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Journal of Nuclear Materials 313–316 (2003) 413–418

journal of nuclear materials

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Erosion of CFC under simultaneous bombardment by helium and deuterium ions

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Abstract

The erosion of 1D-CFC due to the simultaneous bombardment of He^+, D_3^+, D_2^+ and D^+ ions with the known fluxes of the each species has been studied as a function of the ϕ_{He}/ϕ_D flux ratio with sample temperature of 450, 773 and 973 K. It has been commonly observed that for the low energy chemically active species for which the damage production rate is not saturated (<300 eV) the addition of a second energetic species should lead to an increase in the hydrocarbon release rate. However, a mechanism leading to the hydrocarbons molecule/precursor breakup can actually result in a decrease in the erosion yield. The balance between these two processes is a complicated function of flux ratio, projectile energies and concentration. Up to now there are no data available for the case when the energies of both species are not enough for the damage production rate saturation. At the present experiments the energy of deuterium was from 67 eV/deuteron for D_3^+ ions to 200 eV/deuteron for D^+ ions and the energy of He^+ ions was 200 eV. The analytical description of the graphite erosion by hydrogen ions from J. Nucl. Mater. 266–269 (1999) 51 was applied to describe the present experimental data.

PACS: 79.20.R
Keywords: Carbon fiber composite; Deuterium irradiation; Sputtering; Erosion; Ion bombardment; Helium

1. Introduction

The erosion of graphite under ion bombardment is intensively investigated during last two decades because the importance of this process for present and next generation fusion devices. Most of the studies have been dealt with the chemical erosion of graphite by hydrogen ions or atoms that seriously limits the lifetime of the carbon materials. The analytic description for the erosion yield by chemical sputtering has been reported [1,2] that can be fitted well to the existing experimental data. However in the extrapolation to divertor relevant conditions it is necessary to take into account that divertor materials will be subject of multispecies bombardment. In particularly, C and He impurities are always present at a certain level as a result of wall erosion and D–T fusion reaction, respectively. In ITER discharges the ratio of He to D + T flux onto the divertor plates was estimated can be as great as 0.1 [3]. In the preset experiments the erosion of 1D-CFC due to the simultaneous bombardment with He^+, D_3^+, D_2^+ and D^+ ions of known relative fluxes has been studied.

It has been commonly observed for various cases of graphite exposure to $Ar^+ + H^0$ [4,5], $H^+ + H^0$ [6–10], $C^+ + H^0$ [11], $C^+ + H^+$ [12], $He^+ + D^0$ [6], $He^+ + H^+$

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0022-3115/03/$ - see front matter
PII: S0022-3115(02)01401-0

[12,13] that for the low energy chemically active species for which the damage production rate is not saturated (<300 eV for H^+ [12]) the addition of a second energetic species should lead to an increase in the hydrocarbon release rate. However, a mechanism leading to the hydrocarbons molecule/precursor breakup can actually result in a decrease in the erosion yield. The balance between these two processes is a complicated function of flux ratio, projectile energies and concentration. Up to now there are no data available for the case when the energies of both species are not enough for the damage production rate saturation. At the present experiments the energy of deuterium was from 67 eV/deuteron for D_3^+ ions to 200 eV/deuteron for D^+ ions and the energy of He^+ ions was 200 eV. The experiments were carried out at lower temperature when thermally activated processes can be neglected and at higher temperature when those become dominated.

