

Supporting Information

Gas-Phase Synthesis of the Elusive Cyclooctatetraenyl Radical (C_8H_7) via Triplet Aromatic Cyclooctatetraene (C_8H_8) and Non-Aromatic Cyclooctatriene (C_8H_8) Intermediates

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Experimental Procedures

Experimental: The elementary gas phase reaction of atomic carbon ($C({}^{3}P_{j})$) with 1,3,5-cycloheptatriene ($C_{7}H_{8}$; TCI America; > 95 %) was studied under single collision conditions in a crossed molecular beams machine.^[1-4] A pulsed molecular beam of helium-seeded ground state carbon atoms was produced in the primary source chamber by ablating a rotating graphite rod with a 266 nm output of a Nd:YAG laser (Quanta Ray Pro 270, Spectra Physics) operating at 30 Hz and 10-13 mJ per pulse. The ablated species were seeded in helium carrier gas (Airgas; 99.9999 %) with a backing pressure of 3040 Torr released from a piezoelectric pulsed valve operating at a repetition rate of 60 Hz, a pulse width of 80 µs, and a peak voltage of -400 V. The pulsed carbon molecular beam passed through a skimmer and was velocity selected by a four-slot chopper wheel rotating at 120 Hz. The on-axis characterization of the pulsed carbon beam at m/z 12 (C⁺) indicated a peak velocity (v_p) of 2537 ± 122 ms⁻¹ and a speed ratio (S) of 3.6 ± 0.5. The carbon beam was crossed perpendicularly by a pulsed, supersonic cycloheptatriene (C7H8) beam seeded in argon (Airgas; 99.9999 %) generated by a second piezoelectric pulsed valve at a backing pressure of 550 Torr with a repetition rate of 60 Hz, a pulse width of 80 µs, and a peak voltage of -400 V in the secondary source chamber. The cycloheptatriene molecular beam was characterized through v_p and S to be 588 ± 20 ms⁻¹ and 13.7 ± 0.4, respectively, yielding a collision energy of 32 ± 4 kJ mol⁻¹. The reactively scattered products were detected with a rotatable, triply differentially pumped, quadrupole mass spectrometer (QMS) operated in the time-of-flight (TOF) mode. The neutral products were ionized with an electron impact ionizer at an electron energy of 40 eV and an emission current of 1.4 mA. The mass-selected ions were accelerated towards an aluminum coated stainless steel target held at -22.5 kV; an electron cascade was initiated, when an ion hit the aluminum coated target, and was accelerated toward an aluminum coated organic scintillator generating a photon pulse to be detected by a photomultiplier tube (PMT; Burle Model 8850; -1.35 kV). The signal from the PMT was then filtered by a discriminator (Advanced Research Instruments, Model F-100TD) at a level of 1.6 mV prior to directing into a Stanford Research System SR430 Multi-Channel Scaler to record the TOF spectra. The detector is rotatable within the plane as defined by the carbon and cycloheptatriene beams. Up to 2×10⁶ TOF spectra were recorded at each angle. The laboratory angular distribution was acquired by integration of the TOF spectra at each collection angle and normalizing for the accumulation time and fluctuations in the carbon beam intensity. To gain additional information on the chemical dynamics and underlying reaction mechanism, TOF spectra and the laboratory angular distribution were transformed from the laboratory frame into the center-of-mass reference frame using a forward-convolution routine.^[5,6] This approach initially presumes an angular flux distribution $T(\theta)$ and translational energy flux distribution $P(E_T)$ in the center-of-mass system assuming mutual independence. The laboratory data (TOF spectra and laboratory angular distribution) are then calculated from the T(θ) and P(E_T) and convoluted over the apparatus functions to obtain a simulation of the experimental data. The crucial output of this fitting routine is the product flux contour map, $I(\theta, u) = P(u) \times T(\theta)$, which reports the intensity of the reactively scattered products (I) as a function of the center-of-mass scattering angle (θ) and product velocity (u). This plot is called the reactive differential cross section and can be seen as the image of the chemical reaction.

Theoretical: Geometries of the 1,3,5-cycloheptatriene reactant, C₈H₇ radical products, and various triplet intermediates and transition states on the C₈H₈ PES were optimized at the hybrid density functional B3LYP level of theory^[7,8] with the 6-311G** basis set. The same B3LYP/6-311G** method was used to calculate vibrational frequencies, which were utilized to compute zero-point energy (ZPE) corrections, to identify the stationary structures as local minima or first-order saddle points, and to compute rate constants for unimolecular reaction steps. To evaluate more accurate relative energies, single-point energies at the optimized geometries were recalculated using the G3(MP2,CC)//B3LYP approach^[9,10], which provides accuracy for relative energies within 10 kJ mol⁻¹. The electronic structure calculations were carried out using the GAUSSIAN 09^[11] and MOLPRO 2010^[12] program packages. Relative various H loss channels from C₈H₈ under single-collision conditions were vields for computed usina Rice-Ramsperger-Kassel-Marcus (RRKM) theory^[13-15]. The rate constants were calculated as functions of available internal energy, where the internal energy was taken as a sum of the energy of chemical activation in the C(³P) + 1,3,5-cycloheptatriene reaction and the collision energy, assuming that it predominantly converts into internal vibrational energy. Since the RRKM calculations were performed for single-collision conditions at zero-pressure limit, only a single total-energy level was considered throughout.^[16] The harmonic approximation was employed to compute numbers and densities of states. Using the calculated rate constants, branching ratios of various channels were computed by solving first-order kinetic equations within the steady-state approximation for unimolecular isomerization and fragmentation steps of the initial reaction intermediates i1 and i3.



