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Plasmas for ISRU on Mars: fuels, life-support and agriculture

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Abstract

In this work we address the possibility of oxygen production from the CO_2-enriched Martian atmosphere using plasma technology. A local production of oxygen from CO_2 decomposition would reduce the logistics and costs of future missions, while providing a breathable environment for future human outposts, a source of rocket propellant and feedstock and base chemicals for building materials and fertilizers. A combination of experimental and modelling efforts is used to explore different plasma sources. Diversity of different plasma sources allows exploring different energy transfer pathways leading to CO_2 dissociation, including direct electron impact processes, plasma chemistry mediated by vibrationally and electronically excited states, and thermally driven dissociation. Experimental results are validated against volume average 0D self-consistent kinetic models accounting in detail for the very complex plasma chemistry. The model describes the kinetics of electrons and heavy species under Martian environment in terms of pressure and temperature, relying on the LisbOn Kinetics (LoKI) simulation tool to solve the homogeneous two-term electron Boltzmann equation and the system of zero-dimensional rate balance equations for the most relevant charged and neutral species. We analyze simulated electron energy distribution function (EEDF) for DC glow discharges, operated under continuous regime, at a pressure of 5 Torr and discharge current of 50 mA, both for pure CO_2 and 96%CO_2/2%Ar/2%N_2 (synthetic Martian mixture). Moreover, we also demonstrate the versatility of plasmas to synthesize different compounds of interest to ISRU on Mars, including not only CO and O_2 but also NO_x products.

1. Introduction

Space exploration has fascinated mankind for decades, stimulating the interest in science and engineering, while contributing to the advancement of new technologies with crucial roles in improving everyday lives. At the same time, spaceflight programs are expanding towards the conquering and occupation of space. In this context, Mars is often regarded as the next step towards the voyage of humanity into the Universe. The achievement of a manned mission to Mars will mark the next frontier of discovery and the dawn of a new age in planetary exploration. To achieve this endeavour, the harvesting of local resources at the site of exploration is of a special importance. This approach is known as in situ resource utilization (ISRU) and it has the potential to enable humans to thrive beyond Earth, in a self-sufficient way, for extended periods of time. ISRU on Mars becomes particularly relevant when considering the possibility of oxygen production directly from the CO_2-enriched Martian atmosphere. A local production of oxygen would reduce the logistics and costs of manned missions, while providing a breathable environment for future human outposts and a source of rocket propellant. Note that in a typical rocket fuel (e.g., methane-oxygen fuel), the oxygen accounts for 75-80% of the total propellant mass. Integrating an ISRU system into the strategy of future missions to

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Mars could potentially save tens of billions of euros [1]. Exploring such technology would equally promote the development of new methods targeted at CO_2 recycling on Earth, which is one of the great challenges for the 21st century. Until today, the only concrete proposal for oxygen production on Mars is the inspiring MOXIE experiment under the framework of the Mars-2020 rover mission [2]. It is based on solid oxide electrolysis cell technology, in which electricity is provided between two electrodes to electrochemically convert CO_2 into oxygen at temperatures above 1000 K and pressures of about 1 bar. MOXIE has been operating successfully on Mars, while producing pure oxygen (since 18 Abril 2021) with 99.6% purity [3]. In [3] the authors also provide results from an optimization study of a human-scale MOXIE-type system to account for a 6–person Mars Ascent Vehicle. In addition to the issue associated to the scaling, MOXIE has some practical challenges, namely related to the required high operating temperatures and pressures, which bring concerns for lifetime and degradation of electrodes. Indeed, electrode materials are susceptible to degradation under reduction oxidation cycles and carbon deposition, requiring protective CO atmosphere and operating conditions to maintain stable operation. Particularly, under the Martian scenario, the need for high temperatures calls for a careful design of the thermal insulation system. It would be then of special interest to explore a system that decreases reliability concerns associated with electrodes and operation at high temperatures, while providing an efficient CO_2 decomposition with the versatility to produce products of interest for ISRU. In this work we explore the possibility of converting CO_2 into oxygen under Martian conditions through non-thermal plasma technology. The goal is to build and investigate a suitable plasma-based device to produce oxygen under realistic Martian environment. Note that non-thermal plasmas are highly reactive gas mediums sustained by electrical discharges that allow the coexistence of energetic electrons with relatively cold gas molecules. Under these conditions, far from thermodynamic equilibrium, it is possible to channel the discharge power towards chemical reactions of interest. Over the past years, non-thermal plasmas have gained much attention regarding CO_2 decomposition due to their potential to activate CO_2 at reduced energy cost and excite CO_2 vibrations that efficiently contribute to overcome the dissociation barrier. This has led to a growing field of research aimed at combining renewable electricity with plasmas to convert CO_2 emissions into synthetic fuels for energy storage pathways (see e.g., Pietanza et al. [4] and references therein). As a direct outcome of the extensive investigation addressing plasma-based CO_2 decomposition and the production of solar fuels on Earth, it was recently noted that the knowledge acquired on Earth can be transferred to some extent to ISRU on Mars.

