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Carbon Dioxide Reactions in Non-thermal Low Temperature Plasma

Qiang Huang, Diyu Zhang, Aart W. Kleyn

Center of Interface Dynamics for Sustainability, Institute of Materials, China Academy of Engineering Physics, Chengdu, Sichuan 610200, PR China

Recently, worldwide attention has been focused on the increased emissions of greenhouse gases (mainly carbon dioxide) and the decreasing reserves of traditional energy sources. Utilizing sustainable energy to convert carbon dioxide into fuels or value-added chemicals has great potential to handle these two problems. Among the complex reaction steps involved, highly endothermic carbon dioxide dissociation is one of the key processes affecting the overall reaction efficiency. The non-thermal plasma, which could be easily generated via sustainable electricity and contains chemically active species such as high energy electrons, ions, atoms, and excited gas molecules, is expected to achieve high energy conversion efficiency at low gas temperature.

In plasma the electron collisions can excite vibrational modes of molecules, which can cause the carbon dioxide dissociation through multiple vibrational excitation or ladder climbing. A major fraction of the discharge energy could be transferred from plasma electrons to molecule vibrations while the other degrees of freedom rotation and translation are heated less. Meanwhile, the vibrational energy loss through relaxation is relatively slow. The vibrationally excited molecules will further interact with each other or collide with other plasma electrons, leading to a successive "vibrational pumping" up along the vibrational levels, due to their anharmonicity, and will reach the dissociation limit. This could result in high energy efficiency of the carbon dioxide dissociation in the non-thermal plasma, keeping the other degrees of freedom cooler. As shown in figure 1, the non-adiabatic transition $^1\Sigma^+\rightarrow{}^3B^2$ in the point of crossing of the terms (change of spin during the transition) provides a more effective dissociation process stimulated by the vibrational excitation, direct ladder climbing being more efficient. The electron energy distribution function (EEDF) plays an important role on this kind of ladder climbing of vibrational levels, because for carbon dioxide an electron temperature of about 1 eV could transfer a major portion of the discharge energy to the low vibrational levels to start the climbing process. The other ways to induce dissociation by electron impact, direct excitation of the electronically excited $^1B^2$ and $^3B^2$ states leads to excess translational energy in the CO + O fragments and requires high energy electrons.

We have studied carbon dioxide dissociation in inductively coupled radiofrequency plasma and microwave plasma at low gas pressure. Both systems exhibit features of non-thermal plasma. The highest energy efficiency observed is 59.3% (2.13mmol/kJ), exceeding the maximum value of about 45% in case of thermodynamic equilibrium, and a maximum conversion of 80.6% is achieved. Different discharge conditions, such as the source frequency, discharge gas pressure and the addition of argon, will affect the plasma parameters, especially the electron energy distribution. This plays a great role on the energy transfer from the non-thermal plasma to the molecular dissociation reaction channel via realizing the ladder climbing of the carbon dioxide molecular vibration. The results indicate the importance of ladder climbing.

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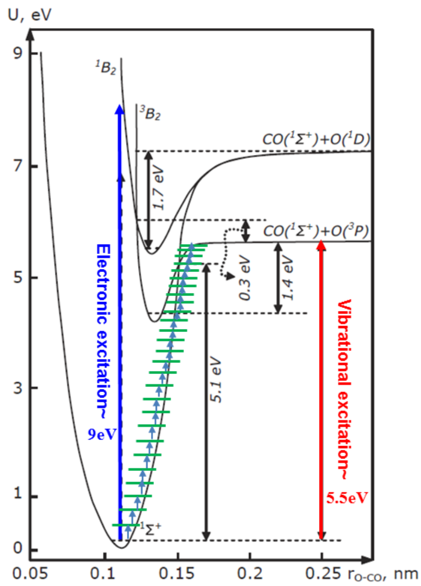

Figure 1. Low electronic terms of carbon dioxide

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