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The role of helium metastable states in radio-frequency driven helium–oxygen atmospheric pressure plasma jets: measurement and numerical simulation

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Abstract

Absolute densities of metastable He(2\textsuperscript{3}S\textsubscript{1}) atoms were measured line-of-sight integrated along the discharge channel of a capacitively coupled radio-frequency driven atmospheric pressure plasma jet operated in technologically relevant helium–oxygen mixtures by tunable diode-laser absorption spectroscopy. The dependences of the He(2\textsuperscript{3}S\textsubscript{1}) density in the homogeneous-glow-like $\alpha$-mode plasma with oxygen admixtures up to 1\% were investigated. The results are compared with a one-dimensional numerical simulation, which includes a semi-kinetical treatment of the pronounced electron dynamics and the complex plasma chemistry (in total 20 species and 184 reactions). Very good agreement between measurement and simulation is found. The main formation mechanisms for metastable helium atoms are identified and analyzed, including their pronounced spatio-temporal dynamics. Penning ionization through helium metastables is found to be significant for plasma sustainment, while it is revealed that helium metastables are not an important energy carrying species into the jet effluent and therefore will not play a direct role in remote surface treatments.

1. Introduction

The rich non-equilibrium chemistry of cold atmospheric pressure plasmas (APPs) opens new opportunities for technological exploitation in controlled treatment of delicate materials, in particular in plasma medicine [1, 2]. Critical confining structures of cold APPs vary between some millimeters down to micrometer scales; hence they are often referred to as microplasmas [3–5]. Microplasmas are highly susceptible to non-linear instabilities initiated through mode transitions and associated variations in plasma ionization mechanisms [6, 7]. The comparatively high mobility of electrons in helium and its high thermal conductivity make helium a preferred gas for efficient and stable operation of cold APPs. For technological applications small admixtures of reactive molecular gases in the range of a per cent have been found to be most efficient [8, 9].

Excited metastable helium plays a key role in plasma ionization and sustainment mechanisms. The excitation energy of helium metastable states, 19.8 eV for He(2\textsuperscript{3}S\textsubscript{1}) and 20.6 eV for He(2\textsuperscript{1}S\textsubscript{1}), is significantly lower than the ionization energy of helium (24.6 eV). Consequently, helium metastables are produced more efficiently and can act as an important energy reservoir for subsequent Penning ionization of gas impurities or minority admixtures, such as nitrogen or oxygen [10]. Direct electron-impact ionization of impurity species, with lower ionization energy, can typically be neglected due to the much higher density of helium. In this work, we investigate the role of metastable helium in a technologically relevant micro-APP jet [11] with significant
admixtures of molecules in the per cent range using direct laser spectroscopic measurements and advanced numerical computer simulations.

2. The micro-APP jet

The μAPPJ produces a cold homogeneous α-mode glow plasma at ambient pressure [12, 13]. The mean electron energy ranges around 2 eV, the electron density up to \(10^{17} \text{ m}^{-3}\) [7]. The setup, shown in figure 1, consists of two plane parallel stainless steel electrodes. Quartz windows enclose the discharge region along the sides. The core plasma channel has a cross-section of 1 mm × 1 mm over a length of 30 mm. Typical operational parameters are a helium feed gas flow up to 2 slm (mean gas velocity of about 17 m s\(^{-1}\)) with a molecular oxygen admixture of 0.5%. One electrode is driven at 13.56 MHz by an rf broadband amplifier via an impedance matching network, while the other one is grounded. Species generated within the rf discharge volume are transported by the gas flow toward the effluent region, where they can be directed onto a surface to be treated. This concept differs from the frequently discussed dielectric barrier APPJ with high-voltage kHz-pulse excitation, where the production zone of the plasma at ambient pressure [12, 13]. The mean electron energy ranges around 2 eV, the electron density up to \(10^{17} \text{ m}^{-3}\) [7].

The operational rf power range depends on the flow and composition of the feed gas. Once ignited, the plasma can be sustained down to an rf generator power of ∼4.5 W for pure helium. Below an rf power threshold of ∼12 W for pure helium, the discharge operates in a homogeneous low-gas-temperature glow-mode, which is the α-mode, but with an increasing γ-mode-like contribution, explained below. Above threshold, the discharge changes to a spatially constricted mode of significantly higher current density, bearing the potential risk of electrode surface damage. The transition back to the α-mode occurs at a lower power due to a thermal hysteresis effect [7]. The operational range between sustaining limit and mode transition shifts to higher powers with increasing feed gas flow, showing that higher flow rate suppresses the thermal instability of the discharge to some extent. Addition of molecular oxygen also shifts the operational range toward higher power. This can be attributed to the energy consumption through molecular vibrational and rotational excitation as well as dissociative processes not directly contributing to the plasma ionization.

3. Laser absorption spectroscopy diagnostic method

For the tunable diode-laser absorption spectroscopy (TDLAS) measurement a commercial diode-laser system (Toptica, DL100 DFB) was used. Using the current modulation of the diode-laser, the system provides a mode-hop-free tuning range of about 30 GHz to scan the \(J = 1, 2\) components of the He(\(2^3\)P\(_j\) ← \(2^3\)S\(_i\)) triplet transition at \(\lambda_0 = 1083\) nm. The laser spectral width of <100 MHz is negligible compared with the several GHz width of the absorption line at atmospheric pressure. Two beam splitters were used to generate two secondary low intensity laser beams, as described in [16]. The main beam crosses the 30 mm long APPJ plasma channel for recording its absorption profile. One of the other two beams is used to calibrate the laser frequency with the aid of a 0.375 GHz free spectral range Fabry–Perot etalon, and the other beam crosses a low pressure helium discharge to provide a reference line profile. These three beams were detected by three photodiodes. The signals were recorded simultaneously and averaged (over 64 scans) in different channels of a digital sampling oscilloscope, triggered with the diode-laser modulation signal.

