the frequency of the twisting motion of the C–Ċ bond, respectively. Since the twisting motion is slower in longer alkanes, the sharpening effect is less marked for the longer acyclic alkanes. However, even for longer alkanes, the spectral sharpening may be observable by increasing the temperature for the ESE measurement.

Fluctuation of the local magnetic field also causes the transverse relaxation. Although the fluctuation of the magnetic field within the spectral region of A spins does not cause the longitudinal relaxation, it does cause the transverse relaxation. The transverse relaxation is therefore much faster than the longitudinal relaxation. Since the spectral diffusion is the result of the local field fluctuation, the transverse relaxation is slower for the A spins near the peak magnetic field. However the sharpening of t_2-dependent ESR spectra is less marked because the transverse relaxation is efficiently produced by other processes such as the fluctuation of superhyperfine interaction with adjacent molecules.

Finally let us point out the possibility of obtaining a high-resolution solid-state CW ESR spectrum. A CW ESR spectrum is generally broadened upon increasing the microwave power because the relaxation rate is faster between the ESR peaks. Defining the power-dependent CW ESR spectrum as $S(h,w)$, where w is the microwave power, the unrelaxed component of the ESR spectrum is given by

$$S_s(h,w) = S(h,w_0) - (w_0/w)^{1/2}S(h,w) \qquad (15)$$

where w_0 is the microwave power necessary to observe the ESR spectrum with no power-saturation effect. The ESR spectrum of the unrelaxed component is sharper than $S(h,w_0)$, because the intensity of the spectrum near the peaks is enhanced.

Acknowledgment. The author thanks Professor H. Yoshida for many helpful discussions during the course of this work.

Computational Study of 2-Aminopyrimidine, 2-Amino-5-nitropyrimidine, and the Corresponding *S,S*-Dimethyl-*N*-sulfilimines

Peter Politzer,* Gary P. Kirschenheuter,

Department of Chemistry, University of New Orleans, New Orleans, Louisiana 70148

and Richard S. Miller

Mechanics Division, Office of Naval Research, Arlington, Virginia 22217 *(Received: June 5, 1987; In Final Form: October 2, 1987)*

In order to better understand why the formation of sulfilimine intermediates facilitates the oxidation of some aminoazines, we have carried out a computational analysis of 2-aminopyrimidine (II) and *S,S*-dimethyl-*N*-(2-pyrimidinyl)sulfilimine (III). We also investigated a system for which the procedure fails: 2-amino-5-nitropyrimidine (VI) and the corresponding sulfilimine (VII). An ab initio SCF approach (GAUSSIAN 82) was used to compute the optimized structures and electrostatic potentials of these molecules. We found that the most negative regions in the sulfilimine intermediates as well as in the original aminoazines are associated with one or both of the ring nitrogens rather than the exocyclic one that is to be oxidized. An important new feature in the case of III is the development of extensive negative potentials above and below the ring plane, which make the exocyclic nitrogen much more accessible to electrophiles than it is in II. Thus the possibility of oxidation occurring at this site is significantly greater in III than in II. The presence of the electron-withdrawing $-NO_2$ in VI prevents the development of extensive, relatively strong negative regions above and below the ring; accordingly there does not occur, in VII, an analogous increase in electrophilic accessibility to the exocyclic nitrogen.

Introduction

The reactions with electrophiles of an exocyclic $-NH_2$ group on an aminoazine are impeded by the tendency of the electrophile to preferentially interact with the ring nitrogen(s).[1] This accordingly represents a significant obstacle to the peroxy acid oxidation of such amines to the corresponding nitroso derivatives, a process that is interpreted as involving an electrophilic interaction.[2]

It was suggested that, by making the exocyclic nitrogen sufficiently negative, this obstacle could be overcome.[3] The approach chosen for testing this hypothesis was based on an initial conversion of the amino group into a sulfilimine:

