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# Difference of the Final Products of the Photochemical Reactions of Azanaphthalenes and Naphthalene in Mixed Crystals of Durene

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Final products of the photochemical reactions of quinazoline, quinoline, and naphthalene in 1,2,4,5-tetramethylbenzene (durene) mixed crystals are investigated on the basis of the phosphorescence and fluorescence spectra of the products. Quinazoline in durene is considered to transform into a benzene derivative, whereas naphthalene and quinoline tranform into anthracene derivatives. These products are different from a pyrazine derivative found in the case of quinoxaline, which is specific to the reaction in a durene mixed crystal. The cause of this difference is discussed in terms of the configurations of guests in the mixed crystals and the difference of the molecular structures.

One of the interesting features of the chemical reactions in single crystals is the generation of reaction products specific to single crystals.[1,2] Yamauchi, Terazima, and Hirota reported a unique product of the photochemical reaction of quinoxaline (Qx) in a single crystal of 1,2,4,5-tetramethylbenzene (durene (D)).[3] The final product of this reaction was found to be a pyrazine derivative with two hydrogen atoms attached to the benzene ring of Qx. In the hydrogen abstraction reactions of Qx in solution, the reaction product is the 1,4-dihydro-1-quinoxalinyl radical (QxH) with a hydrogen attached to the nitrogen of Qx.[4-8] Pyrazine derivatives have never been found in the reactions in solution.

In a series of papers, we have recently discussed the photochemical reactions of naphthalene (N) and azanaphthalenes (Qx, quinazoline (Qz), and quinoline (Q)) in durene mixed crystals schematically shown by the scheme in Fig. 1. In all cases, the initial steps (1) of the reactions are the hydrogen abstraction reactions of the triplet states of these molecules and quantum mechanical tunneling plays an essential role in these steps.[9-12] We have discussed the observed temperature, pressure and deuterium isotope effects on the reaction rates in terms of the Siebrand model of hydrogen tunneling.[13] As for the final products, we have hitherto discussed only the case of Qx mentioned above. In the present work, we have made a spectroscopic investigation on the reaction products of Qz, Q, and N in mixed crystals of durene (hereafter these systems are abridged as Qz/D, Q/D, and N/D). The final products of these reactions are found to be quite different depending on the system. The reaction product of Qz/D gives a phosphorescence spectrum characteristic of a benzene derivative, whereas the products of Q/D and N/D give fluorescence spectra characteristic of anthracene derivatives, indicating that dimerization of Q (N) and durene occurs. The difference in the products is rationalized on the basis of the configurations of the guest molecules in the mixed crystals and the difference in the molecular structures.

## Experimental

Q was purified by vacuum distillation. Qz was purified by vacuum sublimation. N was recrystallized from ethanol and

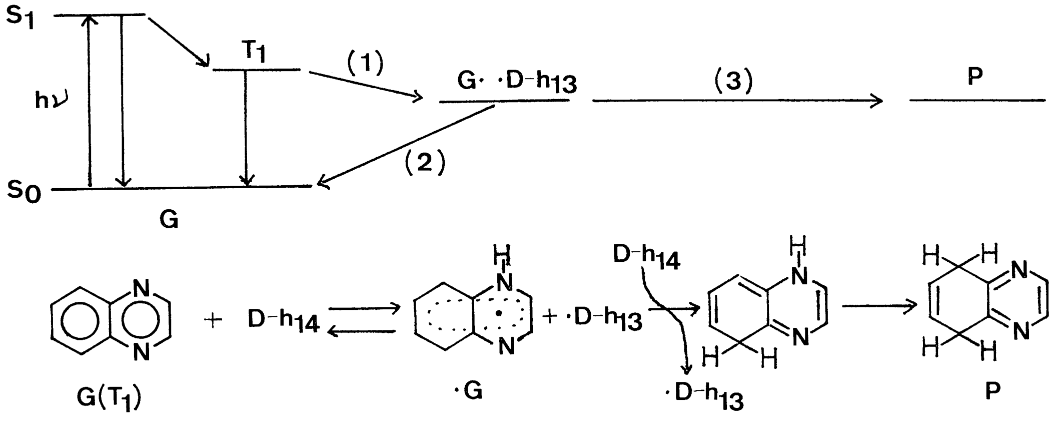

Fig. 1. General reaction scheme of the photochemical reaction of azanaphthalene/durene and naphthalene/durene. Reaction (3) may consist of several steps. G, D-h$_{14}$, D-h$_{13}$, and P represent a guest molecule, durene, 2,4,5-trimethylbenzyl radical, and product, respectively. The reaction process of quinoxaline/durene is also shown below the scheme.