Absolute He(\(2^3\)S\(_i\)) densities were deduced according to the Beer–Lambert law:

\[
\frac{I_v}{I_0} = \exp(-A(v)) = \exp(-k(v)L)
\]

(1)

with \(I_v\) and \(I_0\) the transmitted and incident laser intensity at the laser frequency \(v\) respectively, \(A(v)\) the absorbance profile and \(L\) the absorption length. The generalized absorption coefficient \(k(v)\) comprises the observed fine-structure components \(i \rightarrow f\) with central line positions \(v_{fi}\) and a common normalized spectral line profile \(\Phi(v - v_{fi})\):

\[
k(v) = \sum_{f,i} k_{fi}^0 \Phi(v - v_{fi}).
\]

(2)

The individual absorption coefficients \(k_{fi}^0\) are given by

\[
k_{fi}^0 = \frac{g_f}{g_i} \frac{\lambda_0^2 A_{fi}}{8\pi} \langle n \rangle,
\]

(3)

where \(\langle n \rangle\) denotes the mean absorber density, \(g_f\) and \(g_i\) the statistical weights of the upper \(f\) and lower \(i\) states, respectively, and \(A_{fi}\) the transition probability [17]. A more detailed description can be found in [18].

An example of a measured absorption profile is shown in figure 2. The solid curve represents the best fit to the measuring...
Figure 2. Measured absorption profile (circles) of the He(2^3P_1,2 − 2^3S_1) transition at 2 slm He flux and 11 W rf power. Solid and dotted curves are best fits.

points with a superposition of two Voigt profiles (dotted curves), each for one fine-structure component, with 2.3 GHz spectral separation and 5 : 3 weighting according to theory, and a Doppler broadening $\Delta v_{\text{D}} = \frac{\sqrt{8 \ln(2)} k_B T_g}{M_{\text{He}}/2} \Delta v_{\text{D}}$ of 1.85 GHz, which corresponds to a gas temperature of $T_g = 345$ K, as measured spectroscopically [19]. Within the experimental uncertainties, a Lorentzian width of 13.0 GHz is found, independent of the operational parameters, e.g. independent of an oxygen admixture of up to 1%. Accounting for the $T_g^{0.7}$ dependence found in [20], this would lead to a linear pressure broadening coefficient of 18.9 MHz Torr$^{-1}$ at $T_g = 300$ K. This value is in good agreement with the reported value of 18.5 MHz Torr$^{-1}$ [21] obtained at room temperature.

A notable pressure shift of the resonances was not observed. The He(2^3S_1) detection limit is about $1 \times 10^{15}$ m$^{-3}$ due to small change of the diode-laser intensity when the rf generator is turned on to generate the plasma.

4. Experimental results

4.1. Helium without oxygen admixture

Figure 3 shows the results of an rf power variation for different helium feed gas flows. The helium metastable ($He^*$) density increases slightly overlinear with increasing rf power, from several $10^{15}$ m$^{-3}$ up to $1 \times 10^{16}$ m$^{-3}$ at the smallest 0.5 slm He flow, and up to $4.5 \times 10^{16}$ m$^{-3}$ at the largest 2.0 slm He flow. The trend of higher He* density at larger He feed gas flow saturates at about 1.5 slm. The overlinearity of the He* density dependence on the rf power indicates an increasing $\gamma$-mode-like contribution superimposed on a linear $\alpha$-mode contribution. We call it a ‘$\gamma$-mode-like’ contribution, since it is mainly due to electrons, generated through Penning ionization, which are subsequently accelerated by the high-voltage plasma sheaths during the sheath breakdown phase, rather than by fast secondary electrons [22]. It was possible to estimate the He* density in the constricted mode, despite the ill-defined absorption length, to be higher by more than one order of magnitude than in the homogeneous mode.

4.2. Helium with oxygen admixtures

Figure 4 shows the results of an rf power variation for different O$_2$ admixtures at 1 slm He feed gas flow. The maximum He* density drops from $2.2 \times 10^{16}$ m$^{-3}$ without molecular admixture down to $9 \times 10^{15}$ m$^{-3}$ with 0.5% O$_2$ admixture. One reason for the He* density decrease is that inelastic electron-impact collisions with molecular oxygen and perhaps ozone, produced in the plasma, reduce the electron temperature and hence the He* production rate. The other reason is the enhanced loss of He* through Penning ionization with O$_2$.

Figure 5 shows the measured He* density (circles) as a function of the O$_2$ admixture at 12 W rf power and 1 slm He feed gas flow. The variation is relevant for technological applications, since a density maximum for highly reactive oxygen atoms was found at about 0.5% O$_2$ admixture [8, 9]. The measuring points can be well described by a reciprocal dependence, the dotted curve in figure 5. This indicates (shown below) that the metastable helium atoms are mainly lost through Penning ionization with O$_2$.

Figure 4. Measured He($2^3S_1$) density as a function of the supplied rf power for different He feed gas flows.
intermediate O₂ admixtures continues toward lower degrees of admixture, where a change of the discharge characteristics is expected. In this regime, Penning ionization of He metastable atoms with gas impurities such as molecular nitrogen becomes an important process for the He* destruction as well as for the total ionization.