This paper is organized as follows: in **section 2** we provide an overview on the potential of plasmas for ISRU on Mars. We discuss recently published results while addressing current challenges associated with ISRU with plasma technology. These challenges are mainly associated to gas separation issues required to extract and separate oxygen. In **section 3** we focus our attention on the description of the CO_2 plasma chemistry, while analysing different decomposition channels associated to the production of valuable products, namely oxygen, CO and NO. Modeling results are compared against experimental data with special focus on DC glow discharges. Finally, **section 4** summarizes the main results and concludes the paper.

2. State of the art

The very strong case supporting plasma-based production of oxygen on Mars presented by Guerra et al. [5] predicted theoretically that the Martian atmospheric conditions of pressure, temperature, and gas composition are very favourable to ignite a plasma system and to achieve efficient CO_2 decomposition under a non-thermal regime. It was demonstrated that the cold temperatures of Mars can preserve the asymmetric vibrations of CO_2, which accumulate energy for the decomposition of CO_2 and subsequent oxygen production. These results clearly anticipate that, under Martian conditions, the CO_2 decomposition can be strongly induced by CO_2 vibrational excitation because of the pumping of vibrational quanta towards the dissociation limit, thus providing an efficient way to produce oxygen. The feasibility of oxygen production on Mars through plasma technology was also corroborated by two experimental campaigns. First, a DC glow discharge was cooled down to Martian temperatures by inserting a plasma reactor inside a cold bath of dry ice and ethanol [6]. The cold Martian temperatures did increase the CO_2 vibrational excitation degree, while the reactor provided dissociation fractions in the range of 10-30%. These results are very encouraging considering that this plasma setup was designed for fundamental research and far from suited to the development of a prototype. A second experimental study [7], developed under the framework of a master thesis between Instituto Superior Técnico in Portugal and the Dutch Institute for Fundamental Energy Research in the Netherlands showed that microwave discharges, operated under Martian mixtures, can provide CO_2 conversions of about 35% with roughly 300 W of power. Furthermore, it was estimated that oxygen can be produced at the rate of 14 g/h using a 4 kg reactor with 25x20x5 cm dimension (including main components and shielding). These values are extremely promising in the context of ISRU and further demonstrate the potential of microwave discharges for CO_2 decomposition at low pressure conditions. Considering that this second experimental campaign did not explore the effect of Martian temperatures or any type of fine tuning, the values obtained on CO_2 conversion also reveal large room for improvement. Overall, these studies clearly justify further theoretical and experimental research targeted at exploring CO_2 decomposition under Martian conditions. On one hand, there is still much ground to be covered in relation to the contribution of CO_2

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vibrational excitation (induced by cold Martian temperatures) for an effective CO_2 decomposition. On the other hand, the transition from the early stages of fundamental research to a prototype phase requires the creation of a dedicated experimental framework on which the challenges of ISRU (gas separation, weight requirements, dust removal, etc.) can be explored in detail.

Once the CO_2 decomposition is achieved, it is worth noticing that the extraction of oxygen from the decomposed CO_2 mixture presents a formidable challenge. Gas separation, the process of isolating specific gases from a mixture, becomes crucial to obtain high-purity oxygen. In this context, separating the desired oxygen molecules from the mixture that contains CO, trace impurities, and other residual gases becomes a complex task. For the case of plasma operated in pure CO_2, the products are always a mixture of CO_2, CO, and O_2, necessitating then means of separating these products for further utilization. Although conversion and separation processes can be conducted independently, there is a potential for synergistic effects when they are integrated and operated together. Just as introducing a catalytic surface into a plasma system can influence the distribution of reaction products, the inclusion of an oxygen-permeable membrane surrounding the plasma can induce a beneficial shift in the equilibrium of the CO_2 dissociation reaction, leading to higher concentrations of carbon monoxide (CO) and oxygen (O_2). There are three distinct categories of oxygen-permeable membranes, each with its own characteristics and transport mechanisms:

