

Electron scattering from C₄H₆ and C₄F₆ molecules

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Abstract

Absolute electron-impact total cross sections (TCSs) for C₄H₆ isomers (1, 3-butadiene and 2-butyne) and for hexafluoro-2-butyne (C₄F₆) have been measured in a linear electron-beam transmission experiment as a function of impact energy, between 0.5 and 370 eV. For the C₄H₆ molecule distinct differences in the shape and magnitude of TCS energy dependences of both isomers are apparent at low energies. In the 1, 3-butadiene TCS curve, the enhancement below 1.2 eV, the narrow maximum centred around 3.2 eV and the dominant broad peak at 9–9.5 eV are easily distinguishable; a very weak shoulder is located near 8 eV. The TCS energy function of 2-butyne demonstrates a remarkable resonant-like maximum located near 3.6 eV and much broader enhancement centred at 8 eV. Above 50 eV both C₄H₆ curves merge together. The hexafluoro-2-butyne TCS has the maximum near 8 eV and very broad enhancement spanning between 20 and 80 eV; weak shoulders near 3 and 6 eV are also noticeable. The effect of perfluorination is indicated.

1. Introduction

Electron interaction studies are of fundamental importance in the development of a basic understanding of the kinetics and dynamics of processes in which electrons are involved. Accurate electron-collision cross sections and electron-transport coefficients are important quantities necessary in a variety of applications as diverse as radiation biochemistry (Gohlke and Illenberger 2003), low-temperature processing plasmas (Tanaka and Inokuti 2000), gas discharges (Chutjian 2000), fusion edge plasmas (Summers 1994), plasma chemistry (Morgan 2000) and atmospheric (Shimazaki 1990) and astrophysical (Millar 2000) phenomena.

Studies of electron scattering from hydrocarbons and their substitutes are of interest due to the worldwide applications of these compounds. On the other hand, because of their versatility, hydrocarbons give specific opportunities for the investigation of how various changes in the structure of target molecules reflect in the mechanism of the scattering process. Surprisingly,

in spite of extensive experimental and theoretical electron-collision research, there are still a number of quite simple hydrocarbons for which electron impact studies are rather scarce.

C_4H_6 and C_4F_6 molecules are examples of compounds for which electron-scattering data available in the literature are far from being satisfactory. Franklin and Mogenis (1967) reported relative abundances and appearance potentials of the principal positive ions from 1, 3-butadiene. Electron impact excitation spectra at energies between 20 and 55 eV were taken by Mosher *et al* (1973). Temporary negative ion formation in butadiene has been studied in the transmission experiment by Burrow and Jordan (1975) at energies below 5 eV (see also Jordan and Burrow 1987). Rutkowsky *et al* (1980) measured absolute cross section for attachment of low-energy (below 10 eV) electrons to 1, 3-butadiene and 1-butyne with negative ion formation. The excitation spectra for 1-butyne were obtained over the subionization energy range by Bowman and Miller (1965). For 1-butyne and 2-butyne Dance and Walker (1974) recorded threshold electron energy-loss spectra up to 10 eV while Ng *et al* (1982) obtained derivatives with respect to impact energy of the transmitted electron current. 1, 3-butadiene has many industrial uses and, due to leaks and waste emissions from manufacturing facilities as well as from car exhaust fumes and/or passive cigarette smoke, becomes of environmental and medical interest (see Pacchierotti *et al* 1998 and references therein).

Even less available are works concerning the electron scattering by C_4F_6 molecule. Christodoulides *et al* (1979) obtained electron thermal attachment rates and determined the cross sections, at energies below 1.5 eV, for electron attachment to three isomers of the C_4F_6 molecule. The electron attachment and detachment properties of *c*- C_4F_6 at low impact energies were studied by Datskos *et al* (1993). More recently Bart *et al* (2001) have measured absolute total positive ion electron ionization cross sections for hexafluoro-1, 3-butadiene and hexafluoro-2-butyne up to 220 eV. The C_4F_6 molecule is considered to be a gaseous insulator.

