

主な研究成果

1. 交差分子線法による気相反応の反応動力学

化学反応素がどのように進むかを決定するのは、関与する反応物の原子や分子間の相互作用、またはポテンシャルエネルギー超曲面であるとの考えで、反応の起こる様子を観測し、運動状態をポテンシャルエネルギー面の関連において理解する学問が、反応動力学である。交差分子線法を用いて $A + B-C \rightarrow A-B + C$ 型の素反応の動力学を行ってきた。F 原子と一連の不飽和炭化水素との置換反応：例えば、 $F + C_6D_6 \rightarrow C_6D_5F + D$ の生成物の角度及び速度分布を測定し、反応中間体内で全ての振動モードにエネルギーが移動するかどうかを論じた [11]。また、回転型質量分析器を用いた研究としては、最も詳しい反応動力学的知見得られた反応系である $F + D_2 \rightarrow DF + D$ によって生成した DF 分子の重心系における微分断面積を求めた [29]。アーク放電によって生成した電子励起原子と分子との衝突によって生成した電子励起分子からの発光強度の衝突エネルギー依存性を求めることによって、励起原子の関与する反応の動力学を明らかにした [27, 30]。

2. シンクロトロン放射を用いた分子および半導体表面の真空紫外光励起化学の研究

分子科学研究所極端紫外光実験施設(UVSOR)の真空紫外光吸収分光法によって気相分子や分子クラスターの吸収スペクトルを測定し、Rydberg 状態を含む豊富な励起状態の帰属を行った [M4]。さらに、気相分子の真空紫外光解離によって生成する励起原子や分子からの発光やその偏光特性を観測して、真空紫外領域における分子の解離励起過程の動力学について明らかにした [89]。半導体材料やダイヤモンド表面のシンクロトロン放射励起によるエッチング反応の波長依存性やエッチャントの圧力依存性の測定を行い [38]、その機構について研究した。特に、 SiO_2 (エッチャント: SF_6) [65] やダイヤモンド (エッチャント: O_2) [94] の真空紫外光励起エッチング反応においては、表面層にある原子の内殻励起過程が、エッチング反応に直接的に効くことを示した。

3. 分子 - 表面相互作用と表面反応動力学の研究

シンクロトロン放射励起表面反応過程の研の過程で、清浄表面における反応研究の必要性を痛感し、分子線 - 表面反応解析装置を製作した。これを用いて、超熱 Cl_2 分子による $Si(111)$ のエッチング反応による生成物の速度分布及び表面方位依存性を測定し、脱離生成物の同定を行った [86]。また、原子または分子 - 固体表面衝突散乱による散乱物の散乱角及び速度分布測定を行って、並進 - 振動エネルギー移動過程を明らかにした [112]。北海道大学触媒化学研究センター松島龍夫現名誉教授の、清浄表面上に吸着した原子、分子どうしの Langmuir-Hinshelwood 機構による反応 ($CO(a)$ の酸化反応や NO の還元反応) 生成物の脱離方位角及び速度分布の測定プロジェクトに参画した [71]。この一連の研究によって、生成物

の生成直前の遷移状態の化学種と表面反応場の構造とについて知見を得ることができるようになった[113]。豊田理化学研究所における研究課題では、表面反応物のエネルギーを制御して、反応の PES を直接的に求めるための実験を試みた。これまで、 $^{18}\text{O}(\text{a})/\text{Pt}(111)$ と並進エネルギー1.2 eV の CO 分子との衝突による反応生成物はまだ見出されていない[R22]。今後、CO の衝突エネルギーを、さらに 2.3 eV まで上昇した衝突反応散乱実験を行う予定である。

(注)[]内の文献番号は、業績リストの数に対応する。

論文リスト

List of Publications (正島 宏祐)

2009年9月15日

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