zone refined. Indan was purified by vacuum distillation. Durene was recrystallized from ethanol followed by column chromatography and zone refined. Mixed single crystals were grown from melts containing about 0.1 mol% of guest molecules by the standard Bridgman method.

The UV irradiation was provided by a 900 W Xe lamp whose output was passed through a saturated $NiSO_4$ solution and a Toshiba UV-D33S glass filter. Samples were irradiated at room temperature for about 30 min to induce photochemical reactions. Phosphorescence spectra of the reaction products were obtained at 4.2 K with a Spex Model 1704 1-m monochromator equipped with an EMI 9502B photomultiplier. Phosphorescence lifetimes were measured using a mechanical shutter. Fluorescence spectra were observed at room temperature with a Shimadzu fluorometer (RF-502A).

## Results and Discussion

**Quinazoline/Durene.** When Qz/D is irradiated at room temperature, the green fluorescence of a 2,4,5-trimethylbenzyl radical with a maximum peak at 495 nm appears as the phosphorescence of Qz decreases. The phosphorescence of Qz disappears completely after prolonged (30 min.) irradiation. After irradiation, the ESR signal of a pentamethylcyclopentadienyl radical produced via the 2,4,5-trimethylbenzyl radical was obtained.[14] These experimental results are similar to those found for Qx/D,[3] indicating that the 2,4,5-trimethylbenzyl radical is produced by the photoinduced hydrogen abstraction of Qz from durene.

Figure 2(a) shows the phosphorescence spectrum of "Qz/D" at 4.2 K after irradiation at room temperature. The phosphorescence of Qz (0–0 band: 454.4 nm) is absent, but instead a new spectrum with an 0–0 band at 359.7 nm is obtained. The phosphorescence lifetime is 3.2 s. On the analogy of the result of Qx/D, it may be expected that the final product of the photochemical reaction of Qz/D is a pyrimidine derivative. The observed phosphorescence spectrum, however, cannot be due to a pyrimidine derivative, because the phosphorescence lifetime of pyrimidine is about 12 ms.[15] The long lifetime (3.2 s) and the location of the 0–0 band of the phosphorescence indicate that the phosphorescence is likely to arise from a benzene derivative.[16] The energy of the 0–0 band of the observed phosphorescence (27798 $cm^{-1}$) is very close to that of durene (28000 $cm^{-1}$)[17] and there are two possibilities for the assignment of the observed spectrum.

(1) The energy of the triplet state of the reaction product is lower than that of durene and the observed phosphorescence is genuinely due to the reaction product.

(2) The energy of the triplet state of the reaction product is higher than that of durene and the observed phosphorescence is due to a durene X-trap.

