atmosphere. The solution was stirred for 30 min at room temperature and was closely monitored by thin layer chromatography. The solvent was removed under vacuum and the residue was dissolved in dichloromethane. The solution was filtered through Celite to remove the insoluble material and the solution was subjected to chromatographic workup using silica gel thin layer chromatography plates. Elution with n-hexane yielded a single brown band which contained **3** (30 mg, 25%) or **4** (46 mg, 32%).

Compound **3**: IR (n-hexane): $\tilde{\nu}$(CO) = 2083(s), 2050(vs), 2014(vs), 1999(m), 1919(m) cm^{-1}; EI-MS (70 eV, 180 °C): m/z (%): 579.7 (6) [M^+], 523.7 (24) [$M^+ - 2$CO], 439.9 (26) [$M^+ - 5$CO], 411.6 (9) [$M^+ - 6$CO], 383.7 (22) [$M^+ - 7$CO], 355.7 (100) [$M^+ - 8$CO], 292.7 (52) [$M^+ - 8$CO − PS], 227.7 (25) [$M^+ - 8$CO − PS − Cp]; ^{1}H NMR (250.113 MHz, 298 K, CDCl$_3$, TMS): $\delta = 4.90$ (d, ^3J(P,H) = 1.8 Hz, 5H); ^{31}P NMR (101.256 MHz, 298 K, CDCl$_3$, 85% H$_3$PO$_4$ ext.): $\delta = 260.7$; m.p. 136–137 °C; elemental analysis (%) calcd for $C_{13}H_5CrFe_2O_8PS_3$: C 26.9, H 0.86; found: C 26.2, H 1.3.

Compound **4**: IR (n-hexane): $\tilde{\nu}$(CO) = 2075(s), 2042(vs), 2006(vs), 1991(m), 1985(s), 1920(m) cm^{-1}; EI-MS (70 eV, 180 °C): m/z (%): 721.8 (7) [M^+], 665.8 (20) [$M^+ - 2$CO], 581.7 (21) [$M^+ - 5$CO], 497.8 (100) [$M^+ - 8$CO], 432.7 (12) [$M^+ - 8$CO − Cp], 321.8 (34) [$M^+ - 2$Fe − 8CO − Cp]; ^{1}H NMR (250.113 MHz, 298 K, CDCl$_3$, TMS): $\delta = 4.88$ (d, ^3J(P,H) = 2.01 Hz, 5H); ^{31}P NMR (101.256 MHz, 298 K, CDCl$_3$, H$_3$PO$_4$ ext.): $\delta = 235.1$ (s, J(P,Se) = 95.9 and 269 Hz); m.p. 142–143 °C; elemental analysis (%) calcd for $C_{13}H_5CrFe_2O_8PSe_3$: C 21.6, H 0.69; found: C 21.5, H 1.15.

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[1] For reviews, see: a) J. Wachter, *Angew. Chem.* **1998**, *110*, 782–800; *Angew. Chem. Int. Ed.* **1998**, *37*, 750–768; b) G. W. Drake, J. W. Kolis, *Coord. Chem. Rev.* **1994**, *137*, 131–178; c) A.-J. DiMaio, A. L. Rheingold, *Chem. Rev.* **1990**, *90*, 169–190; d) M. Di Vaira, P. Stoppioni, *Coord. Chem. Rev.* **1992**, *120*, 259–279.

[2] a) F. Weigend, S. Wirth, R. Ahlrichs, D. Fenske, *Chem. Eur. J.* **2000**, *6*, 545–551; b) S. Wirth, D. Fenske, *Z. Anorg. Allg. Chem.* **1999**, *625*, 2064–2070.

[3] G. A. Zank, T. B. Rauchfuss, S. R. Wilson, A. L. Rheingold, *J. Am. Chem. Soc.* **1984**, *106*, 7621–7623. In some polymeric solid state phases, the unit $(EY_3)^{3-}$ has been observed (see ref. [1b]], and references therein).