5. Numerical simulation

5.1. Comparison of measurement with numerical simulation

In the following, experimental results are compared with a numerical simulation of the plasma, in order to obtain further insight into the relevant He* formation mechanisms. The applied model is based on the time-dependent, one-dimensional numerical simulation [22] of the discharge gap. The reaction scheme, originally 116 reactions among 16 species, was revised and extended by four additional species (O₂(ν), O₂(ΣΣ), O₂(ΔΔ), and O₂(ΣΔ)) to 184 reactions among 20 species in total, as listed in the appendix. Most of the cross-section data and reaction rate coefficients were taken from Stafford and Kushner [23], which is related to the O₂(¹Δ) production in He/O₂ discharges for oxygen–iodine lasers [24, 25]. Here, the feed gas, 0.5% molecular oxygen in helium, is treated as constant homogeneously distributed background gas. A fluid approach is chosen to describe the transport of the other species, such as O and O₃ neutrals; He*, He₂*, O(¹D), O₂(¹Δ), O₂(¹Σ) metastables; He⁺, He₂⁺, O⁺, O₂⁺, O₂⁺, O₂⁺ ions; and O₂(ν) vibrationally excited oxygen molecules as total population over the first four vibration states. The electrons are treated semi-kinetically on the basis of a two-term approximation Boltzmann solver [26] to account for the strongly non-Maxwellian electron energy distribution in this kind of plasma [27, 28]. The boundary conditions include wall reactions according to [23] and secondary electron production, where a γ-coefficient of 0.1 and an initial energy of 0.5 eV for γ-electrons released from the electrodes due to the bombardment by positive oxygen and helium ions as well as by helium metastables is assumed.

Figure 6 shows the measured He⁺ density for 0.5% O₂ admixture at 1 slm He feed gas flow as a function of rf power.

The corresponding results of the numerical simulation are in good agreement. The averaged absolute difference is about 25%. The simulation yields a slightly weaker overlinearity than the measurement. A likely reason for this deviation is that the individual power axes are scaled, under the assumption of a linear correlation, to the ratio of the experimental rf supply power, at which the transition to the constricted mode is observed, versus the theoretical rf power, at which the simulated current–voltage characteristic exhibits the related turning point [22, 29]. The significant difference between both power axes indicates that most of the applied rf generator power is actually dissipated by thermal heating of the connecting cables, the matching network unit, and the electrodes, as well as through emission of rf radiation from the antenna-like electrode configuration itself. Another reason might be that the model underestimates the influence of the γ-mode, which is likely, in view of the large uncertainties for the γ-electron emission coefficient and the initial γ-electron energy.

5.2. Impact of metastable helium atoms through Penning ionization

The numerical simulation for 0.5% O₂ admixture yields information on the impact of the He* density on the total plasma ionization. O₂⁺ and O₃⁺ are found to be the dominant ion species. O⁺ is of minor importance, because of rapid conversion to O₂⁺ through the charge transfer reactions (R101)–(R103), see the appendix. The formation of O₂⁺ and O₃⁺ is strongly coupled through the ion conversion reactions (R109) O₂⁺ + O → O₂²⁺ + O, (R107) O₂⁺ + O₂⁺(¹Δ) → O₂²⁺ + O₂ and (R104) O₂⁺ + O₂⁺ + M → O₂²⁺ + M. Since (R104) represents the only O₂⁺ production channel, the main plasma ionization results through O₂⁺ production channels other than (R109) and (R107), so mainly through Penning ionization (R68) He⁺ + O₂ → O₂⁺ + e + He and electron-impact ionization (R12) e + O₂ → O₂⁺ + 2e. Figure 7 shows the O₂⁺ production through the main channels as a function of the rf power. The relative contribution
of Penning ionization to the total O$_2^+$ production, without considering (R109) and (R107), as explained, increases from 28% to 46% with increasing rf power. This reflects the He* density increase. The relative contribution of electron-impact ionization oppositionally decreases from 47% to 30%. The dependence of the total O$_2^+$ ion production on the rf power is found nearly linear, so similar as for the electron density, see [22], and less overlinear as for the He* density.

Because of this important role of He* in the plasma sustainment, the formation of metastable helium atoms was analyzed with the aid of the numerical simulation for 0.5% O$_2$ admixture. Electron-impact excitation from the ground state, e + He $\rightarrow$ He* + e, dominates the He* production by orders of magnitude in comparison all other possible two-body or three-body e + He$_2^+$ recombination channels. As illustrated in figure 8, the He* destruction mainly occurs through He* + M Penning ionization with the neutral oxygen species M = O, O$_2$, O$_2$(1$\Delta$), and O$_3$. Their relative contributions, e.g. the O$_2$ contribution decreasing from 82% to 79% with increasing rf power, simply reflect the weighting of the simulated reactant densities, since a common reaction rate coefficient was used.

Several conclusions for the He* density distribution along the discharge channel can be drawn: a rapid build-up over a short distance from the gas inlet, due to the fast electron-driven production. Then, over the main part of the channel, the He* density is expected to stay more or less constant, since the main destruction through Penning ionization only depends on the total neutral oxygen species density. Note that this total density is mostly independent of the evolution of the individual neutral oxygen species densities on the time scale of the slower chemical reactions. Another important implication is that our line-of-sight integrated absorption measurement actually yields a representative value of the local He* density over the main part of the discharge channel.

5.3. Dynamics of metastable helium atoms

The large contribution of Penning ionization to the total ion production implies that the metastable character of the He* atoms in the investigated atmospheric pressure plasma is significantly reduced; from the pure radiative lifetime of about 10$^4$ s down to an effective lifetime of several tens of ns (typical lifetime of excited resonant states under low pressure conditions), as shown below. This finding excludes a direct role of helium metastable atoms as energy-carrying species in the effluent of the jet, and as such will have no direct significance for technologically relevant surface treatments.

Figure 9(a) shows the time averaged He* density distribution within the inter-electrode distance. The density profile shows off-center peaks near the rf sheath edges, where most of the high energy electrons are produced, and, subsequently, most of the excitation and ionization events occur. The He* production directly in front of the electrodes or inside the plasma bulk is comparably low, since there either the electron density or the mean electron energy is very low, respectively. The observed spatial He* density structure is supported by the results of an independent spatially resolved TDLAS experiment [30] on a similar discharge without molecular admixture.