The second possibility cannot be excluded, if the reaction

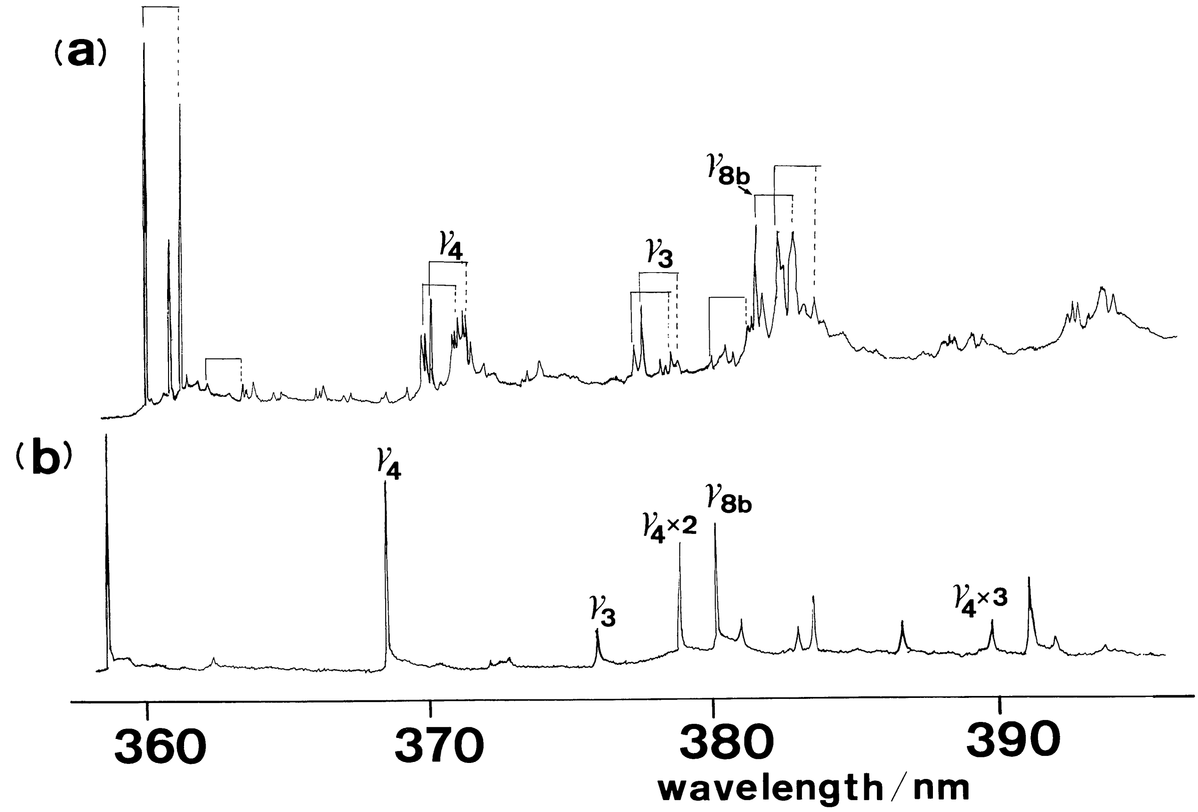

Fig. 2. (a) Phosphorescence spectrum of quinazoline/durene at 4.2 K after irradiation at room temperature. (b) Phosphorescence spectrum of durene X-trap obtained by doping indan in a durene single crystal at 4.2 K.

product is a pyrimidine derivative. This is because the 0–0 energy of $^3(n, \pi^*)$ pyrimidine (28112 cm$^{-1}$ in *p*-xylene and 28240 cm$^{-1}$ in methylcyclohexane[18]) is slightly higher than that of the lowest triplet state of durene.

In order to assign the observed phosphorescence, we have studied the phosphorescence spectrum of a durene X-trap obtained by doping indan in a durene single crystal (Fig. 2(b)). The vibrational analysis and the assignment of the vibronic bands of the phosphorescence are shown in Table 1(a) and (b). The assignment was made on the basis of the Raman data of durene.[19] The energy of the 0–0 band of the durene X-trap is 90 cm$^{-1}$ higher than that of Qz/D after irradiation. Vibronic bands characteristic of the benzene ring can be found in the durene X-trap phosphorescence. The vibronic bands at 760, 1290, and 1582 cm$^{-1}$ are assigned as $\nu_4$, $\nu_3$, and $\nu_{8b}$ modes of the benzene ring,[19,20] respectively. These vibrations are the C–C–C out-of-plane bending, the C–H in-plane bending, and the C–C stretching modes, respectively. As for Qz/D after irradiation, vibronic bands from two different sites are clearly observed with a splitting of 91 cm$^{-1}$. The vibronic bands characteristic of the benzene ring ($\nu_4$, $\nu_3$, and $\nu_{8b}$) are also observed, indicating that the phosphorescence comes from a benzene derivative. There are, however, significant differences between the Qz/D spectrum and the X-trap spectrum. First, in the Qz/D spectrum, there are some bands that are not observed in the durene X-trap, for example, the vibronic bands at 741, 839, 1269 cm$^{-1}$ etc. The band at 741 cm$^{-1}$ may be assigned to the C–H bending mode of an aliphatic skeleton as observed in 1,4-dihydronaphthalene.[21] Second, some overtone (754×2, 754×3) and combination bands in the durene X-trap are not observed in Qz/D. These differences seem to indicate that the phosphorescence of Qz/D after irradiation comes from the reaction product, and not a durene X-trap.