The main purpose of the present work is to supply reliable experimental absolute electron-scattering total cross sections (TCSs) for C_4H_6 and C_4F_6 molecules over a wide energy range—to the best of our knowledge, no electron-scattering TCS data either for C_4H_6 or for C_4F_6 molecules seem to exist in the literature. As the very distinct isomeric effect in TCS energy functions has been very recently observed for C_3H_4 (Szmytkowski and Kwitnewski 2002b) and C_3H_6 (Szmytkowski and Kwitnewski 2002a, Makochehanwa *et al* 2003) molecules, it is also interesting to examine how the structure of the target molecules studied is reflected in the shape and magnitude of their TCS energy dependence.

2. Experimental details

The electron beam transmission apparatus and experimental procedure we employed in the present study were described earlier (Szmytkowski and Możejko 2001) and only a brief outline is given here. An electron beam of given energy, E (with an energy spread ΔE of $\simeq 80$ meV, FWHM), is directed into a scattering cell filled with a target sample. The target pressure is sufficiently close to single-collision conditions, i.e. mean free path of electrons in the medium under study is distinctly longer than the transmission length. Those electrons which pass the interaction volume are energetically discriminated by a retarding field analyser and eventually collected by a Faraday cup detector. The attenuation of the intensity of the transmitted electron beam is measured. To a first approximation, the attenuation factor is given by $\exp(-Q(E)nl)$, where $n = p/kT$ is the target density number (p is the target pressure at temperature T), l is the effective electron path length in the target and $Q(E)$ is the scattering TCS at energy E .

Energy calibration was accomplished by using the oscillatory resonant structure visible in the electron transmission current around 2.3 eV when nitrogen was admitted to the target studied.

Pressure readings of the sample gas in the scattering volume were taken with the MKS Baratron transducer and corrected for the thermal transpiration effect (Knudsen 1910). The background pressure in the electron optics volume was always three orders of magnitude lower than the pressure in the scattering cell and was kept constant, independently of whether the target was present or not in the scattering chamber.

In a more realistic approach (Bederson and Kieffer 1971) one would take into consideration the inhomogeneity of the target pressure across the scattering cell and the presence of target particles outside the cell in the vicinity of its orifices—both effects are associated with the effusion of gas through the cell apertures. One would also account for electrons that are scattered (at least elastically) through small angles in the forward direction. The error associated with the latter effect increases remarkably with the increase in electron impact-energy. Rough estimates show that the lowering of the measured TCS due to imperfect discrimination of electrons scattered forward in the present experiment should not exceed 5% at the highest applied energies.

The final TCS value at each particular energy, E , presented in this work is a weighted mean of results obtained in independent series (5–24) of individual runs (6–10 in a series). The statistical uncertainty (one standard deviation of the weighted mean value) of the measured TCS is nearly 1% below 1.5 eV and well below 1% for all higher investigated energies. Only in the experiment with 2-butyne, at the lowest applied energies, is the scatter of results about 2%. The TCSs obtained at the same energy appeared to be independent, within the random experimental uncertainties, of the applied pressure (0.06–0.14 Pa) and the electron-beam intensity (0.1–10 pA). The systematic uncertainty in the measured cross section was estimated as a direct sum of potential systematic errors of measured individual quantities used for the TCS derivation. The sources and magnitudes of most significant systematic error components are: uncertainty in the product nl in the attenuation relationship ($\sim 1.5\%$); inability to discriminate against electrons scattered at small forward angles (about 1% below 10 eV and 4–5% above 300 eV); electron current drift (0.5–1%); impurity of samples ($\sim 1\%$). The overall experimental uncertainty of TCS (as presented in the figures) was obtained as a combination in quadrature of total systematic and statistical uncertainties; it amounts to about 6% below 2–3 eV, decreases to about 4% around 10 eV and increases to 6–7% at the highest applied energies.

Samples that are gases at room temperature (1, 3-butadiene, 99 + %; hexafluoro-2-butyne, 99%) were used directly from commercially supplied cylinders (Sigma–Aldrich) without any further purification. The 2-butyne (Sigma–Aldrich, 99%) vapours were purified before use by a few repeated freeze–pump–thaw cycles at the liquid-N₂ temperature to remove air and other impurities not frozen at this temperature.