$$-NH_2 \xrightarrow{(CH_3)_2S} \underset{\text{I}}{-N^- - {}^+S(CH_3)_2}$$

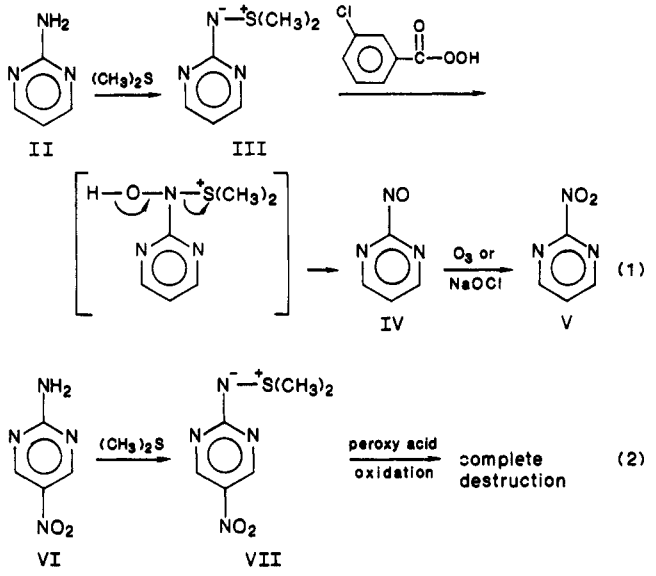
While there has been some controversy as to whether the sulfilimine N–S linkage is best described by a semipolar representation, as in I, or as a double bond,[4] it seems reasonable to anticipate that the negative character of the nitrogen is considerably enhanced relative to the original amine.

It was indeed found that several different aminoazines, upon conversion to sulfilimines, could be oxidized to the nitroso compounds (which are sometimes unstable) and then undergo further transformations, including a second oxidation step to the corresponding nitro derivative.[3] The process, as depicted for 2-aminopyrimidine (II), is shown in reaction 1.[3] 2-Nitropyrimidine (V) was obtained in 33% yield (relative to III), using ozone for the second oxidation.

In the work that is being reported, we have examined in detail the effects of sulfilimine formation upon the exocyclic and the ring nitrogens, so as to achieve a better understanding of how this facilitates the oxidation of the former. We have also investigated

(1) Taylor, E. C. *Principles of Heterocyclic Chemistry*; American Chemical Society: Washington, DC, 1974.
(2) See, for example, Curci, R.; Edwards, J. O. *Organic Peroxides*; Swern, D., Ed.; Wiley-Interscience: New York, 1970; Vol. 1, Chapter IV.
(3) Taylor, E. C.; Tseng, C.-P.; Rampal, J. B. *J. Org. Chem.* **1982**, *47*, 552.
(4) Oae, S.; Furukawa, N. *Sulfilimines and Related Derivatives*; ACS Monograph 179; American Chemical Society: Washington, DC, 1983; Chapter 3.

0022-3654/88/2092-1436$01.50/0



a case in which this procedure does not work;[5] reaction 2, which starts with 2-amino-5-nitropyrimidine (VI), leads to the destruction of the heterocyclic system. Accordingly, a comparison shall be made of the starting molecules and the sulfilimine intermediates in reactions 1 and 2.

Methods

Our approach is a computational one; we use the ab initio self-consistent field molecular orbital GAUSSIAN 82 procedure.[6] For each molecule, we first determined its optimized structure and then used this geometry to calculate the properties of interest. Because of the sizes of III and VII, we carried out the optimizations at the STO-3G* level, which has been found to be effective for predicting geometries.[7] However, since this basis set is known to give poor results for C–NO_2 and N–O bond lengths,[8] we obtained these and the C–N–O angles by a preliminary 6-31G optimization of VI.

Our focus in this study is upon the degrees of negative character and susceptibilities to electrophilic attack of the exocyclic and ring nitrogens. These properties can be analyzed very effectively by means of the electrostatic potentials of the molecules. The potential $V(\vec{r})$ that is created in the space around a molecule by its nuclei and electrons is given rigorously by

$$V(\vec{r}) = \sum_A \frac{Z_A}{|\vec{R}_A - \vec{r}|} - \int \frac{\rho(\vec{r}')\, d\vec{r}'}{|\vec{r}' - \vec{r}|} \qquad (3)$$

Z_A is the nuclear charge of atom A, located at $\vec{R}_A$, and $\rho(\vec{r})$ is the electronic density function of the molecule. Negative values of $V(\vec{r})$ indicate regions in which the effect of the electrons is dominant; it is to these regions, and particularly to the points of most negative potential (the local minima), that an approaching electrophile is initially attracted. The electrostatic potential is well-established as an effective tool for interpreting and predicting molecular reactive behavior.[9,10] An important feature of $V(\vec{r})$ is that it is a real physical property, which can be determined experimentally as well as computationally.[10] In this work, we have calculated $V(\vec{r})$ at the STO-5G level, using our optimized molecular geometries; it has been shown that satisfactory electrostatic potentials are obtained with ab initio SCF procedures using minimum basis sets.[9b,11]