Table 1(a). Vibrational Analysis of the Phosphorescence Spectrum of Quinazoline/Durene at 4.2 K after Irradiation
(As for the vibronic bands from different sites, only those with the lower energy are shown. Weak bands are not shown in the table.)

| $\lambda$/Å | $\nu$/cm$^{-1}$ | $\Delta\nu$/cm$^{-1}$ | Intensity | Assignment |
|---|---|---|---|---|
| 3597.3 | 27798 | 0 | v.s. | 0–0 |
| 3605.6 | 27735 | 63 | s. | |
| 3634.9 | 27511 | 287 | m. | $b_{3g}$ |
| 3659.5 | 27326 | 472 | m. | |
| 3694.8 | 27085 | 733 | m. | |
| 3695.9 | 27057 | 741 | s. | C–H bend.<br>(aliphatic) |
| 3698.5 | 27038 | 760 | s. | $\nu_4$ |
| 3706.1 | 26983 | 815 | m. | |
| 3706.6 | 26979 | 819 | m. | |
| 3709.4 | 26959 | 839 | s. | |
| 3712.6 | 26935 | 863 | m. | |
| 3769.5 | 26529 | 1269 | m. | |
| 3772.5 | 26508 | 1290 | s. | $\nu_3$ |
| 3814.4 | 26216 | 1582 | s. | $\nu_{8b}$ |
| 3815.5 | 26209 | 1589 | m. | |
| 3821.5 | 26168 | 1630 | s. | $b_{2g}$ |
| 3823.0 | 26157 | 1641 | s. | |

Table 1(b). Vibrational Analysis of the Phosphorescence Spectrum of Durene X-Trap at 4.2 K

| $\lambda$/Å | $\nu$/cm$^{-1}$ | $\Delta\nu$/cm$^{-1}$ | Intensity | Assignment |
|---|---|---|---|---|
| 3586.3 | 27884 | 0 | v.s. | 0–0 |
| 3602.9 | 27755 | 128 | w. | |
| 3604.8 | 27741 | 143 | w. | |
| 3623.3 | 27599 | 285 | w. | $b_{3g}$ |
| 3685.9 | 27130 | 754 | v.s. | $\nu_4$ |
| 3701.8 | 27014 | 870 | w. | |
| 3703.6 | 27001 | 883 | w. | |
| 3721.0 | 26874 | 1010 | w. | |
| 3724.0 | 26853 | 1031 | w. | $\nu_{18a}$ |
| 3725.1 | 26845 | 1039 | w. | |
| 3727.4 | 26828 | 1056 | w. | |
| 3759.3 | 26601 | 1283 | m. | $\nu_3$ |
| 3789.3 | 26390 | 1494 | s. | 754×2 |
| 3801.5 | 26305 | 1579 | s. | $\nu_{8b}$ |
| 3809.8 | 26248 | 1636 | m. | $b_{2g}$ |
| 3826.5 | 26134 | 1750 | w. | |
| 3830.0 | 26110 | 1774 | m. | 754+1031 |
| 3833.4 | 26087 | 1797 | w. | |
| 3835.3 | 26074 | 1810 | s. | 754+1056 |
| 3867.2 | 25859 | 2025 | m. | 754+1283 |
| 3898.5 | 25651 | 2233 | m. | 754×3 |
| 3911.9 | 25563 | 2321 | s. | 754+1579 |

This product comes most likely from Qz for the following reason. The reaction products originating from durene are considered to be 2,4,5-trimethylbenzyl and pentamethylcyclopentadienyl radicals. These radicals give, however, no phosphorescence. Furthermore, in the case of Qx/D and Q/D, no phosphorescence similar to that found for the reaction product of Qz/D is observed, though durene is used as the host in all cases. Moreover, durene neat crystal does not give such phosphorescence after irradiation for about 1 h. Intermediate 1,3-dihydro-1-quinazolinyl and 2,4,5-trimethylbenzyl radicals may combine to produce a 3,4-dihydro-3-(2,4,5-trimethylbenzyl)quinazoline like product. The 0–0 band of the phosphorescence spectrum of such a product, however, should be much lower than that of the observed one because of the conjugation of a double bond with the benzene ring as discussed in Reaction Mechanism.