Figure S1. Molecular orbital diagrams (a) for the decomposing intermediate **i2 (**2,4,7-cyclooctatriene, C₁) on the triplet surface, (b) for the 1,2,4,7-cyclooctatetraenyl radical (C₈H₇, C₂) product **p1**, and (c) comparison of molecular orbital energy levels of **i4**, **i2**, and **p1**. The energy levels are shown in kJ mol⁻¹.



(b)

Figure S1. (Continued)



(c)

Figure S1. (Continued-2)



Figure S2. Bond lengths in the main product **p1** and bond lengths and bond angles in the atomic hydrogen loss transition state **i2-p1** are given in pm and degrees, respectively.

Discussion about the singlet surface

Addressing a possible role of the singlet surface, we note that in the previous studies of reactions of ground state atomic carbon with unsaturated hydrocarbons, a spin-flip via intersystem crossing (ISC) was found to play an important role only for the carbon plus acetylene reaction, in which the system crosses over to the singlet C_3H_2 surface and can form the $C_3(X^1\Sigma_q+) + H_2$ products ^[17] This unique behavior was attributed to a relative long lifetime of the decomposing C3H2 complexes caused by the fact that the products on the triplet surface, c- and I-C₃H are nearly thermoneutral or slightly exoergic. Only their relatively long lifetime allows the triplet-singlet ISC to occur. Another reaction of ground state carbon atoms, in which ISC might play a role is carbon plus benzene, but only in bulk conditions, not in gas phase molecular beam experiments.^[17] In the reactions involving larger alkynes and alkenes, much more excergic products can be formed and the lifetime of the triplet intermediates is shorter and hence no evidence of ISC was observed so far. The present reaction is highly exoergic as well, and our statistical (RRKM) calculations indicate that the lifetimes of the triplet intermediates are short, in the range of 25-75 ns at the experimental collision energy. The predominant role of the triplet surface is evidenced by the existence of an exit barrier for the hydrogen atom loss, based on the shape of the translational energy distribution; the hydrogen atom loss from any singlet intermediate is barrierless. Although the role of the singlet surface cannot be completely ruled out, it is unlikely. First, according to our calculations of the singlet surface, the singlet-triplet energy gaps for all critical triplet intermediates involved are large, 161, 135, and 127 kJ mol⁻¹ for i2, i3, and i4, respectively, with singlet states lying lower in energy (Supporting Information). The energy gaps that high make ISC a less likely process; for comparison, in C_3H_2 ISC was found to be efficient between triplet and singlet structures separated only by 50 kJ mol⁻¹ with triplet being more favorable and still only at low collision energies.[18] Second, if singlet isomers are formed via ISC, their calculated lifetimes with respect to hydrogen atom elimination are up to three orders of magnitude longer (2-60 µs). Therefore, the C₈H₈ singlet isomers, if produced, could have survived in the reaction chamber and then be ionized and detected. This however is not the case in the present experiment.



Figure S3. Fragment of the singlet C_8H_8 PES that can be accessed by triplet-singlet intersystem crossing in the vicinity of triplet intermediates **i2-i5**. All relative energies are shown in kJ mol⁻¹ with respect to the C(³P) + C₇H₈ reactants.