3. Results and discussion

In this section we present our absolute electron-scattering TCSs measured for two isomers of C₄H₆ molecules (1, 3-butadiene and 2-butyne) as well as for hexafluoro-2-butyne (isomer of C₄F₆ molecule) over the energy range from about 0.5 to 370 eV. To the best of our knowledge, no other TCS measurements and calculations for these molecules are as yet available in the literature for comparison.

3.1. 1, 3-butadiene, $H_2C=CH-CH=CH_2$

The energy dependence of the electron-impact TCS for 1, 3-butadiene is shown in figure 1 while numerical TCS values are listed in table 1. Over the energy range studied, the TCS curve

Table 1. Absolute experimental TCS for electron scattering on 1, 3-butadiene (C_4H_6) in units of 10^{-20} m^2 .

Energy (eV)	TCS	Energy (eV)	TCS	Energy (eV)	TCS	Energy (eV)	TCS
0.5	37.1	3.1	43.9	8.5	50.4	70	30.3
0.6	37.5	3.2	44.0	9.0	51.2	80	28.6
0.7	37.3	3.3	43.8	9.5	51.3	90	27.4
0.8	37.6	3.4	43.6	10	51.0	100	25.9
0.9	37.7	3.5	43.1	11	49.9	110	25.1
1.0	37.1	3.7	42.5	12	48.8	120	23.2
1.1	36.5	4.0	41.7	14	47.4	140	22.0
1.2	35.7	4.2	41.2	16	45.5	160	19.8
1.4	34.9	4.5	41.3	18	44.0	180	18.3
1.6	34.9	4.7	41.7	20	43.0	200	17.0
1.8	35.5	5.0	43.1	25	41.0	220	16.1
2.0	36.5	5.5	45.0	30	39.1	250	13.9
2.3	38.3	6.0	46.6	35	37.7	275	12.5
2.5	40.4	6.5	47.9	40	36.8	300	11.3
2.7	41.8	7.0	48.8	45	35.5	350	9.80
2.9	43.0	7.5	49.4	50	33.8	370	9.25
3.0	43.6	8.0	49.5	60	31.9		

shows three distinct enhancements: below 1.2 eV, in the vicinity of 3.2 and around 9.5 eV, separated with two minima located near 1.5 and 4.4 eV; more uncertain results, not presented here, suggest that just below 0.5 eV the TCS tends to decrease.

The TCS enhancement visible near 1 eV (figure 1) may be attributed to the formation of negative ion states when the colliding electron is temporarily captured into the lowest unoccupied molecular orbital of the 1, 3-butadiene. The above statement is concluded from earlier experiments of Burrow and Jordan (1975), Rutkowsky *et al* (1980) and calculations of Hoyland and Goodman (1962). Burrow and Jordan obtained the derivative of the transmitted electron current versus energy—the transmission spectrum that is more suitable for detection of subtle variations in TCS—and observed between 0.5 and 1.5 eV the well-developed vibrational pattern attributed mainly to the C=C symmetric stretch mode. They assigned this resonant feature to the lowest anion state, 2A_u , of the parent molecule; the $2a_u(\pi^*)$ orbital is the lower of two states arising from the splitting of the π^* orbitals of the two ethylenic subunits. Even more convincing evidence for this resonance comes from the electron attachment cross section obtained by Rutkowsky *et al* (1980); they observed a substantial increase of the negative ion formation between 0.5–1.2 eV, with a peak at 0.8 eV. Above 1.6 eV the TCS function rapidly increases with the impact energy increase and peaks at 3.2 eV. In this energy region Burrow and Jordan (1975) observed the structureless feature and related it to the second short-lived negative ion state with the extra electron attached to the next unfilled $2b_g(\pi^*)$ orbital. Near 7.5 eV some weak, but repetitive, change in the slope of the TCS curve is discernible. The broad TCS peak ($52 \times 10^{-20} \text{ m}^2$) centred near 9 eV is the most dominant feature over the energy studied. Such maximum located around 8–10 eV is present in TCS for many targets studied so far and is mainly associated with elastic scattering. Some contribution from a number of weak inelastic components, also resonant in nature, allowed at these energies is possible. From 10 eV up to the highest applied energy, 370 eV, the TCS monotonically decreases with the energy increase.