**Quinoline/Durene.** Different from the cases of Qx and Qz, no new phosphorescence appears after irradiation at room temperature when observed at 4.2 K. However, very intense blue fluorescence starts to grow gradually as the green phosphorescence of Q diminishes upon irradiation. Figure 3 shows the fluorescence spectrum of the reaction product of Q/D after irradiation for about 30 min at room temperature. The emission spectrum (obtained by irradiation at 285 nm) starts to rise at 361 nm accompanied by two vibronic bands at 418 and 442 nm and a shoulder at 397 nm. The wavelength range of the emission and the interval of the vibronic

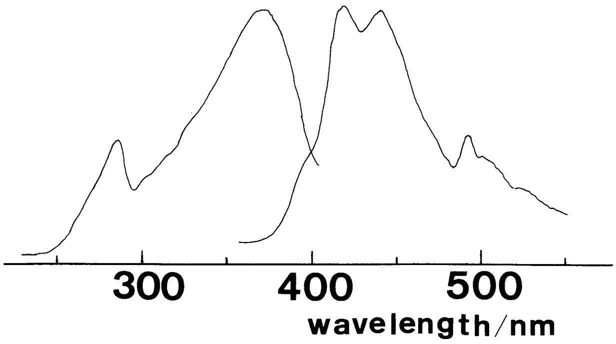

Fig. 3. Fluorescence emission (irradiated at 285 nm) and excitation (monitored at 418 nm) spectra of quinoline/durene at room temperature after irradiation.

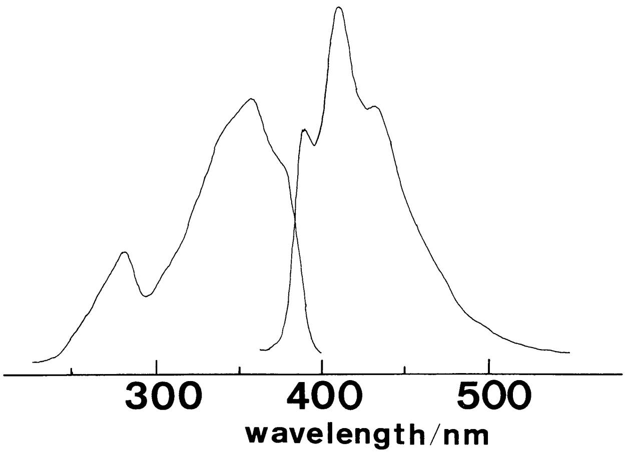

Fig. 4. Fluorescence emission (irradiated at 285 nm) and excitation (monitored at 410 nm) spectra of naphthalene/durene at room temperature after irradiation.

bands (1300 cm$^{-1}$) are similar to those of anthracene,[22] and the excitation spectrum monitored at 418 nm is also similar to the absorption spectrum of anthracene.[23] Because of the broad nature of the spectrum, we cannot ascertain that this fluorescence arises from only one product. But the above mentioned features of the spectrum seems to indicate that the main fluorescent product is an anthracene derivative.

The fluorescence peaks observed lower than 490 nm are due to the 2,4,5-trimethylbenzyl radical which is produced by the hydrogen abstraction reaction.

**Naphthalene/Durene.** Intense blue fluorescence increases during the UV irradiation at room temperature as Q/D. Figure 4 shows the fluorescence emission and excitation spectra of the reaction product after irradiation. The emission (irradiated at 285 nm) starts at about 370 nm with three vibronic bands at 389, 410, and 433 nm. The wavelength range of the fluorescence and the interval of the vibronic bands (1300 cm$^{-1}$) are similar to those of anthracene.[22] The excitation spectrum with a peak at 361 nm (emission at 410 nm) is also similar to the absorption spectrum of anthracene.[23] Therefore, the reaction product of N/D is also considered to be one of the anthracene derivatives as in the case of Q/D.