2. Experimental

All experiments were performed with the super low energy particle irradiation system (SLEIS) [14] that is schematically shown in Fig. 1. The dense arc plasma is produced in the magnetic multicups ion source, and ions are extracted through the specially designed single stage acceleration grid system. Due to the short distance between grids, the low energy beam with particle flux up to 1×10^{20} D/m^2 s can be formed. It has been measured [14] that with the total beam extraction area of 90 mm × 100 mm the uniformity of the beam was ±5% over an area of 60 mm × 100 mm at the sample position. In the previous studies pure H_2 [13,14] or D_2 [16] were used to run the ion source. Under certain gas pressure and arc power more than 85% of the total charged particles could be extracted as H_3^+ or D_3^+ ions, respectively. In the present experiments pure He, pure D_2 or $He + D_2$ simultaneously were introduced into ion source. The species of the charged particles were analyzed using a magnetic mass analyzer (MMA). Initially the ion source was put into operation with pure He and the total gas pressure measured in the target chamber was 27.9 mPa. Then in addition D_2 was introduced and a change of ion beam composition with increase of total pressure was observed, see Fig. 2. Since for both D_2^+ and He^+ ions $M/e = 4$, the contribution of the each species could not be defined from the MMA mass spectrum, but it was possible if H_2 was used instead of D_2, see Fig. 2 doted line. Therewith, the dependence of the relative ion fraction on total pressure for H^+, $H_2^+ + He^+$, H_3^+ was found to be similar within accuracy of 5% as for D^+, $D_2^+ + He^+$, and D_3^+, respectively. Under assumption that a change of He^+ flux with increasing total pressure is also identical for both cases, the D_2^+ ion fraction in the ion beam can be estimated. The summary of the irradiation conditions is shown in Table 1.

A test sample was placed about 10 cm downstream from ion source. Temperature of the sample was

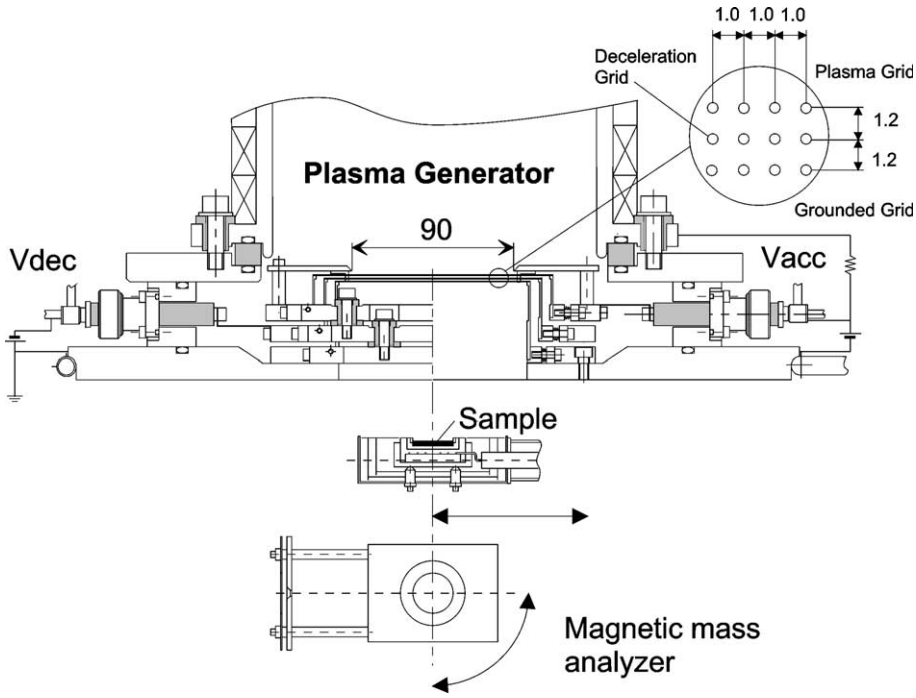

Fig. 1. Schematic of the SLEIS.

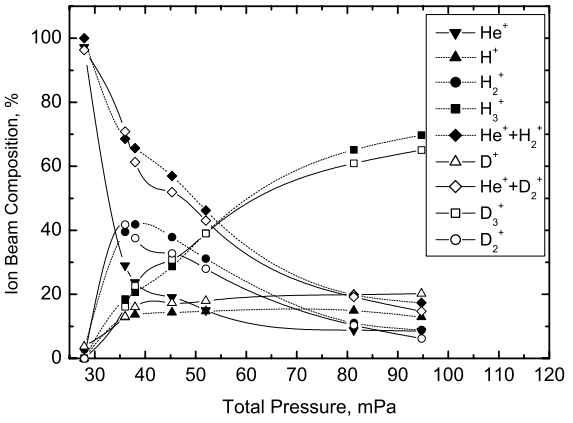

Fig. 2. Comparison of the He + D and He + H ion beam composition.

measured by thermocouples during irradiation. Size of each 1D-CFC sample was 25 mm × 25 mm × 3 mm, that is sufficiently smaller than the size of beam cross-section to provide the uniformity of a flux density. All samples were washed two times in the ethyl alcohol bath with an ultrasonic vibrator to remove completely a carbon powder from the target, and then were baked up to 900 °C in vacuum.