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Structure	Coordinates		Frequencies		
1,3,5-cycloheptatriene	6 0 0.255703 -1.417716	0.680838	214.2007	295.2611	356.2407
	6 0 0.255703 -1.417716	6 -0.680838	417.5620	431.8578	610.4532
	60-0.248870 -0.360149	9 -1.527547	667.8938	725.3930	760.9588
	6.0-0.248870 -0.360149	1.527547	813,2356	890,4976	921,1552
	60-0248870 0951946	5 -1 221424	938 8788	960 4329	977 9240
20	60-0248870 0951946	1 221424	983 2979	1001 3070	1040 0880
÷	6 0 0 456430 1 484915	0,00000	1067 8/27	1210 3505	1221 / 227
		5 1 177061	1247 3754	1260 2/88	1327 5448
		5 1 177061	1247.3734	1/2/ 200	1475 5665
			1305.7004	1424.3000	1475.5005
		1 -2.476233	1480.0003	1572.7814	1003.2048
	10-0.680782 -0.669577	1 2.476233	1670.9291	2992.9632	3088.3701
	10-0.757659 1.654307	-1.875029	3124.8049	3130.6974	3137.5103
	10-0.757659 1.654307	1.875029	3152.1701	3158.4702	3163.1804
	1 0 1.499013 1.140161	0.000000			
	1 0 0.466769 2.575908	3 0.000000			
i0 (ts i3-i3)	6 0 -1.753400 0.083737	7 -0.136878	514.4247 <i>i</i>	74.0510	158.2441
0.0	60-0.424030 -1.456777	7 -0.385587	237.3086	350.3716	399.7129
	6 0 0.954245 -1.472230	0.156966	447.3517	493.9021	634.2829
	6 0 -1.092721 1.347282	2 0.159625	673.2567	701.9547	733.6203
	6 0 1.746492 -0.405500	0.228472	779.6552	816.0227	871.0929
6.90 O	6 0 0.205352 1.703428	0.170440	919.4117	952.7248	966.3657
è	6 0 1.395357 0.939049	-0.375517	969.3894	1006.7080	1037.1564
	10-2.664982 0.172481	-0.739634	1106.3208	1188.6641	1204.2177
	1 0 -0 596465 -1 752569	-1 423320	1241 1901	1258 1401	1284 6884
	1 0 1 268387 -2 418847	0.595822	1358 5495	1374 2058	1386 4509
	1 0 -1 806697 2 138279	0.396771	1421 6130	1475 3600	1641 4935
		0 711424	1698 2803	2977 3152	3015 0408
		0.711424	3055 8010	2077 2313	3084 4204
		0.405410	2100 2240	21/6 7205	2151 2710
		-0.310000	5109.5549	3145.7305	3131.2710
		1 0 470447			
14	60-1.500379 -1.193244	+ 0.472447	457 4400	004 5045	077 5504
11	60-1.435168 -0.628942	2 -0.118694	157.4186	231.5945	277.5564
900 B	60-1.190451 1.016776	0.058444	349.2926	402.3314	462.0406
6-60 0-0	60 0.088109 1.609399	0 -0.331692	533.8174	609.6206	663.9902
V I	6 0 1.335493 1.111556	5 -0.155109	716.7522	809.5799	825.8966
	6 0 0.999542 -1.335525	5 0.178482	862.5362	874.8608	910.0382
- en	6 0 1.697332 -0.200409	0.353567	956.2503	972.1817	991.6852
e e	10-2.384628 -0.847008	3 -0.619055	1000.1256	1012.7027	1023.4316
	10-2.056177 1.598290	0 -0.264047	1094.5326	1207.6960	1222.4359
	1 0 0.009700 2.601474	-0.768474	1247.1607	1258.7794	1301.0822
	1 0 2.166080 1.769560	0 -0.398851	1346.5958	1396.7529	1424.2942
	10 1.385719 -2.249644	0.617997	1461.6995	1472.5682	1604.0922
	1 0 2.645522 -0.260905	5 0.882499	1675.3845	2981.8560	3024.3013
	6.0-1.421674 0.050763	3 1.135944	3053.3692	3060.4089	3125.0512

Table S1. Optimized Cartesian coordinates (in Å) and vibrational frequencies (in cm⁻¹) for all stationary structures.

60.0.2020412 -1.45129 -0.636965 313.0315 3151.5041 3163.6207 10 0.062444 -1.45605 -1.675065 -						
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1 0 -2.919300 -0.000000 -0.000000 3120.8971 3135.5145 3135.6676		1 0 -2.919300 -0.000000 -0.000000	3120.8971	3135.5145	3135.6676	