Neither the fluorescence spectrum of the 2,4,5-trimethylbenzyl radical nor the ESR spectrum of the pentamethylcyclopentadienyl radical was observed. This is probably because the back reaction in the initial hydrogen abstraction process ((2) in Fig. 1) is so efficient that the steady state concentrations of the intermediate 2,4,5-trimethylbenzyl and 1,4-dihydro-1-naphthyl radicals become too low to be detected by ESR.[10] The fluorescence of 2,4,5-trimethylbenzyl radical seems to be much weaker than that of anthracene derivative because of its low steady state concentration and quantum yield.

**Reaction Mechanisms.** There are three questions about the experimental results given above.

(1) What are the exact structures of the reaction products ?

(2) Why is Qz transformed into a benzene derivative, not a pyrimidine derivative ?

(3) Why are Q and N transformed into anthracene derivatives, but not Qx and Qz ?

We discuss these questions on the basis of the configurations of the guest molecules in the mixed crystals and the molecular structures of the guest molecules.

Figure 5 shows the proposed reaction mechanism of Qz/D. In the case of Qz/D, according to the potential energy calculation by McCool et al., there are two sites for Qz in durene, one rotated by 18° and the other by −25° in the molecular plane with respect to the molecular axis of durene.[24] Figure 6 shows the configuration of Qz at the −25° site in durene as a representative. The following discussion can also be applied to Qz at the +18° site. The hydrogen atom from durene is expected to attack the nitrogen of Qz via tunneling in the initial process of the reaction as reported before.[11] The molecular center of Qz deviates by 0.9 Å from that of durene.[24] The durene nearest to the nitrogen at position 1 of Qz is located along the *c* axis of the lattice (2 in Fig. 6). On the other hand, the durene nearest to the nitrogen at position 3 of Qz is located in the *ab* plane of the lattice (3 in Fig. 6). When a reaction proceeds via tunneling, a reaction rate constant depends sensitively on the tunneling distance (the distance between the nitrogen of Qz and the abstracted hydrogen of durene).[13] For example, a decrease of the tunneling distance by 0.1 Å is known to enhance the reaction rate constant by one or two order of magnitude. Because the tunneling distance for the nitrogen at position 3 of Qz is smaller (0.2 Å for the +18° site and 1.5 Å for the −25° site) than that at position 1, the hydrogen atom from durene is predicted to attack position 3 of Qz preferentially to produce a 1,3-dihydro-1-quinazolinyl radical (QzH). Therefore the hydrogen abstraction at position 1 of Qz is negligible to that at position 3 in the initial process of the reaction.

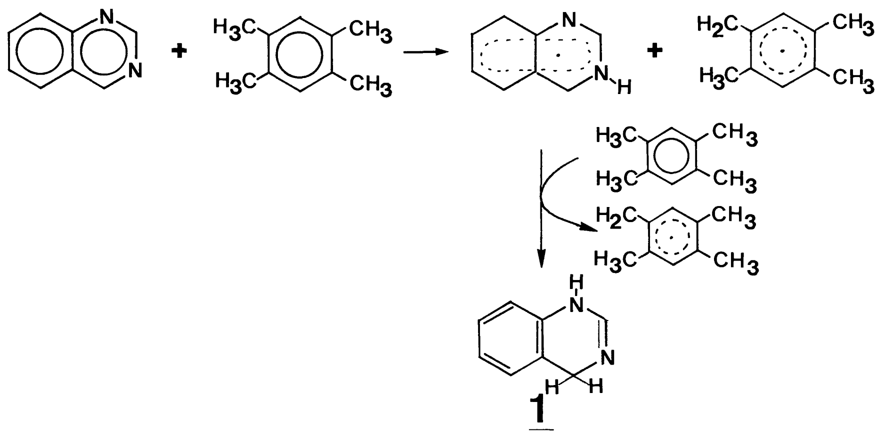

Fig. 5. Proposed reaction mechanism of quinazoline/durene.