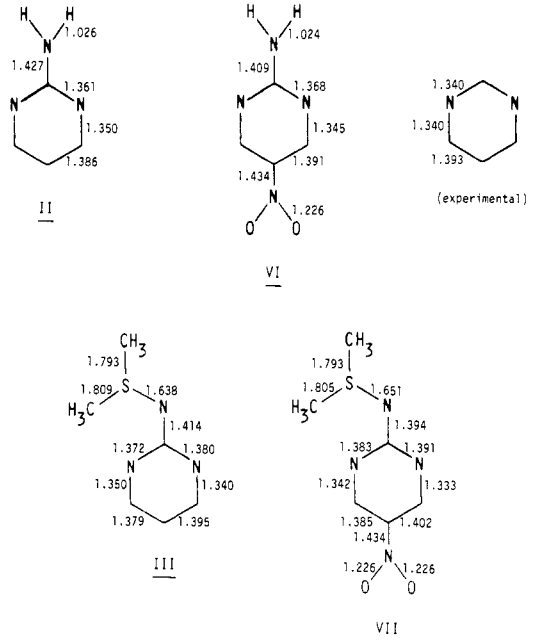


Figure 1. Calculated bond lengths in 2-aminopyrimidine (II), *S,S*-dimethyl-*N*-(2-pyrimidinyl)sulfilimine (III), 2-amino-5-nitropyrimidine (VI), and *S,S*-dimethyl-*N*-(5-nitro-2-pyrimidinyl)sulfilimine (VII). All values are in angstroms.

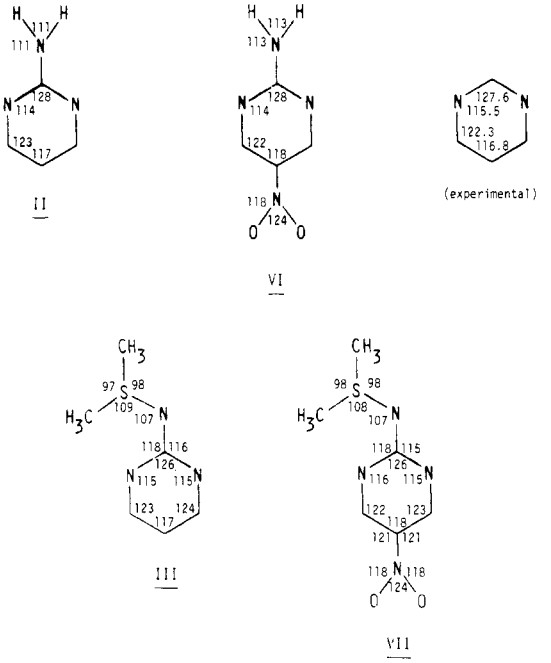


Figure 2. Calculated bond angles in the molecules listed in the caption to Figure 1. All values are in degrees.

Results and Discussion

Structures. Our calculated structures are presented in Figures 1 and 2. For comparison, the experimentally determined geometry of unsubstituted pyrimidine is also included.[12] In general, the bond lengths and bond angles in II and VI are quite similar to

(5) Coburn, M. D., private communication.

(6) Binkley, J. S.; Frisch, M.; Raghavachari, K.; DeFrees, D.; Schlegel, H. B.; Whiteside, R.; Fluder, E.; Seeger, R.; Pople, J. A. GAUSSIAN 82, Release A; Carnegie-Mellon University: Pittsburgh, PA.

(7) Radom, L.; Lathan, W. A.; Hehre, W. J.; Pople, J. A. *J. Am. Chem. Soc.* **1971**, *93*, 5339.

(8) Politzer, P.; Domelsmith, L. N.; Sjoberg, P.; Alster, J. *Chem. Phys. Lett.* **1982**, *92*, 366.

(9) (a) Scrocco, E.; Tomasi, J. *Adv. Quantum Chem.* **1978**, *11*, 116. (b) Politzer, P.; Daiker, K. C. *The Force Concept in Chemistry*; Deb, B. M., Ed.; Van Nostrand-Reinhold: New York, 1981; Chapter 7.

(10) Politzer, P.; Truhlar, D. G., Eds. *Chemical Applications of Atomic and Molecular Electrostatic Potentials*; Plenum: New York, 1981.

(11) (a) Scrocco, E.; Tomasi, J. *Top. Curr. Chem.* **1973**, *42*, 95. (b) Ghio, C.; Tomasi, J. *Theor. Chim. Acta* **1973**, *30*, 151. (c) Almlof, J.; Stogard, A. *Chem. Phys. Lett.* **1974**, *29*, 418.

(12) Katritzky, A. R. *Handbook of Heterocyclic Chemistry*; Pergamon Press: New York, 1985.