Figure 9(b) show the time and space resolved He* density over one rf cycle and across the discharge gap. The He* density, concentrated near the sheath region, shows a specific temporal modulation, as a result of the strongly time and space dependent He* production through electron-impact excitation, shown in figure 9(c). Three distinct maxima per half cycle are found. Each of them can be attributed to a different plasma heating mechanism: the two weaker maxima, occurring simultaneously inside the pre-sheath regions, correspond to the sheath expansion and the corresponding sheath collapse at the opposite electrode side. The strongest is caused by electron acceleration and multiplication in the high-voltage plasma boundary sheath during the so-called sheath breakdown phase. Similar spatio-temporal structures have been observed in independent optical emission measurements [19] and numerical simulations [29].

The temporal evolution of the He* density during an rf cycle can be described by the differential rate equation

$$\frac{\partial n_{He^*}(t)}{\partial t} = k_{exc}(t) n_e(t) n_{He^*} - k_{ion} n_{O_2} n_{He^*}(t),$$  \hspace{1cm} (4)
since the rf period $T = 73.8$ ns is much shorter than the He* diffusion time, estimated as $l^2/D = 135 \mu s$ for a typical gradient scale length $l = 10^{-4}$ m and the binary diffusion constant $D = 0.74 \times 10^{-4}$ m$^2$s$^{-1}$ for He* in He at a gas temperature of $T_g = 345$ K and atmospheric pressure [31]. The formal solution of the rate equation for periodic boundary conditions, $n_{He^*}(t) = n_{He^*}(t + T)$, can be expressed as

$$n_{He^*}(t) = n_{He^*} \left[ \tilde{E}(T) \left( \exp(T/\tau) + 1 \right)^{-1} + \tilde{E}(t) \right] \exp(-t/\tau)$$

(5)

with $\tilde{E}(t) = \int_0^t k_{exc}(t') n_{e}(t') \exp(t'/\tau) \, dt'$ including the effective excitation rate, and $\tau = (k_{ion} n_{O2})^{-1}$ the effective He* lifetime due to the loss through Penning ionization. The effective lifetime $\tau \approx 35$ ns, calculated on the basis of the rate coefficient $k_{ion} = 2.54 \times 10^{-16}$ m$^3$s$^{-1}$ from [32], is shorter than the rf period, but longer than the width of the excitation pulses ($\approx 15$ ns). This explains the modulation of the He* density, as well as the temporal shift ($\approx 10$ ns) and broadening of the corresponding spatio-temporal structures with respect to the excitation pulses.

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**Appendix. He/O2 plasma-chemical reactions**

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<tr>
<td></td>
<td>Electron-impact excitation, dissociation and ionization</td>
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<td>$e + He \rightarrow He + e$</td>
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<td>[26]</td>
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<tr>
<td>(R2)</td>
<td>$e + He^m \rightarrow He^* + e + e$</td>
<td>$f(e)$</td>
<td>[33]</td>
</tr>
<tr>
<td>(R3)</td>
<td>$e + He \rightarrow He^* + e$</td>
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<td>[26]</td>
</tr>
<tr>
<td>(R4)</td>
<td>$e + He \rightarrow He^* + e + e$</td>
<td>$f(e)$</td>
<td>[26]</td>
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<tr>
<td>(R5)</td>
<td>$e + O_2 \rightarrow O_2 + e$</td>
<td>$f(e)$</td>
<td>[34]</td>
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<td>$f(e)$</td>
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<td>$f(e)$</td>
<td>[34]</td>
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(R22) e + O_2(\nu) \rightarrow O(^1D) + O(^1D) + e \hspace{1cm} f(\epsilon) \hspace{1cm} [34]
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(R43) e + O \rightarrow O^- + O_2 \hspace{1cm} f(\epsilon) \hspace{1cm} [35]
(R44) e + O \rightarrow O^+_2 + O \hspace{1cm} f(\epsilon) \hspace{1cm} [35]
(R45) e + O \rightarrow 88\% O(^1D) + O_2(^1\Delta) + e \hspace{1cm} f(\epsilon) \hspace{1cm} [36]
(R46) \rightarrow 12\% O + O_2 + e \hspace{1cm} f(\epsilon) \hspace{1cm} [34]
(R47) e + O \rightarrow O + e \hspace{1cm} f(\epsilon) \hspace{1cm} [34]
(R48) e + O \rightarrow O(^1D) + e \hspace{1cm} f(\epsilon) \hspace{1cm} [34]
(R49) e + O \rightarrow O^+ + e + e \hspace{1cm} f(\epsilon) \hspace{1cm} [34]
(R50) e + O(^1D) \rightarrow O^+ + e \hspace{1cm} f(\epsilon) \hspace{1cm} [34]
(R51) e + O(^1D) \rightarrow O(^1D) + e \hspace{1cm} f(\epsilon) \hspace{1cm} [34]
(R52) e + O(^1D) \rightarrow O^+ + e + e \hspace{1cm} f(\epsilon) \hspace{1cm} [34]

Electron recombination

(R53) He^* + e + e \rightarrow He^* + e \hspace{1cm} 6.0 \times 10^{-32} (T_e/T_0)^{-4.0} \hspace{1cm} [37]
(R54) He^+_2 + e + e \rightarrow 75\% He_2^* + e + h\nu \hspace{1cm} 4.0 \times 10^{-32} (T_e/T_0)^{-4.0} \hspace{1cm} [37]
(R55) \rightarrow 25\% He^* + He + e \hspace{1cm} [37]
(R56) He^+_2 + e + He \rightarrow He^* + He + He + h\nu \hspace{1cm} 5.0 \times 10^{-39} (T_e/T_0)^{-1.0} \hspace{1cm} [37]
(R57) O^+ + e \rightarrow O(^1D) \hspace{1cm} 4.7 \times 10^{-17} T_e^{-0.5} \hspace{1cm} [23]
(R58) O^+ + e \rightarrow O(^1D) + e \hspace{1cm} 1.6 \times 10^{-21} T_e^{-4.5} \hspace{1cm} [23]
(R59) O^+_2 + e \rightarrow O(^1D) + O \hspace{1cm} f(\epsilon) \hspace{1cm} [38, 39]
(R60) O^+_2 + e \rightarrow O_2 + O \hspace{1cm} 6.0 \times 10^{-10} T_e^{-1.0} \hspace{1cm} [40]
(R61) O^+_2 + e + e \rightarrow O_2 + O_2 + e \hspace{1cm} 9.8 \times 10^{-21} T_e^{-4.5} \hspace{1cm} [40]