The sputtering yield was determined by the weight loss method. The target weight change was measured by a Mettler AE240 microbalance with an absolute accuracy 0.01 mg and the ion beam current – by a microammeter with an absolute accuracy better than 15%.

3. Results and discussions

We can estimate experimentally some effective sputtering yield of CFC:

$$Y_{\text{eff}} = N_{\text{C}}/(N_{\text{He}} + N_{\text{D}}), \tag{1}$$

where N_{C}, the total number of carbon atoms leaving the target in any form, was found from weight loss:

$$N_{\text{C}} = -(\Delta m \times N_0)/M_{\text{C}}, \tag{2}$$

where Δm is the weight change of the target, N_0 is Avogadro's number, M_{C} is the carbon atomic mass. N_{He} and $N_{\text{D}} \equiv N_{\text{D}^+} + 2N_{\text{D}_2^+} + 3N_{\text{D}_3^+}$ are the number of helium ions and deuteron, respectively, coming to the target that were calculated from the particle fluences and known ion beam composition.

It should be noted that even in case of pure deuterium bombardment there are some discrepancy in the available experimental data and theoretical prediction. Roth and Garsia-Rosales in Ref. [1] and Mech et al. in Ref. [17] have incorporated by different way the additional processes into the atomic impact model of Kuppers [18,19] in order to model the erosion due to energetic ion: (i) breaking of the C=C double bonds due to radiation damage, (ii) the kinetic removal of a methyl group attached to an sp^3 carbon center, and (iii) the graphitization of carbon that occurs at high temperature. Fitting of these models to experimentally measured erosion yields shows good agreement, while the experimental data itself determined by weight loss measurements in Ref. [1] are higher by up to a factor of ~3 than those obtained by residual gas mass spectrometry in Ref. [20]. This deviation could be attributed to the fact that emitted hydrocarbon radicals partly stick to the vessel surfaces and could not be detected in residual gas

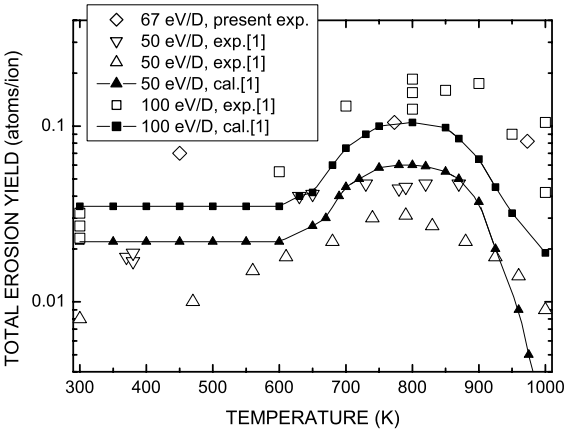

Fig. 3. Temperature dependence of the total erosion yield of carbon due to deuterium bombardment.

Table 1
Irradiation conditions

Total pressure mPa (He + D)/m^2 sec	$N_{\text{He}^+}/N\ \alpha$	$N_{\text{D}^+}/N\ \alpha_1$	$2N_{\text{D}_2^+}/N\ \alpha_2$	$3N_{\text{D}_3^+}/N\ \alpha_3$	$\phi_{\text{He}}/\phi_{\text{D}}$	Flux density × 10^{20}
29	1.00	–	–	–	–	0.15
33	0.333	0.067	0.400	0.200	0.5	0.23
42.7	0.103	0.077	0.359	0.461	0.114	0.42
56	0.061	0.080	0.254	0.601	0.065	0.43
95	0.036	0.085	0.053	0.826	0.037	0.62
108	–	0.040	–	0.960	–	1.37

Acceleration voltage is 200 eV.

analysis [2]. The total erosion yields measured in the present experiment and shown in Fig. 3 are higher than reported in Ref. [1]. The presence of 14% D^+ ions with the relatively high energy, 200 eV/deuteron, together with 86% D_3^+ ions with 67 eV/deuteron in the ion beam, possible contribution in the erosion yields simultaneously arrived neutral species may account for the observed discrepancies.