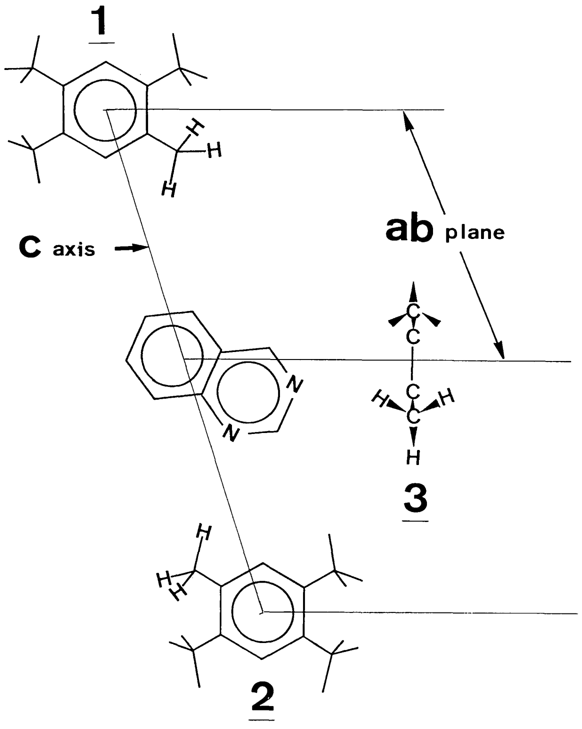

Fig. 6. Configuration of −25° site of quinazoline in durene mixed crystal. Molecules along the *c* axis are not in the same plane in reality.

Hydrogen abstraction via tunneling at the benzene ring of Qz is also negligible to that at the pyrimidine ring because the tunneling distance to the benzene ring is much larger than that to the pyrimidine ring because of the deviation of the molecular center of Qz from that of durene. In order to produce a stable benzene derivative, one more hydrogen must attack the pyrimidine ring of Qz. There are three possible benzene derivatives shown in Fig. 7, but neither product **2** and **3** can be the final product. Because of the conjugation of the double bond with the benzene ring, the triplet energies of products **2** and **3** are expected to be as low as that of 1H-indazole (23590 cm$^{-1}$)[25]. On the other hand, product **1** does not have the conjugaton of the double bond with the benzene ring, producing a triplet energy as high as that of aniline (27659 cm$^{-1}$).[26] Therefore, we consider product **1** as the final product. Though the vibronic bands corresponding to the N–H bond are not found, the existence of the C–H bending mode of an aliphatic skeleton in the phosphorescence spectrum supports this assignment.

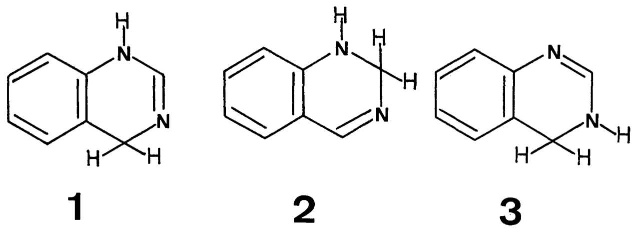

Fig. 7. Possible benzene derivatives produced from quinazoline.

A possible way to explain how the final benzene derivative is produced is the following. The hydrogen of durene along the *c* axis attacks the nitrogen atom at position 1 of QzH, accompanying the transfer of the hydrogen from position 3 to position 4 simultaneously.

Though there is no direct experimental evidence that a pyrimidine derivative is not produced, we consider that the attack of the hydrogen on position 3 of Qz and the deviation of the molecular center of Qz from that of durene inhibit the formation of a pyrimidine derivative. In the case of Qx/D, the molecular axes of Qx is the same as those of durene and the deviation of the molecular center is considered to be small compared to that of Qz. In this system, first, the hydrogen of durene along the *c* axis attacks position 1 of Qx whose tunneling distance is the smallest, producing a 1,4-dihydro-4-quinoxalinyl radical (QxH). Then, the hydrogen attacks position 5 (benzene ring of QxH) which is equivalent to position 1. Finally, the hydrogen attached to position 1 is transferred