Associative ionization

(R62) He^* + He^* \rightarrow 70\% He^+_2 + e \hspace{1cm} 1.5 \times 10^{-15} \hspace{1cm} [37]
(R63) \rightarrow 30\% He^* + He + e \hspace{1cm} 1.5 \times 10^{-16} \hspace{1cm} [37]
(R64) He^* + He^+_2 \rightarrow 85\% He^+_2 + He + e \hspace{1cm} 1.5 \times 10^{-16} \hspace{1cm} [37]
(R65) \rightarrow 15\% He^* + 2He + e \hspace{1cm} 1.5 \times 10^{-16} \hspace{1cm} [37]
(R66) He^*_2 + He^* \rightarrow 85\% He^+_2 + 2He + e \hspace{1cm} 2.6 \times 10^{-16} \hspace{1cm} [32]
(R67) \rightarrow 15\% He^* + 3He + e \hspace{1cm} 2.6 \times 10^{-16} \hspace{1cm} [32]
(R68) He^* + O_2 \rightarrow O^+_2 + He + e \hspace{1cm} 2.6 \times 10^{-16} \hspace{1cm} [32]
(R69) He^* + O_2 + He \rightarrow O^+_2 + 2He + e \hspace{1cm} 1.6 \times 10^{-43} \hspace{1cm} [32]
(R70) He^* + O_2(^1\Delta) \rightarrow O^+_2 + He + e \hspace{1cm} 2.6 \times 10^{-16} \hspace{1cm} [32]
(R71) He^* + O_2(^1\Delta) + He \rightarrow O^+_2 + 2He + e \hspace{1cm} 1.6 \times 10^{-43} \hspace{1cm} [32]
(R72) He^* + O_2(^1\Sigma) \rightarrow O^+_2 + He + e \hspace{1cm} 2.6 \times 10^{-16} \hspace{1cm} [32]
<table>
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<tbody>
<tr>
<td>(R73)</td>
<td>He* + O$_3$($^1\Sigma$) + He $\rightarrow$ O$_3^+$ + 2He + e</td>
<td>$1.6 \times 10^{-43}$</td>
<td>[32]</td>
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<tr>
<td>(R74)</td>
<td>He* + O$_3$ $\rightarrow$ O$_3^+$ + O + He + e</td>
<td>$2.6 \times 10^{-16}$</td>
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<tr>
<td>(R75)</td>
<td>He* + O$_3$ + He $\rightarrow$ O$_3^+$ + O + 2He + e</td>
<td>$1.6 \times 10^{-43}$</td>
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<tr>
<td>(R76)</td>
<td>He* + O $\rightarrow$ O$^+$ + He+ e</td>
<td>$2.6 \times 10^{-16}$</td>
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<tr>
<td>(R77)</td>
<td>He* + O + He $\rightarrow$ O$^+$ + 2He + e</td>
<td>$1.6 \times 10^{-43}$</td>
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<tr>
<td>(R78)</td>
<td>He* + O($^1D$) $\rightarrow$ O$^+$ + He+ e</td>
<td>$2.6 \times 10^{-16}$</td>
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<tr>
<td>(R79)</td>
<td>He$^+$ + O($^1D$) + He $\rightarrow$ O$^+$ + 2He + e</td>
<td>$1.6 \times 10^{-43}$</td>
<td>[32]</td>
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<tr>
<td>(R80)</td>
<td>He$_2^+$ + O$_2$ $\rightarrow$ O$_3^+$ + He + He + e</td>
<td>$3.6 \times 10^{-16}$</td>
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<tr>
<td></td>
<td>Electron detachment</td>
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<tr>
<td>(R81)</td>
<td>O$^-$ + O$_2$ $\rightarrow$ O$_3$ + e</td>
<td>$5.0 \times 10^{-21} T^0.5$</td>
<td>[23, 39]</td>
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<tr>
<td>(R82)</td>
<td>O$^-$ + O$_3$(1$\Delta$) $\rightarrow$ O$_3$ + e</td>
<td>$3.0 \times 10^{-16} T^0.5$</td>
<td>[23, 39]</td>
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<tr>
<td>(R83)</td>
<td>O$^-$ + O$_3$(1$\Sigma$) $\rightarrow$ O + O$_2$ + e</td>
<td>$6.9 \times 10^{-16} T^0.5$</td>
<td>[23, 39]</td>
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<tr>
<td>(R84)</td>
<td>O$^-$ + O$_3$ $\rightarrow$ O$_2$ + O$_2$ + e</td>
<td>$3.0 \times 10^{-16} T^0.5$</td>
<td>[23]</td>
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<tr>
<td>(R85)</td>
<td>O$^-$ + O $\rightarrow$ O$_2$ + e</td>
<td>$2.0 \times 10^{-16} T^0.5$</td>
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<tr>
<td>(R86)</td>
<td>O$^-$ + e $\rightarrow$ O + e + e</td>
<td>$2.2 \times 10^{-20} T^0.5 \exp(-26303/T_e)$</td>
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<tr>
<td>(R87)</td>
<td>O$_2^+$ + O $\rightarrow$ O$_3$ + e</td>
<td>$1.5 \times 10^{-16} T^0.5$</td>
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<td>(R88)</td>
<td>O$_2^+$ + O$_2$(1$\Delta$) $\rightarrow$ O$_2$ + O$_2$ + e</td>
<td>$2.0 \times 10^{-16} T^0.5$</td>
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<tr>
<td>(R89)</td>
<td>O$_2^+$ + O$_2$(1$\Sigma$) $\rightarrow$ O$_2$ + O$_2$ + e</td>
<td>$3.6 \times 10^{-16} T^0.5$</td>
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<tr>
<td>(R90)</td>
<td>O$_2^+$ + He$^+$ $\rightarrow$ O$_2$ + O$_2$ + He + e</td>
<td>$3.0 \times 10^{-16}$</td>
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<tr>
<td>(R91)</td>
<td>O$_2^+$ + O($^1D$) $\rightarrow$ O + O$_2$ + O$_2$ + e</td>
<td>$1.0 \times 10^{-16}$</td>
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<tr>
<td>(R92)</td>
<td>O$_2^+$ + O$_2$(1$\Sigma$) $\rightarrow$ O$_2$ + O$_2$ + O$_2$ + e</td>
<td>$1.0 \times 10^{-16}$</td>
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<td>Positive charge exchange</td>
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<tr>
<td>(R93)</td>
<td>He$^+$ + He + He $\rightarrow$ He$_2^+$ + He</td>
<td>$6.6 \times 10^{-44}$</td>
<td>[37]</td>
</tr>
<tr>
<td>(R94)</td>
<td>He$^+$ + O$_3$ $\rightarrow$ 97% O$^+$ + O + He</td>
<td>$1.1 \times 10^{-15} T^0.5$</td>
<td>[23]</td>