	1 0 2.064257 2.0642	57 -0.000000	3151,7942	3151.8025	3161.4652
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6	6 0 0.433800 1.6381	97 -0.169265	349.9398	388.5809	413.8813
	60-0.969791 1.3968	32 0.061399	465.8956	557.0275	674.6521
	60-1.840915 0.2643	66 0.133283	702.0670	763.8722	769.9828
	60-0.233457 -1.4917	53 -0.328802	798,7918	870.8140	879.6484
Ψ 6 9-6	6 0 -1.605709 -1.0730	66 -0.087984	905.5596	928.6939	972.4380
	1 0 2.357348 -0.9490	43 -0.904281	980.0195	1054.1886	1071.3350
	1 0 0.626649 2.7016	78 -0.331915	1145,4173	1164.3543	1233.4743
	1 0 -1 513212 2 3321	32 0 168866	1254 5038	1301 4629	1339 3551
	1 0 -2 880765 0 5427	78 0 296039	1350 5018	1381 6309	1440 7585
	1 0 0 031559 -1 8817	32 -1 312764	1467 2670	1522 5444	1549 6341
	10-2451013 -17467	59 -0 195993	1656 2376	3016 9981	3058 8405
	6.0 1 568198 0 9014	74 -0 163943	3059 9529	3090 3116	3113 0547
	6 0 0 722504 -1 0800	85 0 518264	3138 8252	3142 8537	3151 2672
	1 0 0 414709 -0 7394	31 1 503208	0100.0202	0112.0001	010112012
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p1	60 0466520 -1 2078	74 -1 282798	162 0550	231 5994	245 5635
	6 0 -0 466520 1 2078	74 -1 282798	323 7634	433 7977	470 7325
	6 0 -0 000000 1 9244	92 -0 104091	551 4793	611 5109	637 3983
	6 0 -0 000000 -1 9244	92 -0 104091	735 7008	751 6977	801 9095
	6.0.0.200438 1.3184	92 1.118549	819.9044	832,1372	875.5913
○- ● ●	6.0-0.200438 -1.3184	92 1.118549	903,2436	969.1877	970.6350
	6.0.000000 -0.0000	00 1.621412	979.2282	981.2413	1110,7879
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_	1 0 1.242714 -1.6457	23 -1.906010	1288.2997	1373.7356	1426.8330
	10-1.242714 1.6457	23 -1.906010	1443.4682	1456.0336	1528.9128
	1 0 0.072787 3.0072	30 -0.147417	1551.6436	1966.8832	3105.8125
	10-0.072787 -3.0072	30 -0.147417	3116.9185	3128.3773	3129.0829
	1 0 0.462419 2.0168	69 1.911406	3135,4608	3152.9946	3156.0729
	10-0.462419 -2.0168	69 1.911406			
	1 0 0.000000 -0.0000	00 2.709056			
p2	6 0 0.326225 -1.3832	91 -0.466860	148.6530	198.0393	253.2148
	60-0.741919 -1.2224	51 0.356868	326.8161	418.0699	432.0069
U. Go	6 0 0.901978 1.4108	95 0.145325	499.9787	606.4770	694.5315
bý L	60-1.919003 -0.4964	41 -0.110337	714.6279	752.3199	793.3899
	6 0 -0.396784 1.5946	92 -0.272679	807.7051	817.3898	854.8598
	6 0 -1.530246 0.7422	13 -0.446869	892.9053	983.3972	986.5726
	10-0.599888 -1.3008	25 1.433387	1006.7617	1053.0480	1114.0004
	10-2.954293 -0.8157	25 -0.047221	1210.1646	1265.6197	1281.8491
	1 0 0.156202 -1.5075	99 -1.534645	1302.8064	1330.7233	1428.7655
	10 1.418291 2.3571	92 0.293637	1459.3787	1496.0961	1569.1003
	10-0.642882 2.6429	51 -0.442785	1606.4189	1625.6371	3092.2331
	6 0 1.653743 -1.0144	34 -0.027060	3114.1743	3115.7661	3120.7597
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b 1+2 6 0 -1.644312 -0.405102 -0.021540 552.68511 201.1333 215.0061 6 0 -0.806477 1.433364 0.078733 272.9084 305.64453 420.8391 6 0 -0.55877 0.705914 0.132217 -0.350238 478.4463 611.7266 630.8096 6 0 -0.558787 0.0541170 0.352047 1.561066 0.72223 821.8742 806.8170 872.4095 1 0 -0.558787 0.0541170 0.352087 0.0641170 0.352874 112.04.9037 100.63027 100.63027 100.63027 100.42214 121.4204 9037 100.7575 100.7575 100.7575 100.63027 100.6		1 0 2.810487 0.596	016 0.645332			