[4] Crystal structure analysis of **3** and **4**: STOE IPDS area-detector diffractometer with $Ag_{K\alpha}$ radiation ($\lambda = 0.56087$ Å) for **3** and $Mo_{K\alpha}$ radiation ($\lambda = 0.71073$ Å) for **4**. The structures were solved by direct methods using SHELXS-86,[13a] and refined by full matrix least squares on F^2 using SHELXL-93,[13b] with anisotropic displacement for non-H atoms. H atoms were placed in idealized positions and refined isotropically using a riding model. **3**: $C_{26}H_{10}Cr_2Fe_4O_{16}P_2S_6$, $M_r = 1160.04$, crystal size $0.40 \times 0.20 \times 0.03$ mm, monoclinic, space group $P2_1/n$, $a = 7.5570(15)$, $b = 21.477(4)$, $c = 24.213(5)$ Å, $\beta = 96.11(3)°$, $V = 3907.5(13)$ Å^3, $T = 213(2)$ K, $Z = 4$, $\rho_{calcd} = 1.972$ Mg m^{-3}, $\mu(Ag_{K\alpha}) = 12.66$ cm^{-1}; 14890 independent reflections ($2\theta_{max} = 52°$, $R_{int} = 0.0669$), 505 parameters, $wR_2 = 0.1400$; R_1 (10513 with $F_o = 4\sigma(F_o)) = 0.0483$. **4**: $C_{26}H_{10}Cr_2Fe_4O_{16}P_2Se_6$, $M_r = 1441.44$, crystal size $0.30 \times 0.15 \times 0.01$ mm, monoclinic, space group $P2_1/n$, $a = 7.5355(2)$, $b = 21.868(2)$, $c = 24.809(3)$ Å, $\beta = 96.16(3)°$, $V = 4172.4(10)$ Å^3, $T = 293(2)$ K, $Z = 4$, $\rho_{calcd} = 2.295$ Mg m^{-3}, $\mu(Mo_{K\alpha}) = 72.31$ cm^{-1}; 8095 independent reflections ($2\theta_{max} = 52°$, $R_{int} = 0.0816$), 505 parameters, $wR_2 = 0.1164$; R_1 (5107 with $F_o = 4\sigma(F_o)) = 0.0480$. Crystallographic data (excluding structure factors) for the structures reported in this paper have been deposited with the Cambridge Crystallographic Data Centre as supplementary publication nos. CCDC-148123 and CCDC-146124. Copies of the data can be obtained free of charge on application to CCDC, 12 Union Road, Cambridge CB21EZ, UK (fax: (+44) 1223-336-033; e-mail: deposit@ccdc.cam.ac.uk).

[5] S. van Houten, E. H. Wiebenga, *Acta. Crystallogr.* **1957**, *10*, 156–160.

[6] O. J. Scherer, G. Kemeny, G. Wolmershäuser, *Chem. Ber.* **1995**, *128*, 1145–1148.

[7] G. J. Penney, G. M. Sheldrick, *J. Chem. Soc. A* **1971**, 245–248.

[8] P. Mathur, S. Ghose, M. M. Hossain, C. V. V. Satyanarayana, S. Banerjee, G. R. Kumar, P. B. Hitchcock, J. F. Nixon, *Organometallics* **1997**, *16*, 3815–3818.

[9] L. Y. Goh, W. Chen, R. C. S. Wong, K. Karaghiosoff, *Organometallics* **1995**, *14*, 3886–3896.

[10] L. Y. Goh, W. Chen, R. C. S. Wong, *Organometallics* **1999**, *18*, 306–314.

[11] W. Hieber, J. Gruber, *Z. Anorg. Allg. Chem.* **1958**, *296*, 91–103.

[12] L. Y. Goh, C. K. Chu, R. C. S. Wong, T. W. Hambley, *J. Chem. Soc. Dalton Trans.* **1989**, 1951–1956.

[13] a) G. M. Sheldrick, SHELXS-86, Universität Göttingen, **1986**; b) G. M. Sheldrick, SHELXL-97, Universität Göttingen, **1997**.

About the Chemistry of Phosphorus Suboxides[**]

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Sigrid D. Peyerimhoff

While the molecular phosphorus subsulfides P_4S_3, P_4S_4, and P_4S_5 are well known,[1] various attempts to prepare the corresponding molecular phosphorus suboxides have failed so far. In its polymeric form phosphorus suboxide "P_4O" was already mentioned in 1832[2] but in molecular form P_4O has been detected only in rare gas matrices.[3] Using IR spectroscopy combined with ab initio calculations[4] it was possible to identify the geometries of three different isomers of P_4O. The energetically most favorable isomer of P_4O possesses a planar five-membered ring (isomer **I** in Table 1), while a terminal (isomer **III** in Table 1) and a bridged-bonded isomer (isomer **II** in Table 1), which are closely related to the tetrahedral structure of the P_4 molecule, were found to lie much higher in energy.[4]