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<tr>
<td>(R95)</td>
<td>$\rightarrow$ 3% O$_2^+$ + He</td>
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<tr>
<td>(R96)</td>
<td>He$^+$ + O$_3$(1$\Delta$) $\rightarrow$ 97% O$^+$ + O + He</td>
<td>$1.1 \times 10^{-15} T^0.5$</td>
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<tr>
<td>(R97)</td>
<td>$\rightarrow$ 3% O$_2^+$ + He</td>
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<tr>
<td>(R98)</td>
<td>He$^+$ + O$_3$ $\rightarrow$ O$^+$ + O$_2$ + He</td>
<td>$1.1 \times 10^{-15} T^0.5$</td>
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<tr>
<td>(R99)</td>
<td>He$^+$ + O $\rightarrow$ O$^+$ + He</td>
<td>$5.0 \times 10^{-17} T^0.5$</td>
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<tr>
<td>(R100)</td>
<td>He$^+$ + O($^1D$) $\rightarrow$ O$^+$ + He</td>
<td>$5.0 \times 10^{-17} T^0.5$</td>
<td>[23]</td>
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<tr>
<td>(R101)</td>
<td>O$^+$ + O$_2$ $\rightarrow$ O$_2^+$ + O</td>
<td>$2.0 \times 10^{-17} T^0.5 \exp(-5030/T_e)$</td>
<td>[23]</td>
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<tr>
<td>(R102)</td>
<td>O$^+$ + O$_3$ $\rightarrow$ O$_2^+$ + O$_2$</td>
<td>$1.0 \times 10^{-16}$</td>
<td>[39]</td>
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<tr>
<td>(R103)</td>
<td>O$^+$ + O + M $\rightarrow$ O$_2^+$ + M</td>
<td>$1.0 \times 10^{-41} T^0.5$</td>
<td>[23, 39]</td>
</tr>
<tr>
<td>(R104)</td>
<td>O$_2^+$ + O$_2$ + M $\rightarrow$ O$_2^+$ + M</td>
<td>$3.9 \times 10^{-42}$</td>
<td>[40]</td>
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<tr>
<td>(R105)</td>
<td>O$_2^+$ + He$^+$ $\rightarrow$ O$_2^+$ + O$_2$ + He</td>
<td>$1.0 \times 10^{-16}$</td>
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<tr>
<td>(R106)</td>
<td>O$_2^+$ + O$_3$ $\rightarrow$ O$_2^+$ + O$_2$ + O$_2$</td>
<td>$3.3 \times 10^{-12} T^{-4.0} \exp(-5030/T_e)$</td>
<td>[42]</td>
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<tr>
<td>(R107)</td>
<td>O$_2^+$ + O$_3$(1$\Delta$) $\rightarrow$ O$_2^+$ + O$_2$ + O$_2$</td>
<td>$1.0 \times 10^{-16}$</td>
<td>[40]</td>
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<tr>
<td>(R108)</td>
<td>O$_2^+$ + O$_3$(1$\Sigma$) $\rightarrow$ O$_2^+$ + O$_2$ + O$_2$</td>
<td>$1.0 \times 10^{-16}$</td>
<td>[40]</td>
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<tr>
<td>(R109)</td>
<td>O$_2^+$ + O $\rightarrow$ O$_2^+$ + O$_3$</td>
<td>$3.0 \times 10^{-16}$</td>
<td>[42]</td>
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<tr>
<td>(R110)</td>
<td>O$_2^+$ + O($^1D$) $\rightarrow$ O$_2^+$ + O + O$_2$</td>
<td>$1.0 \times 10^{-16}$</td>
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<tr>
<td></td>
<td>Negative charge exchange</td>
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<tr>
<td>(R111)</td>
<td>O$^+$ + O$_3$(1$\Delta$) $\rightarrow$ O$_3^- + O$</td>
<td>$1.0 \times 10^{-16} T^0.5$</td>
<td>[36]</td>
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<tr>
<td>(R112)</td>
<td>O$^+$ + O$_3$ $\rightarrow$ 95% O$_3^- + O$</td>
<td>$2.1 \times 10^{-16} T^0.5$</td>
<td>[23]</td>
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<tr>
<td>(R113)</td>
<td>$\rightarrow$ 5% O$_3^- + O_3$</td>
<td></td>
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<tr>
<td>(R114)</td>
<td>O$_2^+$ + O$_3$ $\rightarrow$ O$_3^- + O_3$</td>
<td>$6.0 \times 10^{-16} T^0.5$</td>
<td>[23]</td>
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<tr>
<td>(R115)</td>
<td>O$_2^+$ + O $\rightarrow$ O$^-$ + O$_2$</td>
<td>$1.5 \times 10^{-16} T^0.5$</td>
<td>[23]</td>
</tr>
<tr>
<td>(R116)</td>
<td>O$_2^+$ + O + M $\rightarrow$ O$_3^- + M$</td>
<td>$3.5 \times 10^{-41} T^{-1.0}$</td>
<td>[40]</td>
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<tr>
<td>(R117)</td>
<td>O$_2^+$ + O $\rightarrow$ O$_2^- + O_2$</td>
<td>$2.5 \times 10^{-16} T^0.5$</td>
<td>[23]</td>
</tr>
<tr>
<td>(R118)</td>
<td>O$_2^+$ + O$_2$ $\rightarrow$ O$_2^- + O_2 + O_2$</td>
<td>$2.2 \times 10^{-11} T^{-1.8} \exp(-6300/T_e)$</td>
<td>[40]</td>
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<tr>
<td>(R119)</td>
<td>O$_2^+$ + O$_3$(1$\Delta$) $\rightarrow$ O$_2^- + O_3 + O_2$</td>
<td>$1.0 \times 10^{-16}$</td>
<td>[40]</td>
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<tr>
<td>(R120)</td>
<td>O$_2^+$ + O$_3$(1$\Sigma$) $\rightarrow$ O$_2^- + O_2 + O_2$</td>
<td>$1.0 \times 10^{-16}$</td>
<td>[40]</td>
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<tr>
<td>(R121)</td>
<td>O$_2^+$ + O$_3$ $\rightarrow$ O$_3^- + O_2 + O_2$</td>
<td>$3.0 \times 10^{-16}$</td>
<td>[40]</td>
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<tr>
<td>(R122)</td>
<td>O$_2^+$ + O $\rightarrow$ O$_3^- + O_2$</td>
<td>$4.0 \times 10^{-16}$</td>
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<tr>
<td>(R123)</td>
<td>O$_2^+$ + O($^1D$) $\rightarrow$ O$_2^- + O + O_2$</td>
<td>$1.0 \times 10^{-16}$</td>
<td>[40]</td>
</tr>
</tbody>
</table>
Plasma Sources Sci. Technol. 20 (2011) 055005