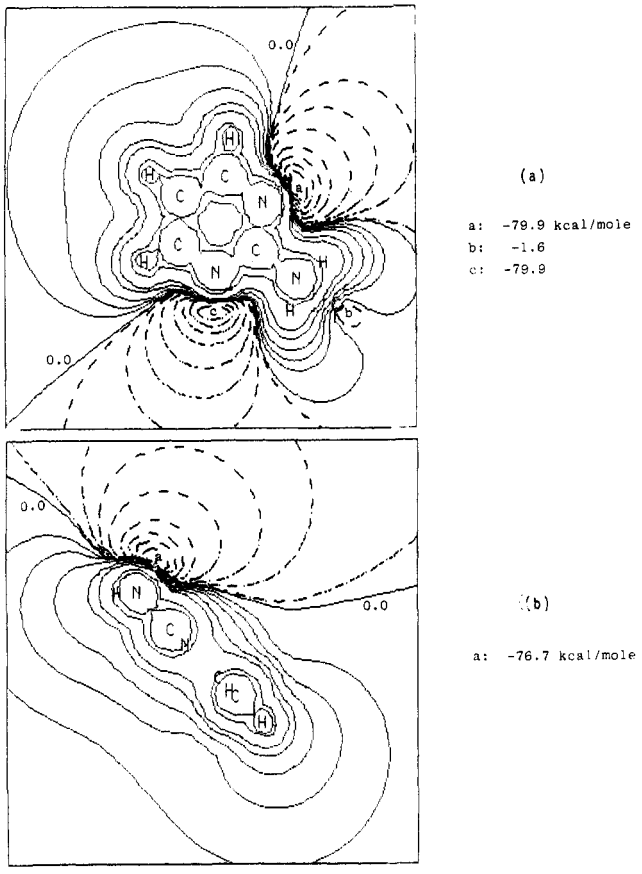


Figure 3. Calculated electrostatic potential in the ring plane of 2-aminopyrimidine (a) and in the perpendicular plane passing through the exocyclic C–N bond (b). Dashed contours correspond to negative potentials; zero contour is indicated. Magnitudes for other contours, in kcal/mol, are −0.6, −3.1, −6.3, −12.6, −25.1, −37.7, −50.2, −62.8, +3.1, +6.3, +12.6, +31.4, +62.8, +313, and +628. The locations of the most negative potentials are indicated, and their values are given at the side of the figure.

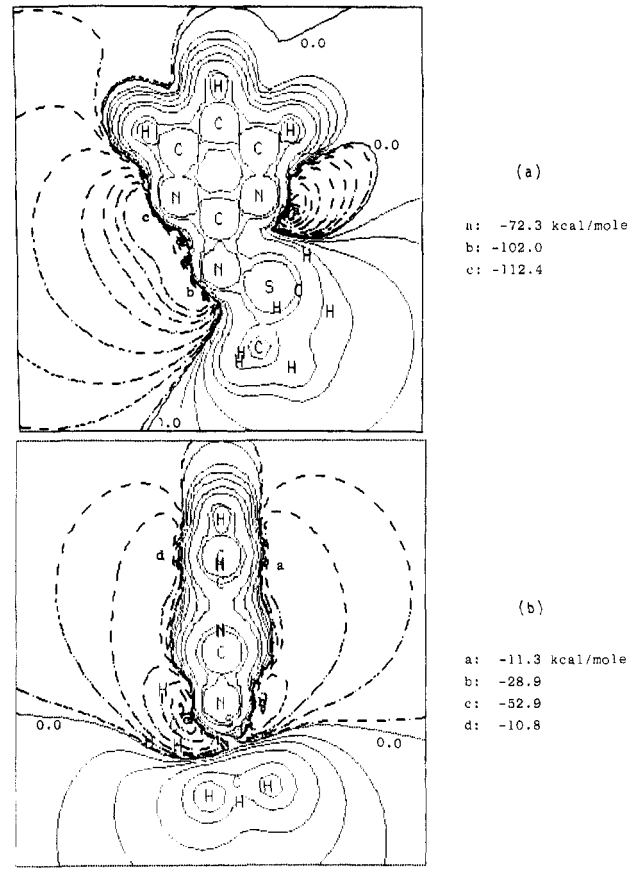


Figure 4. Calculated electrostatic potential in the ring plane of *S,S*-dimethyl-*N*-(2-pyrimidinyl)sulfilimine (a) and in the perpendicular plane passing through the exocyclic C–N bond (b). Dashed contours correspond to negative potentials; zero contour is indicated. Magnitudes for other contours are given in caption to Figure 3. The locations of the most negative potentials are indicated, and their values are given at the side of the figure.