Bit D = 0.500472 1.433344 0.08783 272.9094 305.6433 420.8391 6 0 = 0.534181 1.560217 0.705914 0.132941 665.3294 760.3320 766.4415 6 0 = 0.550317 0.705914 0.132941 665.3294 760.3320 766.4415 6 0 = 0.550317 0.267406 0.525031 0.382254 992.2614 1006.3027 1043.0214 1 0 = 0.2674066 0.525031 0.382254 1300.6770 1178.4771 1204.9037 1 0 0 -0.578317 0.368744 1333.1782 1470.6166 1428.9701 1 0 0 -0.682747 0.368744 1333.1792 1472.5152 121.0135 1 0 0 -0.682747 0.368744 1333.01702 1472.5152 152.0175 1 0 0 -0.682747 0.365759 0.442440 1463.6302 1472.5152 152.0175 1 0 -0.253337 0.0827447 143350 0.247349 305.3736 3065.2294 305.2298 1 0 -0.25337 0.362484 0.227087 344.6238 3129.7770 3145.6386 3168.8938 <t< th=""><th>ts i1-i2</th><th>60-1.644312 -0.405</th><th>102 -0.021540</th><th>552.6851<i>i</i></th><th>201.1393</th><th>215.6061</th></t<>	ts i1-i2	60-1.644312 -0.405	102 -0.021540	552.6851 <i>i</i>	201.1393	215.6061
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10 10 1.1193 1.1193 1.1204 <td< th=""><th></th><th>10-2.674096 -0.525</th><th>031 -0.382954</th><th>972.9614</th><th>1006.3027</th><th>1043.0214</th></td<>		10-2.674096 -0.525	031 -0.382954	972.9614	1006.3027	1043.0214
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$ \begin{array}{c} 6\ 0\ 1.541148\ 1.021412\ -0.033343 & 273.1883\ 320.9582\ 398.6337 \\ 6\ 0\ 0.243490\ 1.690034\ 0.108842 & 434.6949\ 533.7346\ 586.3253 \\ 6\ 0\ -1.080235\ 1.352619\ 0.014616\ 683.0331\ 732.1420\ 740.3156 \\ 6\ 0\ -1.440696\ -1.145851\ -0.032823\ 758.6500\ 810.9870\ 864.8179 \\ 6\ 0\ -1.831468\ 0.127444\ -0.249280\ 871.5112\ 904.6243\ 949.1967 \\ 1\ 0\ 1.114734\ -2.157897\ -1.066420\ 974.9028\ 999.0468\ 1015.0641 \\ 1\ 0\ 2.370404\ 1.728091\ -0.020967\ 1095.7218\ 1124.2184\ 1173.7201 \\ 1\ 0\ 0.385088\ 2.755410\ 0.277798\ 1222.9337\ 1339.9445\ 1343.8887 \\ 1\ 0\ -1.737000\ 2.215139\ 0.107313\ 1381.2819\ 1383.4528\ 1429.0962 \\ 1\ 0\ -2.051863\ -1.986452\ -0.343687\ 1469.2353\ 1510.5578\ 1601.7138 \\ 1\ 0\ -2.051863\ -1.986452\ -0.343687\ 1469.2353\ 1510.5578\ 1601.7138 \\ 1\ 0\ -2.831202\ 0.291834\ -0.647279\ 1631.7449\ 1985.1117\ 3097.1443 \\ 6\ 0\ 1.878644\ -0.265771\ -0.243187\ 3109.0596\ 3120.8772\ 3138.2474 \\ 6\ 0\ -0.123147\ -1.307079\ 0.581576\ 3145.3061\ 3165.2483\ 3180.8680 \\ \end{array}$	ts i2-i5	6 0 1.119892 -1.464	941 -0.234318	1964.5193 <i>i</i>	172.4095	212.8337
6 0 0.243490 1.690034 0.108842 434.6949 533.7346 586.3253 6 0 -1.080235 1.352619 0.014616 683.0331 732.1420 740.3156 6 0 -1.440696 -1.145851 -0.032823 758.6500 810.9870 864.8179 6 0 -1.831468 0.127444 -0.249280 871.5112 904.6243 949.1967 1 0 1.114734 -2.157897 -1.066420 974.9028 999.0468 1015.0641 1 0 2.370404 1.728091 -0.020967 1095.7218 1124.2184 1173.7201 1 0 0.385088 2.755410 0.277798 1222.9337 1339.9445 1343.8887 1 0 -1.737000 2.215139 0.107313 1381.2819 1383.4528 1429.0962 1 0 -2.051863 -1.986452 -0.343687 1469.2353 1510.5578 1601.7138 1 0 -2.831202 0.291834 -0.647279 1631.7449 1985.1117 3097.1443 6 0 1.878464 -0.265771 -0.243187 3109.0596 3120.8772 3138.2474 6 0 -0.123147 -1.307079 0.581576 3145.3061 3165.2483 3180.8680	©	6 0 1.541148 1.021	412 -0.033343	273.1883	320.9582	398.6337