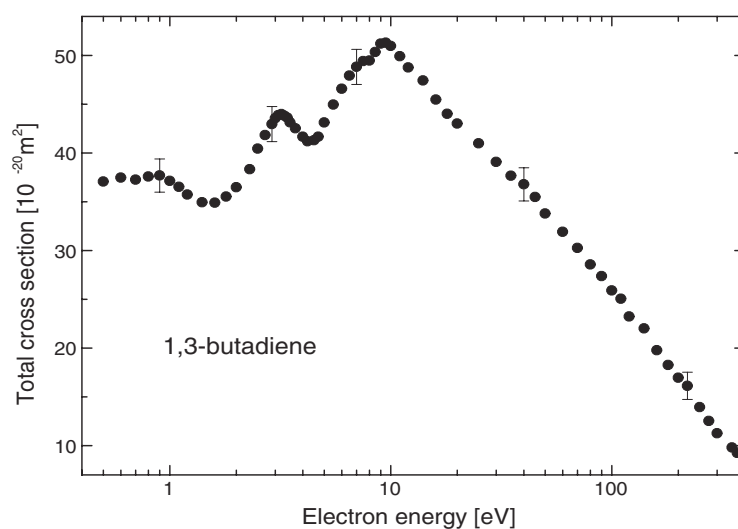


Figure 1. TCS for electron scattering from 1, 3-butadiene. The error bars indicate the overall (combination of systematic and random) experimental uncertainties at selected points.

3.2. 2-butyne, $H_3C-C\equiv C-CH_3$

Figure 2 shows the absolute TCS measured in the present experiment for electron-2-butyne scattering. Numerical values of TCS data are provided in table 2. The TCS energy function has two distinct features: the first one is located around 3.5 eV as a narrow peak superimposed on the left-hand slope of the very broad maximum centred near 8 eV. The structure at 3.5 eV is most probably associated with a shape resonance arising from temporarily trapping the impinging electron into the lowest antibonding orbital of the 2-butyne molecule. The evidence for the resonant character of the electron scattering in the vicinity of 3.5 eV comes from the threshold electron energy-loss spectrum of Dance and Walker (1974) in which they observed the 3.6 eV feature associated with resonance-enhanced vibrational excitation of the parent molecule. Another support for the resonant origin of the 3.5 eV TCS structure results from the feature observed around 3.4 eV by Ng *et al* (1982) in the derivative of the transmitted electron current. Beyond the main maximum, over the energy range from 10 up to 370 eV, the cross-section energy function monotonically decreases.

3.3. Isomeric effect

The electron-scattering TCS energy functions for two C₄H₆ isomers (1, 3-butadiene and 2-butyne) are compared in figure 3. Both isomers are open-chain hydrocarbons. However, whereas the 1, 3-butadiene ($H_2C=CH-CH=CH_2$) is a compound with conjugated carbon-carbon double bonds and with a CH₂ group on either end of the chain, the 2-butyne ($H_3C-C\equiv C-CH_3$) has one triple bond surrounded with two single C-C bonds and methyl groups attached on the edges of the molecule. From figure 3 it is evident that various arrangements of the same constituent atoms in the isomers of the C₄H₆ molecule influences the shape and magnitude of their low-energy electron scattering TCS. The most significant difference concerns the shape of the TCS curves at the lowest applied energies. Between 0.6 and 1.5 eV the TCS for 2-butyne monotonically descends when the impact energy decreases while for 1, 3-butadiene the TCS function distinctly increases when going from 1.5 down