K Niemi et al

**Mutual neutralization**

(R124) \( \text{He}^+ + \text{O}^- \rightarrow \text{He} + \text{O} \)

(R125) \( \text{He}^+ + \text{O}^- + \text{M} \rightarrow \text{He} + \text{O} + \text{M} \)

(R126) \( \text{He}^+ + \text{O}_2^- \rightarrow \text{He} + \text{O}_2 \)

(R127) \( \text{He}^+ + \text{O}_3^- \rightarrow \text{He} + \text{O}_3 \)

(R128) \( \text{O}^+ + \text{O}^- \rightarrow \text{O} + \text{O} \)

(R129) \( \text{O}^+ + \text{O}^- + \text{M} \rightarrow \text{O} + \text{O} + \text{M} \)

(R130) \( \text{O}^+ + \text{O}_2 \rightarrow \text{O} + \text{O}_2 \)

(R131) \( \text{O}^+ + \text{O}_3 \rightarrow \text{O} + \text{O}_3 \)

(R132) \( \text{O}_2^+ + \text{O}^- \rightarrow \text{O} + \text{O}_2 \)

(R133) \( \text{O}_2^+ + \text{O}^- + \text{M} \rightarrow \text{O} + \text{O}_2 + \text{M} \)

(R134) \( \text{O}_2^+ + \text{O}^- \rightarrow \text{O} + \text{O} + \text{O} \)

(R135) \( \text{O}_2^+ + \text{O}^- + \text{O}_2 \rightarrow \text{O}_3 + \text{O}_2 \)

(R136) \( \text{O}_2^+ + \text{O}_2 \rightarrow \text{O}_2 + \text{O}_2 \)

(R137) \( \text{O}_2^+ + \text{O}_2 \rightarrow \text{O} + \text{O} + \text{O}_2 \)

(R138) \( \text{O}_2^+ + \text{O}_3 \rightarrow \text{O}_3 + \text{O}_2 \)

(R139) \( \text{O}_2^+ + \text{O}_3 \rightarrow \text{O} + \text{O} + \text{O}_3 \)

(R140) \( \text{O}_2^+ + \text{O}_3 + \text{He} \rightarrow \text{O}_3 + \text{He} \)

(R141) \( \text{O}_2^+ + \text{O}^- + \text{He} \rightarrow \text{O} + \text{O}_2 + \text{He} \)

(R142) \( \text{O}_2^+ + \text{O}_3 + \text{He} \rightarrow \text{O}_3 + \text{He} \)

(R143) \( \text{O}_2^+ + \text{O}_3 + \text{He} \rightarrow \text{O}_3 + \text{O}_2 + \text{He} \)

(R144) \( \text{O}_2^+ + \text{O}_3 + \text{He} \rightarrow \text{O}_4 + \text{He} \)

\( \rightarrow \)

**Chemical conversion**

(R145) \( \text{O}^{(1)} \text{D} + \text{O} \rightarrow \text{O} + \text{O} \)

(R146) \( \text{O}^{(1)} \text{D} + \text{O}_2 \rightarrow 90 \% \text{ O} + \text{O}_2^{(1 \Sigma)} \)

(R147) \( \rightarrow 5 \% \text{ O} + \text{O}_2^{(1 \Delta)} \)