TABLE I: Potential Minima Associated with Systems II, III, VI, and VII[a]

system	values of minima, kcal/mol	site with which minimum is associated
2-aminopyrimidine (II)	−82.5	ring nitrogens
	−79.2	exocyclic nitrogen
S,S-dimethyl-*N*-(2-pyrimidinyl)-sulfilimine (III)	−112.8	ring nitrogen on side away from $-S(CH_3)_2$
	−104.4	exocyclic nitrogen
	−76.2	ring nitrogen on same side as $-S(CH_3)_2$
2-amino-5-nitropyrimidine (VI)	−68.9	ring nitrogens
	−57.2	exocyclic nitrogen (amino group)
	−54.2	oxygens
	−37.3	oxygens
S,S-dimethyl-*N*-(5-nitro-2-pyrimidinyl)sulfilimine (VII)	−99.7	ring nitrogen on side away from $-S(CH_3)_2$
	−89.0	exocyclic nitrogen (amino group)
	−66.7, −67.1	oxygens
	−62.0	ring nitrogen on same side as $-S(CH_3)_2$
	−50.9, −53.0	oxygens

[a] The values given in this table are for the absolute minima in the regions indicated. Thus they will not, in general, be the same as the minima in Figures 3–6, which refer only to the planes shown in the figures.

those given for pyrimidine; some differences are of course to be anticipated due to the effects of the substituents in the former.

In both II and VI, the amino group is calculated to be pyramidal in configuration. It is interesting to note that the presence of the $-NO_2$ in VI is not sufficient to force the $-NH_2$ into a planar configuration, in contrast to what we have found for the nitroanilines, also at the STO-3G level.[13] In the latter the amino group is in the plane of the ring, unlike aniline itself, which the STO-3G basis correctly predicts to have a pyramidal $-NH_2$ conformation.[13] (It has been found, by us and by others as well, that split-valence basis sets, such as the 6-31G, are not reliable for predicting $-NH_2$ conformations.[14]) When the $-NH_2$ is coplanar with the ring, it can act more effectively as an electron donor (through resonance), in response to the strongly electron-withdrawing $-NO_2$. Evidently $-NH_2$ does not conjugate to as great an extent with the pyrimidine ring as with the phenyl, although the relatively short C–NH_2 bond lengths in II and VI reflect some degree of double bond character and indicate that some conjugation does take place. (Typical aliphatic C–N bond lengths are about 1.47 Å; in aniline, however, this distance is 1.402 Å.[13]) These findings are consistent with the decrease in aromatic character that accompanies the replacement of carbons by nitrogens in the benzene ring.[12]

The formation of the sulfilimines III and VII causes a further shortening of these C–N bonds (Figure 1), which suggests some shifting of electronic charge from the $-N^{-}-{}^{+}S(CH_3)_2$ group toward the ring. This inference will be confirmed by the electrostatic potential analysis. Crystallographic structure determinations have been carried out for a number of sulfilimines,[4] all of them un-

(13) Politzer, P.; Abrahmsen, L.; Sjoberg, P. *J. Am. Chem. Soc.* **1984**, *106*, 855.

(14) (a) Pulay, P.; Fogarasi, G.; Pang, F.; Boggs, J. E. *J. Am. Chem. Soc.* **1979**, *101*, 2550. (b) Lane, P., private communication.