While the structure of P_4O is known, very little is known about its chemistry. For the other phosphorus suboxides P_4O_n ($n = 2$–5), even structural information is missing. The question why phosphorus subsulfides have been prepared, but analogous oxides could not be isolated is generally answered by the statement that P–O bonds are more stable than P–S bonds, however, detailed thermodynamical data of phosphorus suboxides are not known. In order to extract a building-up

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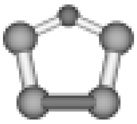
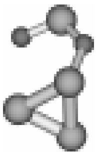
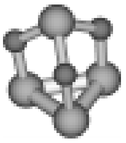
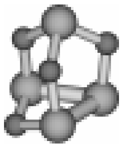
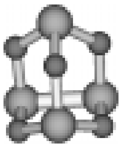
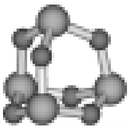
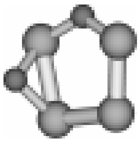
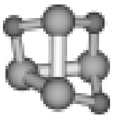
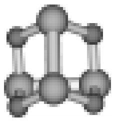
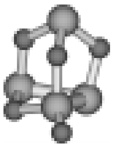
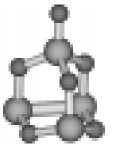
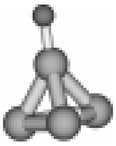
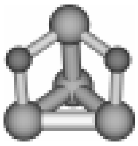
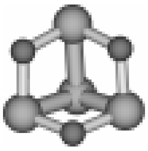
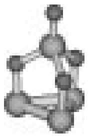
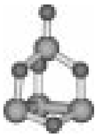
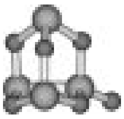
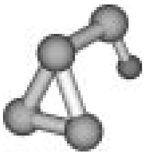
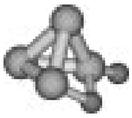
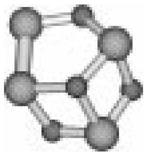
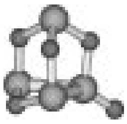
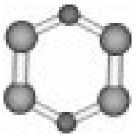
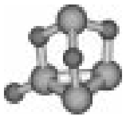
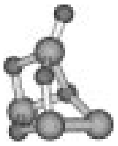
Table 1. Structures and calculated energies [kcal mol^{-1}]. The energies are given relative to the lowest lying isomer of the respective summation formula. The dark centers represent oxygen.

Isomer	P_4O		P_4O_2		P_4O_3		P_4O_4		P_4O_5		P_4O_6	
I		0		0		0		0		0		0
II		15		4		28		1		21		15
III		32		6		29		9		23		20
IV		31		15		29		15		30	–	–
V	–	–		25	–	–		24		39	–	–

principle for phosphorus suboxides the present study determines the geometrical structure and the relative energy of various isomers of compounds with the summation formula P_4O_n ($n = 1-6$); the corresponding heats of formation are computed to gain more insight into the chemistry of phosphorus suboxides.

Due to its unusual bonding a prediction of the most stable isomers of P_4O_n ($n = 2-5$) represents a difficult task. P_4O already illustrates the problem. Its most stable isomer possesses an entirely different structure than that of the tetrahedral P_4, while those isomers structurally related to the tetrahedron lie higher in energy. To ensure that the lowest energy isomer is found in the present study a simulated annealing procedure[5] is used as a first step to obtain the geometrical parameters of a large number of local minima on the potential energy surface (PES) of the respective summation formula. In the second step the energetically low-lying isomers of this set are determined by single point density functional theory (DFT) calculations.[6] Finally, the geometries of the lowest lying isomers are refined using the DFT approach.[7, 8]