1. Non-electrochemical membranes: These materials facilitate oxygen transport without relying on charged species. A notable example is silver, which possesses a high diffusion coefficient for atomic oxygen, particularly along grain boundaries.
2. Mixed ionic-electronic conductors (MIECs): This class of materials exhibits electrical conductivity for both electrons and oxygen ions. The relative conductivity of these two species can vary significantly depending on factors such as the specific material used and the operating conditions (e.g., temperature and oxygen partial pressure). Perovskite materials tend to exhibit higher electron conductivity compared to other MIECs.
3. Pure oxygen ion conductors: These materials predominantly exhibit electrical conductivity through the movement of oxygen ions within the material's lattice structure. For a material to be classified as a pure ionic conductor, its ionic conductivity must be at least two orders of magnitude greater than its electronic conductivity. Achieving significant electrical conductivity in pure oxygen ion conductors typically requires high operating temperatures due to substantial energy barriers for atomic diffusion through the lattice.

Several works addressing the coupling of the previously mentioned materials with plasma reactors are available in literature. A notable illustration of a non-electrochemical membrane (first gas separation method above) specifically tailored for Mars ISRU purposes can be found in the research conducted by Outlaw and colleagues [8]. In their work, they focused on the development of a membrane utilizing silver due to its selective affinity for oxygen species. The study involved investigating the thermally activated behaviour of the diffusion coefficient of oxygen atoms within silver across a temperature range spanning from 673 K to 1073 K. Furthermore, they explored the integration of these membranes with both O_2 and CO_2 glow discharges. In case of MIECs (second method above) a hollow fiber mixed conductor (perovskite-type, La0:6Ca0:4Co0:5Fe0:5O3δ or LCCF: a dominantly electronic conductor) was placed in the effluent of a CO_2 microwave plasma torch by Chen et al [9]. The interior of the Fiber was continuously flushed by pure Ar, ensuring the highest possible O_2 partial pressure to enhance separation. Also, with the purpose of achieving oxygen separation in CO_2 plasmas, a mixed conductor (La0:6Sr0:4Co0:3Fe0:8O3δ, or LSCF, another dominantly electronic conductor) of 1 mm thickness was used as an end cap on a cylindrical Dielectric Barrier Discharge (DBD) by Zheng et al. [10]. Synergetic effect between membrane and the plasma was observed when the reactor was operated at 15 W in air with temperatures of about 873 K. Finally, in case of oxygen separation through ionic conductor systems, it is worth mentioning experiment of Mori and Tun [11]. They used a hybrid system consisting of a dielectric barrier discharge (DBD) and a solid oxide electrolysis cell (SOEC). The DBD was operated on top of the SOEC, where yttria-stabilized zirconia (YSZ) served as the electrolyte. The exterior electrode of the SOEC functioned as the ground electrode for the DBD, and power was independently supplied to both devices. To facilitate efficient oxygen ion conduction, the entire assembly was placed inside an oven, allowing the YSZ to reach a high temperature of approximately 700 K. This elevated temperature enabled significant oxygen ion conduction across the cell when a voltage difference of 5-10 V was applied. By integrating this hybrid system, the researchers demonstrated a remarkable enhancement in CO_2 conversion. The maximum conversion rate, which previously reached 40% without the SOEC, was substantially increased to 90% with the presence of the SOEC under the applied power. Overall, these studies have already demonstrated the feasibility and potential of these membranes to separate oxygen from gas mixtures, including the decomposed CO_2 obtained from plasma-based CO_2 dissociation. The coupling of plasmas with gas membranes for oxygen separation represents a growing research field with numerous ongoing experiments. The utilization of MIECs and ion conductors holds great promise in enhancing the efficiency and performance of plasma-gas membrane systems. The imminent results from recent experiments are expected to contribute significantly to the scientific community's understanding of oxygen separation techniques, enabling advancements in ISRU for oxygen production on Mars. More details about the potential to oxygen separation in plasmas can be found in the perspective

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paper recently published [12]. In parallel with gas separation studies, it is also imperative to develop a comprehensive understanding of the plasma chemistry of Martian plasmas. This necessitates conducting research that encompasses both experimental campaigns and modelling work dedicated to the understanding of the main mechanisms responsible for CO_2 decomposition at low pressure (~ 5 Torr) conditions and low gas temperatures (<500 K). In the following section, we address this crucial aspect and emphasize the significance of exploring plasma chemistry.

