigations on the influence of discharge conditions and supply conditions on the tuning characteristics of the microwave plasma source of surfaguide type were conducted. In this type of source, the plasma of a form of a column is generated inside a dielectric discharge tube, and the surfaguide itself is commonly used for gas conversion, e.g. carbon dioxide and for hydrogen production from gaseous substances (such as methane). The results obtained indicate that the surfaguide's tuning characteristics are significantly affected by the height of the reduced-height section, the diameter of the coupling gap and the presence of a metal cylinder shielding the plasma column, while these characteristics are not very sensitive to changes in the incident wave power, working gas flow rate and to the presence of liquid cooling of the discharge tube.

In the second part of the dissertation, the qualitative and quantitative estimation of the power radiated by a microwave plasma source of TIAGO type was proposed. Similar to surfaguide, TIAGO is used for gas conversion, however the plasma generated in it has a shape of flame formed at the tip of a conical metal nozzle. The results showed that the value of radiated power from the unshielded TIAGO is more than 30% of the power entering the discharge. Due to such high power losses, the danger to people and the environment and the negative impact of this phenomenon on plasma stability, it is therefore necessary to use metal shields that shield the plasma source. The research carried out showed that such a metal tube plays a shielding role, does not have to be closed at the top and its radius must be smaller than the critical radius (cut-off radius) for a circular waveguide operating at a given microwave operating frequency. The use of a tube with a radius greater than the cut-off radius not only does not assure shielding, but on the contrary causes an increase in the power radiated by the plasma source.

In the next two parts of the dissertation, the results of experimental investigations on hydrogen production by the plasma conversion of methane and vapours of liquid substances are presented. The studies were carried out using a surfaguide type microwave plasma sources powered by microwaves at a frequency of 2.45 GHz and 915 MHz and operating at atmospheric pressure. No catalyst was used in the studies. In the case of plasma methane conversion, such processes as pyrolysis, dry reforming and combined steam reforming were investigated. The highest values of hydrogen production rate and energy yield of hydrogen

production were obtained for combined methane steam reforming and amounted to 192 g(H₂)/h and 43 g(H₂)/kWh, respectively. In this case, the energy yield of hydrogen production is competitive compared to other plasma methods of hydrogen production using, for example, dielectric barrier discharge and arc discharge. In the process of plasma conversion of liquid vapors, ethanol, isopropanol and kerosene were used as hydrogen substrate. In the study, the highest value of the energy yield of hydrogen production of 22 g(H₂)/kWh was obtained in the case of plasma conversion of mixture of ethanol and water vapours lifted to the plasma by nitrogen gas. A comparison of the energy yield of hydrogen production for different plasma methods of hydrogen production from different liquids showed that there are solutions that provide higher values of this quantity. However, the almost one hundred percent degree of conversion of the liquid substances used makes the proposed method using microwave discharge plasma potentially competitive with other plasma methods of hydrogen production.