60-1.080235 1.352619 0.014616 683.0331 732.1420 740.3156 60-1.440696 -1.145851 -0.032823 758.6500 810.9870 864.8179 60-1.831468 0.127444 -0.249280 871.5112 904.6243 949.1967 10 1.114734 -2.157897 -1.066420 974.9028 999.0468 1015.0641 10 2.370404 1.728091 -0.020967 1095.7218 1124.2184 1173.7201 10 0.385088 2.755410 0.277798 1222.9337 1339.9445 1343.8887 10-1.737000 2.215139 0.107313 1381.2819 1383.4528 1429.0962 10-2.051863 -1.986452 -0.343687 1469.2353 151.5578 1601.7138 10-2.831202 0.291834 -0.647279 1631.7449 1985.1117 3097.1443 60 -0.123147 -1.307079 0.581576 3109.0596 3120.8772 3138.2474 60 -0.25562 -0.662279 1.436306 3145.3061 3165.2483 3180.8680		6 0 0.243490 1.690	034 0.108842	434.6949	533.7346	586.3253
60-1.440696 -1.145851 -0.032823 758.6500 810.9870 864.8179 60-1.831468 0.127444 -0.249280 871.5112 904.6243 949.1967 10 1.114734 -2.157897 -1.066420 974.9028 999.0468 1015.0641 10 2.370404 1.728091 -0.020967 1095.7218 1124.2184 1173.7201 10 0.385088 2.755410 0.277798 1222.9337 1339.9445 1343.8887 10 -1.737000 2.215139 0.107313 1381.2819 1383.4528 1429.0962 10 -2.051863 -1.986452 -0.343687 1469.2353 1510.5578 1601.7138 10 -2.831202 0.291834 -0.647279 1631.7449 1985.1117 3097.1443 6 0 1.878464 -0.265771 -0.243187 3109.0596 3120.8772 3138.2474 6 0 -0.123147 -1.307079 0.581576 3145.3061 3165.2483 3180.8680		6 0 -1.080235 1.352	619 0.014616	683.0331	732.1420	740.3156
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101.114734 -2.157897 -1.066420 974.9028 999.0468 1015.0641 102.370404 1.728091 -0.020967 1095.7218 1124.2184 1173.7201 100.385088 2.755410 0.277798 1222.9337 1339.9445 1343.8887 10-1.737000 2.215139 0.107313 1381.2819 1383.4528 1429.0962 10-2.051863 -1.986452 -0.343687 1469.2353 1510.5578 1601.7138 10-2.831202 0.291834 -0.647279 1631.7449 1985.1117 3097.1443 601.878464 -0.265771 -0.243187 3109.0596 3120.8772 3138.2474 60-0.123147 -1.307079 0.581576 3145.3061 3165.2483 3180.8680		6 0 -1.831468 0.127	444 -0.249280	871.5112	904.6243	949.1967
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100.3850882.7554100.2777981222.93371339.94451343.888710-1.7370002.2151390.1073131381.28191383.45281429.096210-2.051863-1.986452-0.3436871469.23531510.55781601.713810-2.8312020.291834-0.6472791631.74491985.11173097.1443601.878464-0.265771-0.2431873109.05963120.87723138.247460-0.123147-1.3070790.5815763145.30613165.24833180.8680100.055562-0.6622791.436306145.30613165.24833180.8680		1 0 2.370404 1.728	091 -0.020967	1095.7218	1124.2184	1173.7201
1 0 -1.7370002.2151390.1073131381.28191383.45281429.09621 0 -2.051863-1.986452-0.3436871469.23531510.55781601.71381 0 -2.8312020.291834-0.6472791631.74491985.11173097.14436 0 1.878464-0.265771-0.2431873109.05963120.87723138.24746 0 -0.123147-1.3070790.5815763145.30613165.24833180.86801 0 0.055562-0.6622791.436306		1 0 0.385088 2.755	410 0.277798	1222.9337	1339.9445	1343.8887
1 0 -2.051863 -1.986452 -0.343687 1469.2353 1510.5578 1601.7138 1 0 -2.831202 0.291834 -0.647279 1631.7449 1985.1117 3097.1443 6 0 1.878464 -0.265771 -0.243187 3109.0596 3120.8772 3138.2474 6 0 -0.123147 -1.307079 0.581576 3145.3061 3165.2483 3180.8680 1 0 0.055562 -0.662279 1.436306 1436306 3145.3061 3165.2483 3180.8680		10-1.737000 2.215	139 0.107313	1381.2819	1383.4528	1429.0962
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6 0 -0.123147 -1.307079 0.581576 3145.3061 3165.2483 3180.8680 1 0 0.055562 -0.662279 1.436306		6.0 1.878464 -0.265	771 -0.243187	3109.0596	3120.8772	3138.2474
1 0 0.055562 -0.662279 1.436306		6.0-0.123147 -1.307	079 0.581576	3145.3061	3165,2483	3180.8680
		1 0 0.055562 -0.662	279 1.436306			