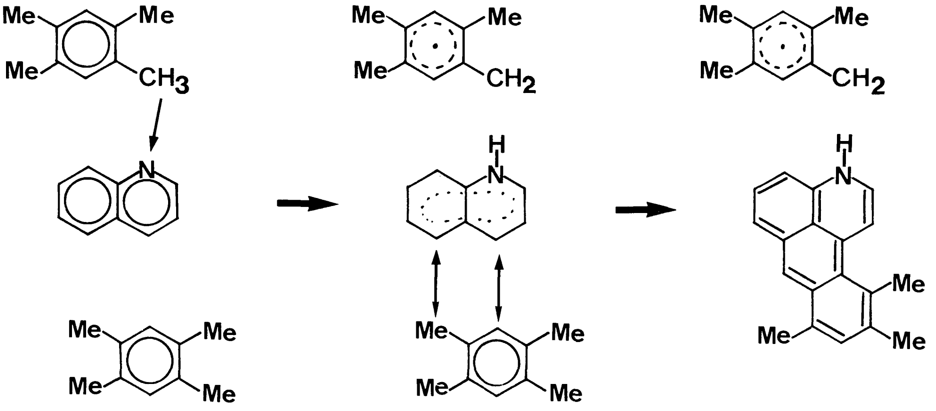

Fig. 8. Probable reaction mechanism of quinoline/durene. The orientation of quinoline and durene in the figure is almost the same as that in durene mixed crystal.

to the neighboring position 8 to produce the pyrazine derivative. The crucial process in proceeding the pyrazine derivative is the attack of the hydrogen of durene on the benzene ring of Qx (position 5). In the case of Qz/D where the hydrogen is attached at position 3, the second hydrogen cannot attack position 5 or 8 because the attachment to these positions destroys the conjugation of the system. The positions in the benzene ring of QzH on which the hydrogen can attack are 6 and 7. The distances from durene to these positions are, however, larger than that of position 5 or 8 on which the hydrogen attacks in Qx/D. Moreover, because the molecular center of Qz deviates by 0.9 Å from that of the replaced durene, the distance between the benzene ring of Qz and durene becomes larger than that for Qx. Therefore, the second hydrogen is expected to attack on the second nearest nitrogen at position 1 of Qz to produce the benzene derivative.

Figure 8 shows a possible reaction mechanism of Q/D. On the analogy of the reaction in solutions[27] and on the basis of the crystal structure,[28] the hydrogen from durene attacks position 1 of Q to produce a 1,4-dihydro-4-quinolinyl radical. After the hydrogen transfers from durene to Q, the 1,4-dihydro-4-quinolinyl radical and durene located at the other position along the *c* axis may dimerize to produce a benzo[*de*]anthracene like derivative. We cannot discuss this dimerization process in detail because the orientation of the intermediate radical is not known. When the orientation of the radical is the same as that of Q, the location of Q and durene along the *c* axis becomes favorable for the dimerization, because Q and durene need not change their orientation greatly to dimerize. However, five hydrogen atoms are released after dimerization. These hydrogen atoms may attach to the neighboring durene molecules to produce pentamethylcyclopentadienyl radicals. On the basis of the fact that the orientation of N is almost the same as that of Q,[29] the photochemical reaction of N/D is also expected to proceed in the same way.

The reason why Qx and Qz do not produce anthracene derivatives may be found in the fact that these molecules have two nitrogen atoms. If such a reaction should occur, durene would attack the nitrogen at position 4 of QxH to form a dimerized product. QxH and durene, however, cannot dimerize because a conjugated system including the nitrogen cannot be formed. In the case of Qz/D, there can be two ways to produce a dimerized product in view of the configuration in the mixed crystal (Fig. 6.) and the attachment of the hydrogen at position 3 of QzH.

(a) Durene 2 attacks the nitrogen at position 1 of QzH.

(b) Durene 1 attacks the carbon at position 4 of QzH. In the case of (a), the dimerization of QzH and durene 2 does not occur for the same reason as given for Qx. In the case of (b), because of the deviation of the molecular center and the rotation in the molecular plane, the distance between durene 1 and position 4 of QzH becomes much larger than that between durene 2 and position 1 of QzH. Therefore the hydrogen abstraction from durene 2 to QzH is considered to occur more efficiently than the dimerization of QzH with durene 1.

In summary, it is found that the final products of the photochemical reactions of Qx/D, Qz/D, Q/D, and N/D are quite different depending on the system despite the fact that the initial steps of the reactions are similar. This difference can be rationalized on the basis of the difference in the molecular structures and the configuration of the guest molecules in the mixed crystals.

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