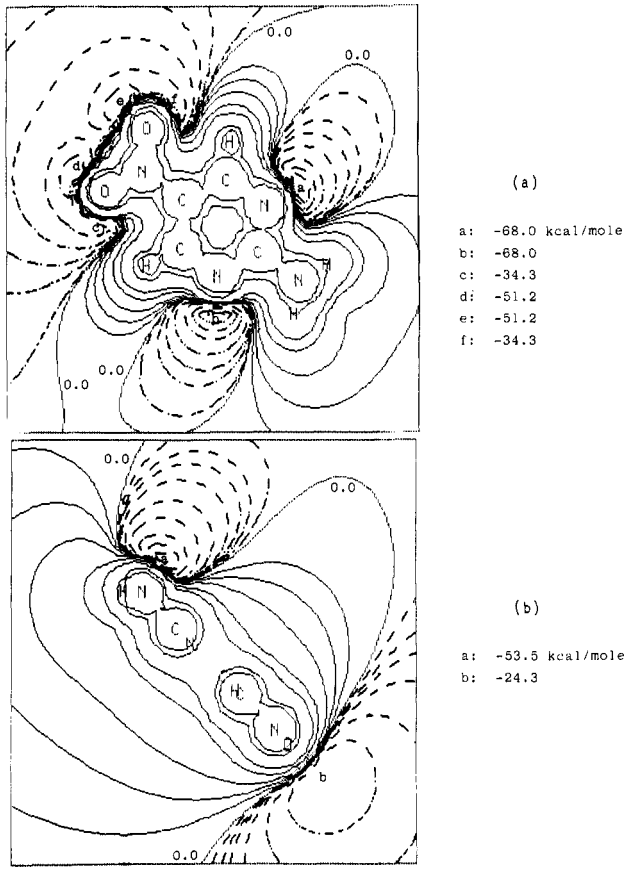


Figure 5. Calculated electrostatic potential in the ring plane of 2-amino-5-nitropyrimidine (a) and in the perpendicular plane passing through the exocyclic C–N bonds (b). Dashed contours correspond to negative potentials; zero contour is indicated. Magnitudes for other contours are given in caption to Figure 3. The locations of the most negative potentials are indicated, and their values are given at the side of the figure.

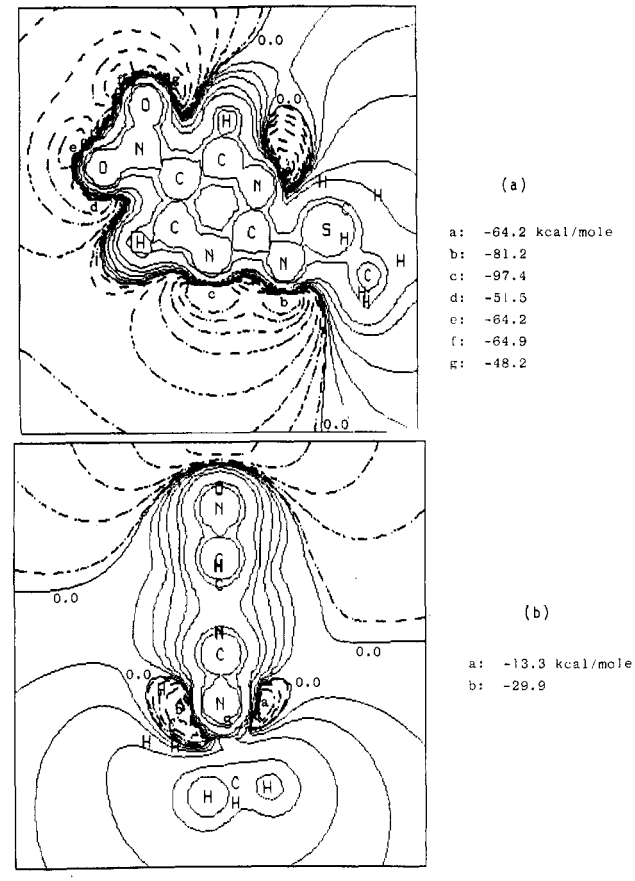


Figure 6. Calculated electrostatic potential in the ring plane of *S,S*-dimethyl-*N*-(5-nitro-2-pyrimidinyl)sulfilimine (a) and in the perpendicular plane passing through the exocyclic C–N bonds (b). Dashed contours correspond to negative potentials; zero contour is indicated. Magnitudes for other contours are given in caption to Figure 3. The locations of the most negative potentials are indicated, and their values are given at the side of the figure.

fortunately quite different in composition from III and VII. However, the S–N distances were almost always found to be within the 1.63–1.68-Å range, with which our calculated values are in excellent agreement.

A final structural point of interest concerns the possibility of intramolecular hydrogen bonding in the sulfilimines III and VII. The methyl hydrogens in $(CH_3)_2S$ are known to be somewhat acidic,[15] and it is conceivable that one of them may form a hydrogen bond to one of the ring nitrogens. Our calculated structures are consistent with a weakly attractive interaction; the shortest separation between a methyl hydrogen and the nearest ring nitrogen is about 2.71 Å in both III and VII, slightly less than the sum of the van der Waals radii of hydrogen and nitrogen (1.20 and 1.56 Å, respectively[16]).

Electrostatic Potentials. The calculated electrostatic potentials of the four systems of present interest (II, III, VI, and VII) are shown in Figures 3–6, and their most negative values (the potential minima) are summarized in Table I. In general, there are relatively strong minima associated with the ring nitrogens, the exocyclic amino or sulfilimino nitrogens, and the oxygens of the nitro groups.