4. Plasma chemistry results

An understanding of plasmas sustained under Martian conditions necessitates a thorough comprehension of various kinetics, including vibrational, chemical, and electronic interactions involving the constituent species present in the Martian atmosphere, such as CO_2, N_2, and Ar, as well as the by-products formed within the reactor. Developing and experimentally validating self-consistent models plays a crucial role in gaining insights into the interconnected kinetics within these plasmas, particularly for quantities that are challenging to measure directly. In this study, we employed an average 0D self-consistent kinetic model to describe the dissociation of CO_2 in plasmas sustained under Martian conditions. This model accounts for the intricacies of plasma chemistry in detail and is based on the work initially developed by Ogloblina et al. [6]. In [6] the modelling results were compared against a DC glow discharge ignited in a 2 cm inner-diameter cylindrical Pyrex tube with pressures of about 5 Torr and gas flow rates of about 10 sccm. The model utilizes the LisbOn KInetics (LoKI) simulation tool [13] and solves the electron Boltzmann equation with two terms and the system of zero-dimensional (0D) rate balance equations, which provide volume-averaged representations, for the most important charged and neutral species. In [6] the simulation outcomes exhibited excellent agreement with experimental data (in terms of species densities, gas temperature, vibrational temperature, etc.), while revealing that the pressure and temperature conditions found on Mars can amplify the extent of vibrational nonequilibrium and increase oxygen decomposition given the higher degree of electron impact dissociation. To illustrate this point, we show the simulated electron energy distribution function (EEDF) for a DC glow discharge operating continuously at a pressure of 5 Torr and a discharge current of 50 mA (see figure 1). These experimental conditions are based on the operating conditions of the DC glow discharge described in [6]. The EEDF is presented for both pure CO_2 and a synthetic Martian mixture consisting of 96% CO_2, 2% Ar, and 2% N_2. The gas temperature considered in the simulations are based on experimental measurements. The EEDFs exhibit a notable enhancement in the high-energy tail for the Martian mixture compared to pure CO_2. This enhancement primarily arises from the disparities in the cross-sections of the different gases incorporated in the model and the consequential modifications in the self-consistently sustained reduced electric field. Specifically, the sustained electric field increases from 59 Td in pure CO_2 to 67 Td in the Martian CO_2/Ar/N_2 mixture. As the threshold for electron-impact dissociation exceeds 6 eV, this effect justifies the observed augmented dissociation in a Martian atmosphere as compared to a pure CO_2 discharge, as reported in [6]. In the present conditions, CO_2 dissociation proceeds mainly by direct electron impact on CO_2 ground-state and vibrationally excited molecules via:

$$e + CO_2(\nu_1\nu_2^{l2}\nu_3) \rightarrow CO(X) + O(^1D), \quad (1)$$

where the CO_2 vibrationally excited levels (00^00), (01^10), (02^20), and (10^00 + 02^00) have the most important contributions.

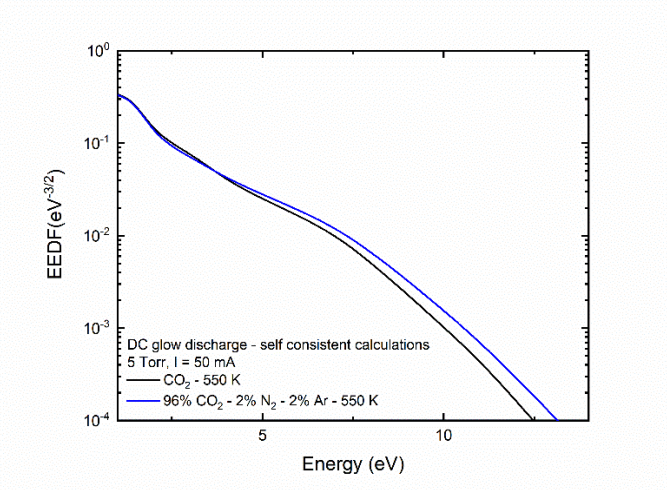

Figure 1 Electron energy distribution functions calculated for DC glow discharges at p = 5 Torr and I = 50 mA for pure CO_2 and for 96%CO_2/2%Ar/2%N2 (Martian mixture) at 550 K temperature.