	1 0 0 849581 -2 231044 0 784438			
to i2 i4	6.0.1.200224 1.262810 0.002282	1225 0001	161 1500	206 0465
15 13-14	0.0-1.300324 -1.202010 0.003302	1233.99017	101.1302	200.9405
	0 0 1.300315 -1.202815 0.003385	479.2593	479.9457	500.8535
9-0-0	6 0 1.921782 -0.074656 -0.391545	514.6335	561.2093	588.7415
	6 0 -1.921784 -0.074645 -0.391544	677.3650	728.7433	758.2713
	6 0 1.322203 1.147322 -0.140430	768.2148	799.0296	857.0159
9-9-9	6 0 -1.322194 1.147326 -0.140431	881.8930	957.6118	964.2532
6 6 G	6 0 0.000005 1.305088 0.470842	1016.6447	1032.3724	1054.3048
	10-1.858909 -2.196161 -0.013385	1126.7055	1138.1707	1212.5723
	1 0 1.858894 -2.196170 -0.013389	1227.4915	1268.5661	1354.1254
	1 0 2 941849 -0 100680 -0 760033	1377 7600	1382 4173	1415 1966
	10-2941851 -0 100660 -0 760031	1424 8703	1453 0093	1463 5983
	1 0 1 03/066 2 0/1836 -0 10811/	1522 2520	1522 5600	3028 0162
		2110 2722	2122.0009	3020.0102
	10-1.934949 2.041044 -0.190110	3119.2722	3123.0903	3147.3247
	10 0.000001 0.054317 1.079275	3149.9555	3172.3327	3173.2248
	10 0.000008 2.189310 1.117834			
	6 0 -0.000006 -1.213748 0.544001			
ts i 3-p1	6 0 1.306602 -1.194498 0.469659	817.0972 <i>i</i>	135.9328	209.9448
0.0.0	6 0 -1.117278 -1.394462 -0.391215	252.6986	263.7517	315.4424
~~~Q	6 0 -1.802325 -0.224524 -0.219815	364.9072	438.1839	474.9135
	6 0 1.808247 -0.019739 -0.284114	594.8832	616.5703	683.4220
	6 0 -1.290976 0.966340 0.459774	706.1935	767.1426	778.5382
	6 0 1.163531 1.151971 -0.375928	825,9203	879.6463	885.1486
	6 0 -0 083450 1 579030 0 304664	929 3887	953 9570	958 6140
•	1 0 1 968059 -1 650364 1 204814	1000 4480	1007 9364	1050 9320
	10-1557072 -2179678 -1001353	1104 9523	1235 7340	1259 7110
		1202 2061	1200.7040	1/10 0677
		1292.2001	1592.2542	1604 1921
		1440.0461	1020.0042	1004.1831
	10-2.020539 1.516007 1.041435	1004.0330	1/01.0202	3103.0149
	10 1.664451 1.951899 -0.915835	3107.6444	3121.2368	3123.9981
	1 0 0.051333 2.493318 0.881761	3132.7265	3143.1033	3144.9787
	6 0 0.096232 -1.647311 0.247346			
	10-0.558346 2.821846 -1.100383			
ts <b>i4-p2</b>	6 0 0.328776 -1.402089 -0.414982	884.5394 <i>i</i>	149.8632	189.9789
6 9	6 0 -0.716530 -1.294129 0.431630	228.0337	302.7330	339.5028
0-0	6 0 0.860273 1.437083 -0.010370	379.3859	433.3882	456.4487
	6 0 -1.935128 -0.622953 -0.059395	478.8702	606.9742	668.5585
	6 0 -0.491849 1.576037 -0.181449	716.8033	739.9778	777.3498
	6.0 -1.626225 0.680811 -0.196253	793,4810	804,7939	886.6231
6	10-0.544938 -1.307772 1.506292	915.3683	963,7065	989.0747
	1 0 -2 931055 -1 038029 -0 164671	1004 5843	1050 4618	1098 6439
	1 0 0 129003 -1 435837 -1 484818	1210 9209	1242 6492	1271 4754
		1317 2/72	1335 2000	1/16 5222
		1451 0224	1533.2099	1540 0291
		1401.0224	1002.9440	1040.0301
		1002.4035	1029.30/3	3101.9740
	0 1.071015 -0.983694 -0.012300	3111.5103	3119.8765	3120.7316
	10 2.529241 -1.647329 0.042035	3136.9229	3153.2233	3165.8295
	6 0 1.822930 0.346307 0.179229			

	4 0 0 00 4050 0 704				
	10 2.824259 0.7014	179 0.414056			
ts i4-i5	6 0 -1.900781 -0.712	805 -0.141421	1783.8539 <i>i</i>	166.6953	199.1734
0 0	6 0 -0.623791 1.5622	221 0.204146	255.7894	339.6239	381.4359
	6 0 0.764685 1.4933	395 -0.132655	428.8496	478.0992	567.9823
	6 0 1.750519 0.4644	401 -0.182537	615.2970	687.3586	740.8663
I I I I	6.0.0.439838 -1.524	324 0.355770	764 6123	790 7113	802 1503
	6.0 1.706609 -0.890	S65 0 128124	832 1621	873 0001	006 7025
		000 1 052121	060 6071	070.0001	000.7323
·			909.0971	979.4395	900.4201
	10-0.897191 2.532	211 0.621547	1048.5445	1090.9920	1178.9186
	1 0 1.213593 2.4822	290 -0.185158	1245.7430	1274.4208	1285.7436
	1 0 2.751681 0.8627	782 -0.338098	1343.9740	1364.8997	1427.0851
	1 0 0.252257 -2.096	094 1.264315	1466.4908	1506.0365	1522.8594
	10 2.644613 -1.394	190 0.343609	1594.9755	2106.6911	3063.8088
	6 0 -1.694257 0.695	743 0.112567	3083.9368	3106.2754	3111.8081
	6.0-0.563221 -1.184	958 -0 491510	3135 4958	3151 3857	3181 2150
	10-0232379 -0705	766 -1 415737	0100.1000	0101.0001	010112100
	10-2872367 -1070	455 _0 458512			
to iE m2		714 0.052994	247 5500;	100 6740	192 41 41
IS 13-p2	60 1.947555 -0.439	714 -0.053664	247.0000	130.0740	102.4141
	60 0.371399 1.6193	330 -0.199722	203.8371	229.5117	255.2189
	6 0 -0.946620 1.3910	685 0.123106	334.6521	428.0094	441.3948
	6 0 -1.836592 0.238	771 0.261893	499.5826	617.3848	690.0555
6-9-6-90	60-0.258929 -1.381	523 -0.470720	722.5670	735.1197	790.4003
•••	60-1.617645 -1.060	090 -0.063657	801.8595	814.0358	884.7415