Looking first at 2-aminopyrimidine and its sulfilimine (II and III), which are involved in reaction 1, our results confirm the basic premise that the formation of the latter renders the exocyclic nitrogen more negative; its potential minimum changes from −79.2 kcal/mol in II to −104.4 in III. The ring nitrogens are also affected; the one closest to the $-S(CH_3)_2$ group is made somewhat more positive, presumably reflecting the proximity of the methyl hydrogen, but the other becomes considerably more negative, with a minimum of −112.8 kcal/mol. This remains, therefore, a more attractive site for the initial approach of an electrophile than is the exocyclic nitrogen.

However the most striking consequences of forming the sulfilimine, in the case of 2-aminopyrimidine, are found above and below the plane of the ring (compare Figures 3b and 4b). In 2-aminopyrimidine (II) as in pyrimidine itself,[17] these regions are almost entirely positive, with the only exception being due to the lone pair of the $-NH_2$ group. (The fact that the presence of the $-NH_2$, a strong resonance electron donor, does not produce a negative ring potential in 2-aminopyrimidine is further evidence of a relatively low degree of conjugation.) In the sulfilimine III, on the other hand, there is an extensive negative electrostatic potential above and below the ring. This is a very significant development, which greatly expands the spatial regions in which an electrophile will undergo an attractive interaction with the molecule and should considerably increase the likelihood of the system undergoing electrophilic attack. While such attack is most favored at the ring nitrogen having the strongest negative electrostatic potential, its likelihood at the exocyclic nitrogen should also be much enhanced.

The situation is quite different when the nitro group is present, in VI and VII. The potentials associated with the amino nitrogen and with one of the ring nitrogens are again made more negative by the formation of the sulfilimine, but the magnitudes are now smaller, due to the electron-withdrawing nature of $-NO_2$ (Figures 5 and 6). The amino nitrogen potential changes from −57.2 to

(15) (a) Rothstein, E. *J. Chem. Soc.* **1937**, 309; *J. Chem. Soc.* **1940**, 1558; *J. Chem. Soc.* **1953**, 3991. (b) Woodward, R. B.; Eastman, R. H. *J. Am. Chem. Soc.* **1946**, *68*, 2229. (c) Streitwieser, A., Jr.; Ewing, S. P. *J. Am. Chem. Soc.* **1975**, *97*, 190.

(16) Bondi, A. *J. Phys. Chem.* **1964**, *68*, 441.

(17) Almlof, J.; Johansen, H.; Roos, B.; Wahlgren, U. *J. Electron Spectrosc. Relat. Phenom.* **1973**, *2*, 51.

-89.0 kcal/mol, while the ring nitrogen goes from -68.9 to -99.7 kcal/mol. Most significant, however, is that the sulfilimine VII does not show extended negative regions above and below the ring (Figure 6b). This is a striking difference between the two sulfilimines that are being studied and indicates that III should be considerably more reactive toward electrophiles than is VII.

Summary and Conclusions

The analysis that has been presented shows that the most negative regions in these molecules are associated with one of the ring nitrogens and not with the exocyclic nitrogen that is to be oxidized. This is true in the sulfilmine intermediates as well as in the original aminoazines. An important new feature in the case of sulfilimine III is that the exocyclic nitrogen has become much more accessible to electrophiles, now from both sides of the ring plane as well as in the plane, whereas the only channel of approach in the original azine II was from one side of the ring plane (compare Figures 3 and 4). The increase in accessibility is much greater for the exocyclic nitrogen than for the one in the ring; the latter could be approached from either side of the ring plane even before formation of the sulfilimine, since its lone-pair potential extends above and below the ring plane, unlike that of the exocyclic nitrogen.

Thus, while the most reactive site for initial electrophilic attack is the ring nitrogen, in III as well as in II, the possibility of the reaction taking place at the exocyclic nitrogen is significantly greater in the sulfilimine. Indeed, as mentioned earlier, a 33% yield of 2-nitropyrimidine (V) was obtained from sulfilimine III.[3]

The introduction of the $-NO_2$ group in position 5 prevents the development of extensive, relatively strong negative electrostatic potentials above and below the pyrimidine ring (Figure 6b). Accordingly, there does not occur, in VII, the considerable increase in electrophilic accessibility to the exocyclic sulfilimine nitrogen that is found in III. We suggest that this is at least part of the reason why oxidation of the latter nitrogen in VII is not observed.[5]

Acknowledgment. We greatly appreciate very helpful discussions with Dr. Jane S. Murray and Dr. Michael D. Coburn. We are grateful for the support of this work by the Office of Naval Research.

Registry No. II, 109-12-6; III, 54214-58-3; VI, 3073-77-6; VII, 108733-91-1.