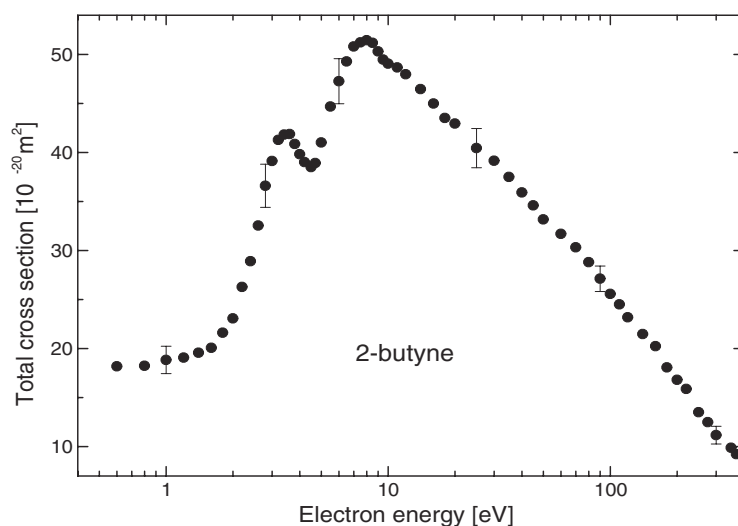


Figure 2. TCS for electron scattering from 2-butyne. The error bars at selected points represent the overall experimental uncertainties (systematic and statistical).

Table 2. Absolute TCS measured for electron impact on 2-butyne (C_4H_6) in units of 10^{-20} m^2 .

Energy (eV)	TCS	Energy (eV)	TCS	Energy (eV)	TCS	Energy (eV)	TCS
0.6	18.2	3.8	40.9	10	49.0	80	28.8
0.8	18.2	4.0	39.8	11	48.7	90	27.1
1.0	18.8	4.2	39.0	12	48.0	100	25.6
1.2	19.1	4.5	38.5	14	46.5	110	24.5
1.4	19.6	4.7	38.9	16	45.0	120	23.2
1.6	20.1	5.0	41.0	18	43.5	140	21.5
1.8	21.6	5.5	44.7	20	43.0	160	20.2
2.0	23.1	6.0	47.3	25	40.4	180	18.1
2.2	26.3	6.5	49.3	30	39.2	200	16.8
2.4	28.9	7.0	50.8	35	37.5	220	15.9
2.6	32.5	7.5	51.2	40	35.9	250	13.5
2.8	36.6	8.0	51.4	45	34.6	275	12.5
3.0	39.1	8.5	51.2	50	33.2	300	11.2
3.2	41.3	9.0	50.3	60	31.7	350	9.88
3.4	41.8	9.5	49.5	70	30.3	370	9.22
3.6	41.9						

to 0.9 eV and then seems to decrease very slowly. Another difference in this energy range concerns the TCS magnitude: below 1.5 eV, the TCS value for 1, 3-butadiene is nearly twice as large as that for 2-butyne. Between 3 and 10 eV this difference is less pronounced and mainly regards the location of the TCS maxima—it is evident that the energy distance between two maxima of 1, 3-butadiene exceeds that of 2-butyne. Both isomers demonstrate a distinct resonant-like maximum near 3 eV: centred at 3.2 eV for 1, 3-butadiene and at 3.6 eV for 2-butyne. Similar behaviour was already observed for double-bonded and triple-bonded isomers of the C_3H_4 molecule (Szmytkowski and Kwitnewski 2002b, Makochekanwa *et al* 2003). For intermediate energies, above 40 eV, no difference between two $e^-C_4H_6$ TCS curves

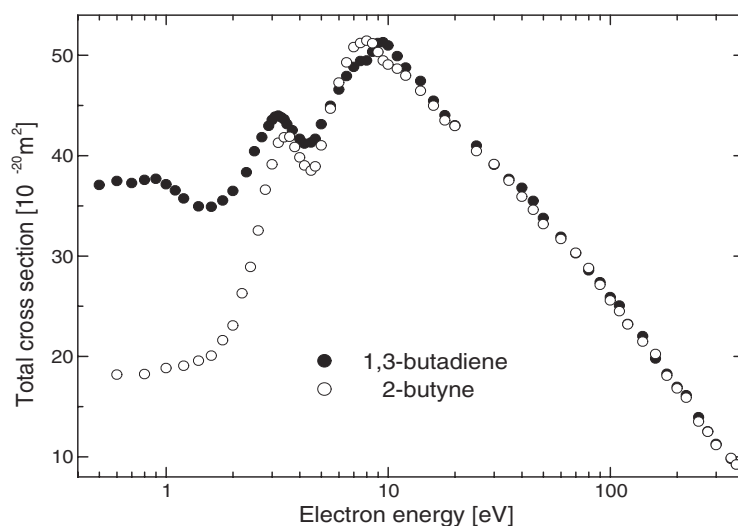


Figure 3. Comparison of the present TCSs for electron scattering from two isomers of the C₄H₆ molecule: (●), 1, 3-butadiene; (○), 2-butyne.