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More details about the contribution of different levels on the CO_2 conversion under DC glow discharge conditions can be found in [6]. Moreover, in addition to electron-impact reactions such as (1), it is worth mentioning, that under our conditions, electronically excited molecules such as CO(a) can also have an important contribution to the overall CO_2 decomposition. Indeed, the presence of CO(a) can promote CO_2 formation via bimolecular reactions:

$$CO(a) + O_2(X) \rightarrow CO_2 + O(^3P). \quad (2)$$

The effect of electronically excited CO(a) on CO_2 decomposition has been analyzed in detail in [15]. At the same time, it is worth mentioning that for small O_2 concentrations, CO_2 dissociation can be stimulated through the bimolecular reaction:

$$CO(a) + CO_2 \rightarrow 2CO + O(^3P), \quad (3)$$

attesting the complex role that this electronically excited state can play in the overall kinetics. This point also reveals the importance of removing the oxygen from the gas mixture to enhance the CO_2 conversion. Indeed, the detrimental effect of oxygen due to recombination mechanisms is very well known and it has been reported in literature (see e.g. [3]). In addition to the presented findings, we also provide the calculated concentrations of the primary neutral species generated within the plasma under the previously mentioned experimental conditions (see **figure 2**). Here we can clearly observe the versatility of plasma sources for ISRU given the possibility of producing valuable species (CO, O_2, NO, etc.). Indeed, these plasma sources can be employed to synthesize diverse compounds by utilizing different gases, thereby enabling the adaptation of the same power sources and principles outlined for CO_2 decomposition to various gas-conversion applications. In relation to the model initially published in [5], it is important to notice that for the results shown in figure 2, the CO_2 plasma chemistry has been updated (see details in [14]) in order to include: (i) vibrational exchanges between N_2 and CO, (ii) quenching of CO and CO_2 vibrations by oxygen atoms, (iii) V–V and V–T exchanges involving CO molecules (iv) vibrational exchanges between CO and CO_2, (v) several electron impact, vibrational energy exchanges, and reactions involving N_2 and Ar, and (vi) chemistry associated to the production and loss of NO and NO_2 and N_2O species. In relation to this last point, it is worth noticing the low amount of NO_x species reported/calculated in figure 2 (about 0.03% of total final mixture). This low amount of NO_x is the result of the low concentration of N_2 considered in the initial gas mixture (2% under Martian conditions). Higher concentrations of NO_x are usually obtained in N_2-O_2 plasmas. N_2-O_2 plasmas have been extensively studied in the past (see e.g. [16]) motivated by applications associated to the production of fertilizers and nitrogen fixation. These processes are extensively pursued in agriculture and the food industry on Earth. Similarly, on Mars, a similar adaptation can be anticipated, utilizing nitrogen from the Martian atmosphere. Indeed, and given a plasma reactor coupled with gas separation membrane, higher concentrations of NO_x (compared to the values shown in figure 2) could be achieved by exposing the extracted oxygen to activated N_2.

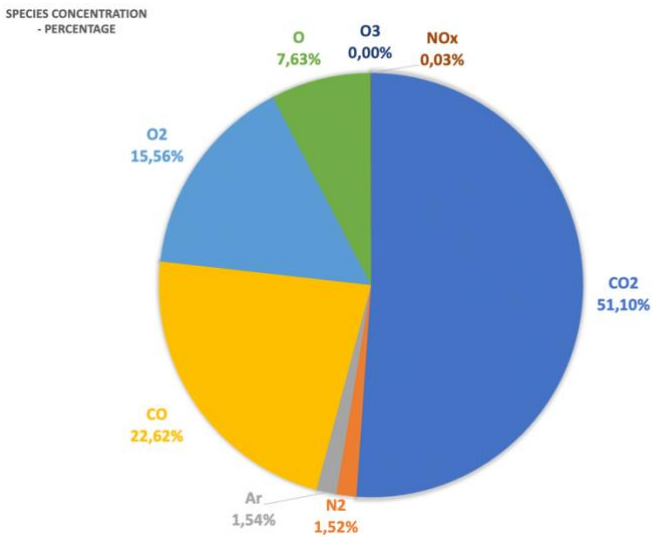

Figure 2 Chart with calculated concentrations for the main species in the plasma in a DC glow discharge operating at p = 5 Torr and I = 50 mA for a 96%CO_2/2%Ar/2%N_2 gas mixture. Concentration of species shown in this chart include the contributions of ground and electronically excited states.