	10 2.975613 -0.7793	390 -0.113695	894.5217	979.3768	987.5312
	1 0 0.603451 2.6789	916 -0.307710	1006.7664	1052.2601	1105.4171
	10-1496225 2320	983 0 257543	1210 5510	1251 1645	1275 7928
	10-2852171 0516	367 0.536211	1312 0511	1331 0051	1424 3105
		364 -1 538133	1/58 1306	150/ 5108	1566 0111
	10-2450769 -1756	122 -0 112573	160/ 1806	1620 1781	3003 6310
		122 -0.112373	2446 6460	1020.1701	3093.0319
	60 1.530315 0.798		3110.0400	3117.9783	3134.4818
	60 0.770919 -1.206	150 0.388443	3135.3745	3149.7166	3161.6252
	10 0.589877 -1.269	50 1.458309			
	1 0 2.921995 -0.037	332 2.065913			
i2s	60-1.027071 -1.323	551 -0.501240	101.9384	241.2124	256.6913
	60 1.414394 -1.219	334 0.240586	307.5236	403.8603	466.9868
	6 0 1.905179 0.1359	991 0.060055	575.0806	595.8497	651.8875
	6 0 1.185156 1.2816	60 -0.092950	724.3102	770.7804	799.7970
T O	6 0 -1.428026 1.039	949 0.079946	832.8474	869.0281	890.3485
	6.0 -0.228606 1.642	158 -0 122131	902 0386	953 2430	993 6480
	6.0.0.220811 -1.537	366 -0.223332	1005 0733	1016 0428	1060 0627
	60-1 772076 -0 375	053 0 457467	1130 9095	1108 1727	1274 0878
		768 _1 /31253	1280 /120	1305 5538	1352 8010
		161 0 760406	1/1/5 0701	1440 0010	1470 2196
			1440.2701	1440.0910	14/3.3100
			1493.5349	1589.2023	1002.4040
	10 1.809922 2.161	16 -0.228452	1987.1002	3047.3414	3085.5970
	1 0 -2.288130 1.6854	401 -0.084122	3103.7374	3117.9590	3131.2707
	1 0 -0.337362 2.6973	337 -0.366675	3137.1934	3145.7949	3169.5528
	10-1.464957 -0.601	684 1.479905			

	10-2.855087 -0.51	9489	0.403245			
i3s	6 0 1.000113 -1.45	51398	-0.285141	150.8039	227.6670	253.3731
6	6 0 -1.428351 -1.04	19852	0.486524	302.2531	414.2074	454.4968
o I 🦻	6 0 -1.861257 0.08	31882	-0.363898	526.5034	559.5041	679.6499
	6 0 1.897891 -0.29	95593	-0.134161	713.5968	753.3316	785.7746
	6 0 -1.082146 1.17	2636	-0.458173	808.3978	866.5504	871.6061
G-€ 9-0	6 0 1.504185 0.96	6770	0.093445	942.7173	976.3278	987.9181
	6 0 0.108268 1.48	89191	0.436458	992.3921	998.4964	1053.7859
	6 0 -0.208697 -1.45	56521	0.225519	1119.3402	1213.9342	1252.2659
	10 1.317748 -2.25	53486	-0.947988	1267.9472	1289.9544	1354.6229
	10-2.038801 -1.43	30581	1.300939	1394.7698	1417.7238	1435.6499
	10-2.782989 0.01	5226	-0.937527	1478.3645	1652.9359	1665.5998
	10 2.954378 -0.47	75012	-0.313397	1981.9330	3012.3382	3059.2858
	10-1.368567 1.95	51273	-1.161251	3120.2840	3121.7685	3134.3819
	1 0 2.283787 1.72	3366	0.109245	3135.3918	3146.1354	3157.1023
	1 0 0.182186 2.57	8487	0.495071			
	10-0.127778 1.14	18034	1.451469			
i4s	60 0.375710 -1.56	64202	0.669220	179.1433	233.8262	263.8547
	6 0 0.375652 -1.56	64301	-0.669060	291.9058	363.6961	363.6961
	6 0 0.375595 1.56	64311	0.669069	649.9608	649.9608	673.1933
	60-0.375670-0.66	69221	-1.564212	683.7027	775.0243	812.5860
	6 0 0.375686 1.56	64212	-0.669211	812.5860	887.8073	941.3255
	6 0 -0.375613 0.66	69059	-1.564311	957.4068	957.4068	980.9307
	1 0 0.930905 -2.35	55212	-1.171197	1002.8831	1002.8831	1012.1621
	10-0.930949 -1.17	71437	-2.355054	1047.1945	1238.3034	1240.7181
	10 0.931009 -2.35	55031	1.171437	1254.9157	1254.9157	1387.8830
	1 0 0.930977 2.35	5055	-1.171414	1431.4549	1431.4549	1469.4725
	10-0.930847 1.17	1197	-2.355235	1694.9738	1694.9738	1699.9728
	6 0 -0.375634 -0.66	69069	1.564301	1717.7148	3100.8922	3104.3945
	10-0.930874 -1.17	71221	2.355212	3104.3945	3110.0730	3120.9842
	1 0 0.930816 2.35	5236	1.171220	3128.2777	3128.2777	3135.1456
	6 0 -0.375726 0.66	69211	1.564203			
	10-0.931037 1.17	′1413	2.355031			

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Table S2. Calculated Branching Ratios of Various Channels in the  $C_7H_8$  Reaction

							Colli	sion Energ	ıy, kJ mol⁻¹						
		0			10			20			32			40	
	from i1	from i3	2/3 <b>i1</b> , 1/3 <b>i3</b>	from <b>i1</b>	from i3	2/3 <b>i1</b> , 1/3 <b>i3</b>	from i1	from <b>i3</b>	2/3 <b>i1</b> , 1/3 <b>i3</b>	from i1	from i3	2/3 <b>i1</b> , 1/3 <b>i3</b>	from i1	from i3	2/3 <b>i1</b> , 1/3 <b>i3</b>
<b>p1</b> from <b>i2</b>	100.00	0.00	66.67	100.00	0.00	66.66	99.99	0.00	66.66	99.99	0.00	66.66	99.99	0.00	66.66
p1 from i3	0.00	2.04	0.68	0.00	2.49	0.83	0.00	2.99	1.00	0.00	3.65	1.22	0.00	4.16	1.39
p1 from i4	0.00	97.96	32.66	0.00	97.51	32.51	0.00	97.01	32.34	0.01	96.35	32.12	0.01	95.84	31.95
<b>p2</b> from <b>i4</b>	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
<b>p2</b> from <b>i5</b>	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

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