An Analysis of π-Bond Formation Using Hartree–Fock Theory

Hiroshi Ichikawa,* Yukiko Ebisawa, and Atsushi Shigihara

Hoshi College of Pharmacy, Shinagawa, Tokyo 142, Japan *(Received: June 11, 1987)*

A Hartree–Fock MO theoretical method of energy analysis of chemical binding which might be applied to large organic systems is proposed. An application of the method to the π-bond formation in the ethylene system shows that a release of the kinetic energy pressure of 2p valence electrons leads to delocalization between 2p atomic orbitals and, as a consequence, both carbon atoms move closer with a concomitant lowering of the potential energy and the total energy.

Seeking the reason for chemical binding has been a central problem in chemistry. This extends to prediction of reactivity and reaction products in synthetic chemistry. The concept of atomic or molecular orbital interaction has been prevalent in the theories of chemical binding (orbital interaction theories). Examples can be seen in molecular orbital theory,[1] frontier orbital theory,[2,3] charge-transfer theory, etc.[4] However, such theories raise further questions regarding the physical sources of attractive or repulsive interaction between orbitals. Although orbital interaction theories are useful in application, it is fruitful to proceed beyond this level and to look for explanations in terms of fundamental physical quantities.

Chemical phenomena always involve energetic changes in a given system. The basic energies which govern chemical phenomena are electrostatic potentials (between electrons, between electrons and nuclei, and between nuclei) and kinetic energies of nuclei and electrons. Since the Born–Oppenheimer approximation[5] holds in chemical processes (unless it is applied to transition states), the kinetic energy of nuclear motion may be considered separately. Therefore, major and important energies in molecule are the potential energy and the kinetic energy of electrons.

Ruedenberg and his co-workers have performed a quantum mechanical analysis on the formation of the covalent bond in the hydrogen molecule ion.[6-8] They came to the conclusion that electron sharing leads to chemical binding as the result of a subtle interplay between the uncertainty principle and the nuclear attractions: "Delocalization of the valence electrons from one atom to other atoms leads to a lowering of the kinetic energy pressure and, as a consequence, there results a firmer attachment of these electrons to the nuclei with a concomitant lowering of the potential as well as the total energy".

We consider their method of approach to be the most fundamental one among those so far proposed since it clarifies the roles of the potential energy and the kinetic energy of electrons in chemical binding. We also believe this kind of analysis to be effective for the fundamental interpretation of chemical phenomena. However, it does not seem to be widely known among organic chemists. This may be because it is difficult to apply the same method or similar methods[9] to large systems such as those

(1) Early history of molecular orbital theory is summarized in: Parr, R. G. *Quantum Theory of Molecular Electronic Structure*; Benjamin: New York, 1963; Slater, J. C. *Quantum Theory of Molecules and Crystals*; McGraw-Hill: New York, 1963; Vol. 1.

(2) (a) Fukui, K.; Yonezawa, T.; Shingu, H. *J. Chem. Phys.* **1952**, *20*, 722. (b) Fukui, K.; Yonezawa, T.; Nagata, C.; Shingu, H. *J. Chem. Phys.* **1954**, *22*, 1433. (c) Fukui, K.; Yonezawa, T.; Nagata, C. *Bull. Chem. Soc. Jpn.* **1954**, *27*, 423.

(3) (a) Woodward, R. B.; Hoffmann, R. *J. Am. Chem. Soc.* **1965**, *87*, 395. (b) Longuet-Higgins, H. C.; Abrahamson, E. W. *J. Am. Chem. Soc.* **1965**, *87*, 2045.

(4) (a) Mulliken, R. S. *J. Am. Chem. Soc.* **1952**, *74*, 811. (b) Mulliken, R. S. *J. Chem. Phys.* **1951**, *19*, 514. (c) Briegelb, C. *Elektronen-Donator-Acceptor-Komplexe*; Springer-Verlag: Berlin, 1961.

(5) Born, M.; Oppenheimer, J. R. *Ann. Phys.* **1927**, *84*, 457.

(6) Ruedenberg, K. *Rev. Mod. Phys.* **1962**, *34*, 326.

(7) Feinberg, M. J.; Ruedenberg, K. *J. Chem. Phys.* **1971**, *54*, 1495.

(8) Feinberg, M. J.; Ruedenberg, K.; Mehler, E. L. *Adv. Quantum Chem.* **1970**, *5*, 27.

(9) (a) Wilson, Jr., C. W.; Goddard III, W. A. *Theor. Chim. Acta* **1972**, *26*, 195. (b) Goddard III, W. A.; Wilson, Jr., C. W. *Theor. Chim. Acta* **1972**, *26*, 211.

0022-3654/88/2092-1440$01.50/0