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To further explore the possibility of forming NO_x products from Martian plasma we simulated the concentrations of NO and N_2O obtained in N_2-O_2 plasmas (see figure 3). The study of N_2-O_2 plasmas under temperatures and pressures of Mars consists of a first step towards the validation of the plasma chemistry of Martian plasmas targeted at producing NO. To simulate N_2-O_2 plasmas under Martian conditions, while quantifying the production of valuable products, we relied on the same simulation tool as described in [5] and the plasma chemistry developed in [14]. Modeling results were then compared against experimental data available in N_2-O_2 plasma literature [17]. In this paper, the authors ignited N_2-O_2 discharges with a reactor composed of an inductive coil connected to a radio frequency power supply and a matching network and a quartz tube with a heating mantle. Experimental conditions involving a porous Pt film deposited on a tubular YSZ membrane (not studied in this work) was also explored in [17]. The comparison between modelling and experimental data in terms of NO production is shown in figure 3. Overall, we obtained a good agreement on the NO trend as function of the oxygen dilution, which provides a first step towards a validation of the N_2-O_2 chemistry under Martian conditions. These results also demonstrated the importance of vibrational excitation for the formation of NO. More specifically, NO is mainly formed in collisions of O atoms with vibrationally excited $N_2(X^1\Sigma_g, v> 12)$ or metastable states $N_2(A^3\Sigma_u^+)$, via:

$$N_2(X^1\Sigma_g, v> 12) + O \rightarrow NO(X) + N(^4S), \quad (4)$$

$$N_2(A^3\Sigma_u^+) + O \rightarrow NO(X) + N(^2D), \quad (5)$$

Other NO formation processes (e.g., involving collisions between N atoms and O_2) were considered (see details in [13]) but they were much less efficient compared to (4) and (5), partly because of the lower density of N atoms compared with O (typically, the N atomic density is about 10 times lower than that of O). The decrease of N atoms density with the increase of the O_2 dilution is also responsible for the fast increase of NO production observed in figure 3 around $[O_2]/([O_2] + [N_2]) \sim 0.01$. A future publication targeted at describing in detail the various volume and surface mechanisms associated with production of NO and N_2O in plasmas operated under Martian conditions (also including the presence of CO_2) is now in preparation.

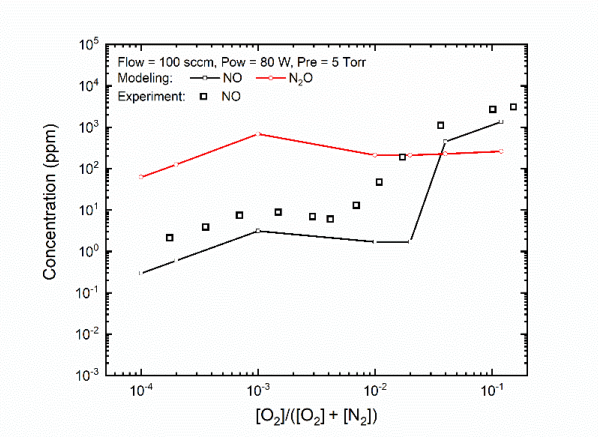

Figure 3 Calculated concentrations (in ppm) of NO and N_2O obtained with LoKI simulation tool, assuming a total flow of 100 sccm and pressure of 5 Torr, for different initial mixtures of O_2 - N_2. Experimental points are taken from reference [17]. Experimental error (based on standard deviation of different measurements) can be about 5%.

Conclusion

In situ resource utilization (ISRU) is an exciting research topic and it has the potential to enable the production of propellants, fuel cell reactants, and life support resources (such as water, oxygen, and buffer gases) from celestial bodies like the Moon and Mars. In this paper we discussed the benefits of plasma based ISRU for decomposition of CO_2 in the production of oxygen on Mars, which can be used for various purposes, including propellant production and the creation of fertilizers for agriculture. This paper presents an overview of the current advancements in plasma based ISRU technologies, specifically for decomposing CO_2 and producing oxygen on Mars. The effectiveness of plasma technology in ISRU is reliant on integrating plasma with separation membranes. The emerging plasma-membrane technology discussed here opens up new possibilities for a versatile electrochemical conversion technology that can be employed in different Earth environments and various ISRU applications. This paper also examines recent research on modeling of CO_2 plasma chemistry on Mars. Modelling results were compared against existing experimental data in the literature, while validating different plasma chemistries associated to 96%CO_2/2%Ar/2%N_2 and O_2-N_2 plasma mixtures.

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