is distinguishable so the magnitude of TCS at these energies seems to be insensitive to the geometry of the target. The behaviour of both TCSs at intermediate energies can be well reproduced with the energy function: $\sim E^{-0.5}$.

3.4. Hexafluoro-2-butyne, F₃C–C≡C–CF₃

Figure 4 shows our TCS for electron scattering from hexafluoro-2-butyne together with cross sections for electron attachment available in the literature (Christodoulides *et al* 1979) and total ionization (Bart *et al* 2001) processes. Table 3 presents our TCS data in numerical form. The e[−]–C₄F₆ TCS curve rapidly increases above 1.5 eV and reaches the first maximum (of nearly $47 \times 10^{-20} \text{ m}^2$) at about 8 eV. Then the TCS decreases to the minimum placed near 12 eV and again enhances up to $49 \times 10^{-20} \text{ m}^2$ between 25 and 40 eV. Such energy dependence, with a very broad intermediate energy hump, is a distinctive characteristic of the perfluorinated molecules studied so far (e.g. Szymkowski *et al* 2000). Above 50 eV, towards higher energies, the cross section monotonically decreases and can be satisfactory approximated with the energy formula, $\sim E^{-a}$, where $a = 0.46$.

More thorough inspection of the e[−]–C₄F₆ TCS energy function reveals that below 8 eV two very weak, but nevertheless repetitive, shoulders are distinguishable around 3 and 6.5 eV, respectively. It is worth noting that no TCS structure is visible around 1 eV whereas Christodoulides *et al* (1979) observed in this energy region distinct enhancement in the electron attachment cross section (see figure 4), associated with the formation of long-lived parent negative ions. The contribution of the electron attachment to the total scattering is, at 0.8 eV, relatively high and amounts to $13 \times 10^{-20} \text{ m}^2$, nearly half that of the TCS at this energy. Figure 4 shows also that the contribution from electron-induced ionization (Bart *et al* 2001) to the total scattering process is relatively small and does not exceed 30% near 120 eV.

To show the perfluorination effect on the electron-scattering cross section, the TCSs for hexafluoro-2-butyne and its perhydrogenated counterpart (2-butyne) are compared in figure 5. This figure clearly demonstrates substantial differences in the magnitude and shape of the compared TCSs. Beneath 2.5 eV the TCS for perfluorocarbon is distinctly higher ($\sim 50\%$)

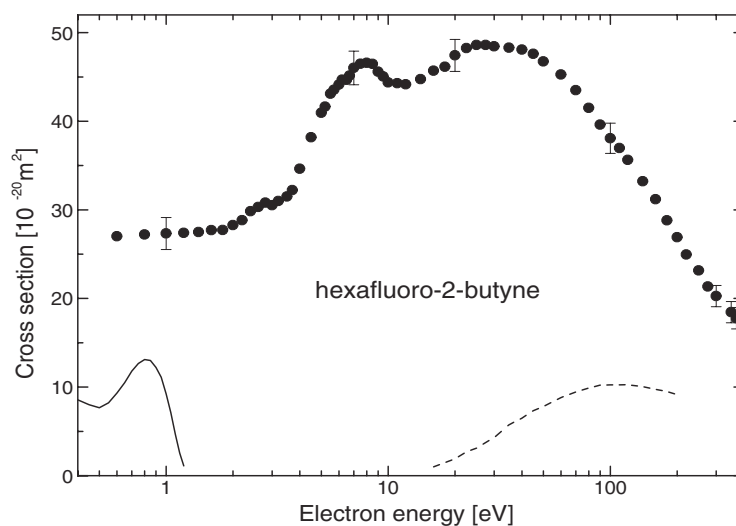


Figure 4. Electron-scattering cross sections from hexafluoro-2-butyne: (●), total, this work, the error bars at selected points indicate the overall (systematic and statistical) experimental uncertainties; (—), electron attachment, Christodoulides *et al* (1979); (---), ionization total, Bart *et al* (2001).