(R148) \( \rightarrow 15 \% \text{ O} + \text{O}_2 \)

(R149) \( \text{O}^{(1)} \text{D} + \text{O}_3 \rightarrow 50 \% \text{ O} + \text{O} + \text{O}_2 \)

(R150) \( \rightarrow 50 \% \text{ O}_2 + \text{O}_2 \)

(R151) \( \text{O}^{(1)} \text{D} + \text{He} \rightarrow \text{O} + \text{He} \)

(R152) \( \text{O}_2^{(1 \Delta)} + \text{O} \rightarrow \text{O} + \text{O}_2 \)

(R153) \( \text{O}_2^{(1 \Delta)} + \text{O}_2 \rightarrow \text{O}_2 + \text{O}_2 \)

(R154) \( \text{O}_2^{(1 \Delta)} + \text{O}_2 \rightarrow \text{O} + \text{O}_3 \)

(R155) \( \text{O}_2^{(1 \Delta)} + \text{O}_2 \rightarrow 50 \% \text{ O}_2 + \text{O}_2 \)

(R156) \( \rightarrow 50 \% \text{ O}_2^{(1 \Delta)} + \text{O}_2 \)

(R157) \( \text{O}_2^{(1 \Delta)} + \text{O}_3 \rightarrow \text{O} + \text{O}_2 + \text{O}_2 \)

(R158) \( \text{O}_2^{(1 \Delta)} + \text{He} \rightarrow \text{O}_2 + \text{He} \)

(R159) \( \text{O}_2^{(1 \Sigma)} + \text{O}_2^{(1 \Sigma)} \rightarrow \text{O}_2^{(1 \Delta)} + \text{O}_2 \)

(R160) \( \text{O}_2^{(1 \Sigma)} + \text{O}_2 \rightarrow 90 \% \text{ O}_2^{(1 \Delta)} + \text{O}_2 \)

(R161) \( \rightarrow 10 \% \text{ O}_2 + \text{O}_2 \)

(R162) \( \text{O}_2^{(1 \Sigma)} + \text{O} \rightarrow 90 \% \text{ O}_2^{(1 \Delta)} + \text{O} \)

(R163) \( \rightarrow 10 \% \text{ O}_2 + \text{O} \)

(R164) \( \text{O}_2^{(1 \Sigma)} + \text{O}_3 \rightarrow 33 \% \text{ O} + \text{O}_2 + \text{O}_2 \)

(R165) \( \rightarrow 33 \% \text{ O}_2^{(1 \Delta)} + \text{O}_3 \)

(R166) \( \rightarrow 33 \% \text{ O}_2 + \text{O}_3 \)

(R167) \( \text{O}_2^{(1 \Sigma)} + \text{He} \rightarrow \text{O}_2^{(1 \Delta)} + \text{He} \)

(R168) \( \text{O} + \text{O} + \text{O}_2 \rightarrow 93 \% \text{ O}_2 + \text{O}_2 \)

(R169) \( \rightarrow 7 \% \text{ O}_2^{(1 \Delta)} + \text{O}_2 \)

(R170) \( \text{O} + \text{O} + \text{O}_2 \rightarrow \text{O}_3 + \text{O} \)

(R171) \( \text{O} + \text{O} + \text{O} \rightarrow 93 \% \text{ O} + \text{O}_2 \)

(R172) \( \rightarrow 7 \% \text{ O} + \text{O}_2^{(1 \Delta)} \)

(R173) \( \text{O} + \text{O} + \text{He} \rightarrow 90 \% \text{ O}_2 + \text{He} \)

(R174) \( \rightarrow 10 \% \text{ O}_2^{(1 \Delta)} + \text{He} \)

(R175) \( \text{O} + \text{O}_2 + \text{O}_2 \rightarrow \text{O}_3 + \text{O}_2 \)

(R176) \( \text{O} + \text{O}_2 + \text{He} \rightarrow \text{O}_3 + \text{He} \)

(R177) \( \text{O} + \text{O}_2 + \text{O}_3 \rightarrow \text{O}_3 + \text{O}_3 \)

2.3 \times 10^{-11} \exp(-1057/T_\beta) \)
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<tr>
<td>(R178)</td>
<td>O + O₃ → O₂ + O₂</td>
<td>1.5 \times 10^{-17} \exp(-2250/T)</td>
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<tr>
<td>(R179)</td>
<td>O + O₃ → O + O + O₂</td>
<td>9.4 \times 10^{-17} \exp(-11400/T)</td>
<td>[23]</td>
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<tr>
<td>(R180)</td>
<td>O₂ + M → O + O₂ + M</td>
<td>1.6 \times 10^{-15} \exp(-11400/T)</td>
<td>[23]</td>
</tr>
<tr>
<td>(R181)</td>
<td>O₂ + O₃ → O₂ + O₂</td>
<td>1.6 \times 10^{-15} \exp(-11400/T)</td>
<td>[23]</td>
</tr>
<tr>
<td>(R182)</td>
<td>He⁺ + He + He → He₂⁺ + He</td>
<td>1.5 \times 10^{-6}</td>
<td>[43]</td>
</tr>
<tr>
<td>(R183)</td>
<td>O₂^{(v)} + M → O₂ + M</td>
<td>1.0 \times 10^{-20} T_0^{0.5}</td>
<td>[23]</td>
</tr>
<tr>
<td>(R184)</td>
<td>O₂^{(v)} + O → O₂ + O</td>
<td>1.0 \times 10^{-20} T_0^{0.5}</td>
<td>[23]</td>
</tr>
</tbody>
</table>

* M denotes background gas species He and O₂.

* Rate coefficient in m³ s⁻¹ for two-body reaction or in m⁶ s⁻¹ for three-body reaction, with Tₑ and T₀ in K, and T₀ = Tₑ/300. $f(\epsilon)$ indicates result of Boltzmann solver.

References