In accordance with the analytical model [2] the following formula for the total sputtering yield is proposed:

$$Y_{\text{tot}} = Y_{\text{phys}} + Y_{\text{surf}} + Y_{\text{therm}}(1 + DY_{\text{dam}}), \tag{3}$$

where Y_{phys} is the physical sputtering yield of carbon calculated by the revised Bohdansky formula:

$$Y_{\text{phys}}(E_0) = QS_{\text{n}}(E_0)\left[1 - (E_{\text{th}}/E_0)^{2/3}\right](1 - E_{\text{th}}/E_0)^2, \tag{4}$$

where E_0 is the incident energy, Q and the threshold energy E_{th} are fitting parameters. $S_n(E_0)$ is the nuclear stopping power, which can be approximated by:

$$S_n(E_0) = 0.5\ln(1 + 1.2288(E_0/E_{\text{TF}}))\Big/\Big[E_0/E_{\text{TF}} + 0.1728(E_0/E_{\text{TF}})^{1/2} + 0.008(E_0/E_{\text{TF}})^{0.1504}\Big], \tag{5}$$

where E_{TF} is the Thomas–Fermi energy.

Y_{surf} is the physical sputtering of weakly bound sp^3 CD_n groups at the surface:

$$Y_{\text{surf}}(E_0, T) = c^{\text{sp3}}Y_{\text{des}}(E_0)/[1 + \exp((E_0 - E_1)/40)], \tag{6}$$

where Y_{des} is the physical sputtering yield of carbon as given by formula (4), however with lower threshold energy E_{des}. The term in the denominator restricts this process to energies below E_1. At energy above E_1 the erosion yield resulting from this process decreases owing to the higher penetration depth of the incident particles and sp^3 formation at the and of the ion range. The concentration of sp^3 complexes can be calculated by formula:

$$c^{\text{sp3}} = C[2 \times 10^{-32}\phi_{\text{D}} + \exp(-E_{\text{therm}}/kT)] / \{2 \times 10^{-32}\phi_{\text{D}} + [1 + (2 \times 10^{29}/\phi_{\text{D}}) \times \exp(-1.8/kT)]\exp(-E_{\text{therm}}/kT)\} \tag{7}$$

with

$$C = 1/[1 + 3 \times 10^7 \exp(-1.4/kT)], \tag{8}$$

where ϕ_{D} is given in deuterons $\times$ m^{-2} $\times$ s^{-1} and all the activation energies are in electronvolts. The thermally activated process, Y_{therm}, which leads to a maximum of the chemical yields at temperatures between 700 and 950 K, is described by formula:

$$Y_{\text{therm}} = c^{\text{sp3}} \times 0.033\exp(-E_{\text{therm}}/kT)/[2 \times 10^{-32}\phi_{\text{D}} + \exp(-E_{\text{therm}}/kT)]. \tag{9}$$

The thermal chemical erosion is enhanced by damage production with Y_{dam} is given by formula (4), however with lower threshold energy for damage production E_{dam}. The constant D in formula (3) varies with the mass of the projectiles used.

To apply the analytical model of Ref. [2] for the description of the present experimental data several assumptions were accepted: (1) the physical sputtering of weakly bound sp^3 CD_n groups at the surface by helium ions will contribute to the sputtering yield Y_{surf}, (2) the value $E_1 = 150$ eV for He^+ is chosen taking into account that at this energy the helium penetration depth in carbon calculated by TRIM code is same as that for 65 eV D^+, (3) the energy deposition due to impact of helium ions will change the multiplicative term $(1 + DY_{\text{dam}})$ in the formula for the chemical erosion yields.

Under simultaneous bombardment by helium and deuterium ions N_{C} can be expressed as:

$$\begin{aligned} N_{\text{C}} = {} & N_{\text{He}}(Y_{\text{He-phys}} + Y_{\text{He-surf}}) \\ & + N_{\text{D}^+}[Y_{\text{D-phys}}(200\text{ eV}) + Y_{\text{D-surf}}(200\text{ eV})] \\ & + 2N_{\text{D}_2^+}[Y_{\text{D-phys}}(100\text{ eV}) + Y_{\text{D-surf}}(100\text{ eV})] \\ & + 3N_{\text{D}_3^+}[Y_{\text{D-phys}}(67\text{ eV}) + Y_{\text{D-surf}}(67\text{ eV})] \\ & + N_{\text{D}}Y_{\text{therm}}[1 + (N_{\text{He}}/N)\mathbf{D}_{\text{He}}Y_{\text{He-dam}} \\ & + (N_{\text{D}^+}/N)D_{\text{D}}Y_{\text{D-dam}}(200\text{ eV}) \\ & + (2N_{\text{D}_2^+}/N)D_{\text{D}}Y_{\text{D-dam}}(100\text{ eV}) \\ & + (3N_{\text{D}_3^+}/N)D_{\text{D}}Y_{\text{D-dam}}(67\text{ eV})]. \end{aligned} \tag{10}$$

Here Y_{He}, Y_{D} are the erosion yields due to impact of He and deuterium, respectively. The weak energy dependence of the total sputtering yield in the energy range from 50 to 200 eV/D has been observed [15,21], but the contributions of each sputtering mechanism strongly depend on the ion energy, that is why formula (10) includes terms for all the deuteron energies at the present experimental conditions. Then the effective erosion yield can be calculated as:

$$\begin{aligned} Y_{\text{eff}} = {} & \alpha(Y_{\text{He-phys}} + Y_{\text{He-surf}}) + \alpha_1[Y_{\text{D-phys}}(200\text{ eV}) \\ & + Y_{\text{D-surf}}(200\text{ eV})] + \alpha_2[Y_{\text{D-phys}}(100\text{ eV}) \\ & + Y_{\text{D-surf}}(100\text{ eV})] + \alpha_3[Y_{\text{D-phys}}(67\text{ eV}) \\ & + Y_{\text{D-surf}}(67\text{ eV})] + (1 - \alpha)Y_{\text{therm}}[1 \\ & + \alpha D_{\text{He}}Y_{\text{He-dam}} + \alpha_1 D_{\text{D}}Y_{\text{D-dam}}(200\text{ eV}) \\ & + \alpha_2 D_{\text{D}}Y_{\text{D-dam}}(100\text{ eV}) \\ & + \alpha_3 D_{\text{D}}Y_{\text{D-dam}}(67\text{ eV})], \end{aligned} \tag{11}$$

where α, α_1, α_2, α_3, are the relative fractions of each species that are the function of the $\phi_{\text{He}}/\phi_{\text{D}}$ ratio, see Table 1. In the formula (11) underlined terms are the results of interactive processes that appear only in case of the simultaneous bombardment and can be associated

with: (1) the physical sputtering of weakly bound sp^3 CD_n groups by helium ions at the surface and (2) the energy deposition leading to atomic displacement with corresponding breaking of C–C bonds resulting in the creation of active sites for reaction with incoming deuterium, respectively. All parameters needed for calculation are given in Table 2. The calculated and measured effective sputtering yields gradually increase with increasing of ϕ_{He}/ϕ_D up to 0.5 at temperature of 450 K and almost stable when the ϕ_{He}/ϕ_D increases above 0.1 at 973 K, whereas the experimental results do not agree with model even in tendency at 773 K, as it is seen in Fig. 4. The analytical description allows to explain the observed dependence of the erosion yield on helium/deuterium flux ratio at temperatures of 450 and 973 K, as shown in Fig. 5. In this figure only the erosion yields due to interactive processes are plotted.

The physical sputtering of CD_n groups by helium ions, $Y_1 = \alpha Y_{\text{He-surf}}$ is the process responsible for erosion yield enhancement with increasing of the ϕ_{He}/ϕ_D ratio at 450 K, when the concentration of these groups at the surface is high. At this temperature Y_3, open rectangles with solid line, almost coincides with Y_1, filled rectangles with dot line. With increasing temperature above 700 K incoming deuterium atoms may recombine with absorbed atoms, thus interrupting the hydrogenation process and reducing the sp^3 concentration [1]. Therefore, at 973 K the contribution of Y_{surf}, filled triangles with dot line, is negligible due to low concentration of sp^3 complexes, whereas, the term $Y_2 = \alpha(1-\alpha)Y_{\text{therm}}D_{\text{He}}Y_{\text{He-dam}}$, filled triangles with dash line, decreases with increasing relative He fraction in the ion beam $\phi_{He}/\phi_D > 0.1$. The concentration of sp^3 complexes still is high at 773 K. The filled rectangles with dot line are overlapped with filled circles with dot line, but the dominant erosion yield at this temperature is the term Y_2, filled circles with dash line, that increases with increasing flux ratio at this temperature. The contribution of the Y_3 in the effective erosion yield at 773 K in Fig. 4 is counterbalanced by decrease of the calculated chemical erosion with increasing of ϕ_{He}/ϕ_D ratio. As a result, the calculated effective erosion yield at 773 K shows the same trend as for 973 K, that is not observed experimentally. The presence of the deuterium neutrals in the ion beam that is not taking into account in Eqs. (10) and (11) may leads to the observed discrepancy.