Table 3. Experimental absolute total electron scattering cross section (TCS) for hexafluoro-2-butyne (C_4F_6) in units of 10^{-20} m^2 .

Energy (eV)	TCS	Energy (eV)	TCS	Energy (eV)	TCS	Energy (eV)	TCS
0.6	27.0	4.0	34.6	10	44.4	70	43.5
0.8	27.2	4.5	38.2	11	44.3	80	41.5
1.0	27.3	5.0	41.0	12	44.2	90	39.6
1.2	27.4	5.2	41.6	14	44.8	100	38.1
1.4	27.5	5.5	43.1	16	45.7	110	37.0
1.6	27.7	5.7	43.6	18	46.1	120	35.6
1.8	27.7	6.0	44.1	20	47.4	140	33.2
2.0	28.3	6.2	44.7	22.5	48.2	160	31.2
2.2	28.8	6.5	44.7	25	48.6	180	28.8
2.4	29.8	6.7	45.2	27.5	48.6	200	26.9
2.6	30.3	7.0	46.0	30	48.4	220	25.0
2.8	30.8	7.5	46.5	35	48.3	250	23.2
3.0	30.5	8.0	46.6	40	48.1	275	21.3
3.2	31.0	8.5	46.4	45	47.6	300	20.3
3.5	31.5	9.0	45.6	50	46.8	350	18.4
3.7	32.2	9.5	45.1	60	45.3	375	17.8

than that for the hydrocarbon molecule. Above 2.5 eV it becomes lower and again is distinctly higher above 16 eV; the difference near 60 eV reaches 50%. It is worth noting that a similar relation between TCS for hydrocarbons and their perfluorinated analogues was already observed (Szmytkowski *et al* 2000).

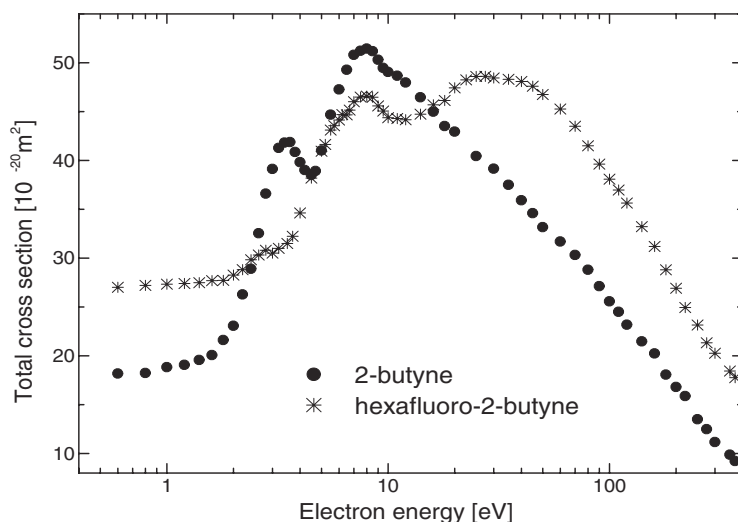


Figure 5. Comparison of the TCSs for electron scattering from 2-butyne and hexafluoro-2-butyne: (●), C_4H_6 , present; (★), C_4F_6 , present.

4. Conclusions

The present results provide absolute TCSs for electron scattering from 1, 3-butadiene and 2-butyne, two isomers of the C_4H_6 molecule, and for hexafluoro-2-butyne (C_4F_6) over an impact energy range between 0.5 and 370 eV. The results show that the differences in the arrangement of atoms in the target molecule lead to significant differences (isomeric effect) in the TCS mainly at low impact energies. Substitution of fluorine atoms for hydrogen atoms in the molecule changes the TCS (substitutional effect) over all the energy range investigated. At low impact energies a number of resonant-like features are observed in TCS energy functions; for an explanation further measurements in particular scattering channels, as well as calculations, would be desirable.

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