Table 2
Parameters for the sputtering yields calculation

Parameter	Deuterium	Helium	Remarks
E_{th} (eV)	27	25.4	Ref. [22,23]
Q	0.1	0.169	Ref. [22,24]
E_{TF} (eV)	447	1087	Ref. [22,23]
E_{dam} (eV)	15	15	Ref. [2]
E_{des} (eV)	2	2	Ref. [2]
D	125	62.5	Ref. [1,2]
E_1 (eV)	65	150	Ref. [2]
E_{therm} (eV)	1.7	–	Ref. [1]

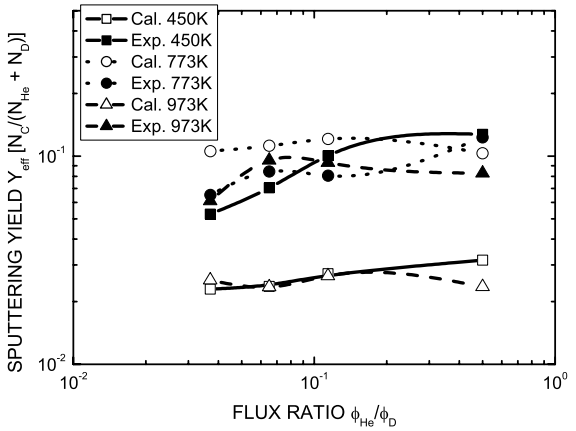

Fig. 4. The effective erosion yield of 1D-CFC measured by weight loss method and calculated using analytic model from Ref. [2].

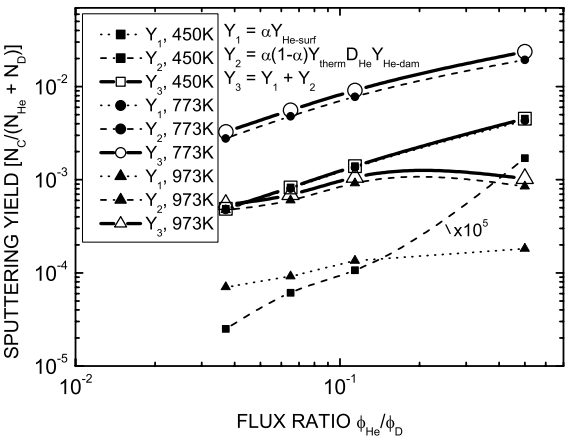

Fig. 5. Contribution of the interactive processes in the total erosion yield under simultaneous bombardment by helium and deuterium ions.

4. Conclusions

The erosion behavior of 1D-CFC due to the simultaneous bombardment of He^+, D_3^+, D_2^+ and D^+ ions with the known fluxes of the each species has been studied as a function of the ϕ_{He}/ϕ_D flux ratio.

Enhancement of the effective erosion yield with increasing of the ϕ_{He}/ϕ_D flux ratio was observed at 450 K and 773 K, whereas at the sample temperature of 973 K the effective erosion yield was almost stable when the ϕ_{He}/ϕ_D increased above 0.1.

The analytical description of the graphite erosion by hydrogen ions from Ref. [2] was applied to describe the present experimental data, that allows to explain the observed experimental results. Enhancement of the effective erosion yield are the results of interactive processes that appear only in case of the simultaneous bombardment and can be associated with: (1) the physical sputtering of weakly bound sp^3 CD_n groups by helium ions at the surface and (2) the energy deposition leading to atomic displacement with corresponding breaking of C–C bonds and creation of active sites for reaction with incoming deuterium.

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