Table 1 contains the calculated relative stabilities of various P_4O_n ($n = 1-6$) isomers and includes sketches of the respective geometrical structures.[9] In addition to the known isomers of P_4O[4] Table 1 presents data for a structure (isomer **IV**) closely related to the most stable isomer of P_4O_2 (see below). For P_4O_6 we included in addition to the well-known cage structure information on two possible low-lying isomers (Table 1). According to our calculations, the most stable isomer of P_4O_2 has an unexpected structure. It is composed of an isosceles P_3 triangle and one O-P-O chain. The base of the triangle has a bond length of 2.05 Å, while the two other bonds are somewhat longer (2.2 Å). The P–O bond connecting the P_3 triangle with the O-P-O fragment is found to be 1.75 Å. The P–O bond lengths within the O-P-O fragment are 1.64 Å (inner) and 1.50 Å (terminal), respectively. The reason for the stability of this isomer may lie in the P_3 triangle which was found in many stable P_n clusters.[10] Isomer **II** of P_4O_2 is about 4 kcal mol^{-1} less stable than isomer **I**[11] according to the present calculations. Its structure is closely related to the most stable isomer of P_4O. Isomer **III** of P_4O_2, which is about 6 kcal mol^{-1} less stable than the most stable isomer, can be related to the P_4 tetrahedron by replacing two P–P bonds by P-O-P units. Isomer **IV**, which possesses one terminal P–O bond, is less stable by about 15 kcal mol^{-1}. In their study, McCluskey and Andrews[3] computed further isomers of the type of isomer **V**, but as shown by our computations such structures are even less stable than the structures discussed so far. While the structural relationships between the isomers of P_4, P_4O, and P_4O_2 are quite complicated, the building-up principle for the higher phosphorus suboxides P_4O_n ($n \geq 3$) is straightforward. The energetically low-lying isomers of these phosphorus suboxides are related to the most stable isomer of P_4O_3, which is composed of a PO_3 unit and a P_3 ring. The structures of the low-lying isomers of P_4O_n ($n > 3$) are obtained either by inserting the additional oxygen atom into one of the P–P bonds, that is by replacing a P–P bond by a P-O-P unit or by adding an oxygen atom in a terminal position. Energetically, the former isomers are found to be always lower than the latter. Examples for these rules are P_4O_4 (isomer **I** in comparison to isomer **III**) and P_4O_5 (isomer **I** compared to isomers **II**–**IV**). Similarly, for P_4O_6 only the cage structure (isomer **I**) but not the structure with terminally bonded oxygen (e.g. isomer **II**) is known.[12] Isomer **II** of P_4O_4 is not directly related to isomer **I** of P_4O_3 but to its isomer **II**. However, while for P_4O_3 isomer **II** is about 28 kcal mol^{-1} higher in energy than isomer **I**, for P_4O_4 the energy gap between isomers **I** and **II** is about zero.

From our computed data, the basis for the structural and energetic rules discussed above become clear. For oxygen-rich compounds P_4O_n ($n > 4$) the stability of the P-O-P unit (i.e. two P–O single bonds) in comparison to the stability of one P–P single and one terminal P=O double bond, determines the structures of the energetically most favorable isomers. According to our calculations isomers possessing terminal oxygen atoms lie about 15–25 kcal mol^{-1} above those isomers with a P-O-P unit. For P_4O_n clusters possessing fewer oxygen atoms the P_3 ring represents an additional stabilizing structural unit, which can be seen from the energy difference between isomer **I** and **II** of P_4O_3 (28 kcal mol^{-1}). Both isomers possess the same number of P–O and P–P bonds but only isomer **I** possesses a P_3 ring. Both, isomers **I** and **II** of P_4O_4 possess also the same number of P–O and P–P bonds; in contrast to P_4O_3 neither of these isomers contains a P_3 ring, and in line with our arguments, both structures are found to be essentially equal in energy. In addition the similar stability of both isomers suggests that the question whether the P–P bonds are adjacent or not is of minor importance.

The P_3 ring stabilizes also isomer **III** of P_4O_4. Formally isomer **III** is obtained from isomer **I** by replacing one P-O-P unit by one P–P single bond and one terminal P–O bond. While for P_4O_5 and P_4O_6 such a replacement leads to a destabilization of about 20 kcal mol^{-1}, in the case of isomer **I** and **III** of P_4O_4 a destabilization of less than 10 kcal mol^{-1} is computed. Nevertheless, the stabilization effect of the P_3 ring strongly depends on its substituents. One example is the energy difference between isomer **III** and **V** of P_4O_4 ($\approx$15 kcal mol^{-1}). In isomer **III** the P_3 ring is an equilateral triangle with P–P bond lengths of about 2.24 Å. In isomer **V** the C_{3v} symmetry is distorted by the terminal oxygen atom. While the P–P bonds adjacent to the terminal P–O bond do not change (2.25 Å) significantly, the base of the triangle elongates by about 0.2 Å. As already discussed, the energetical ordering of oxygen-rich phosphorus suboxides P_4O_n ($n > 4$) is determined by the stability of the P-O-P unit. Nevertheless, in these compounds the stabilizing effect of the P_3 ring is also found. One example is P_4O_5, which possesses three isomers differing only in the position of the terminal P–O double bond. While the stabilities of isomer **II** and **III** (in which the P–O bond is adjacent to two or three P-O-P units, respectively) are nearly equal, the energy gap to isomer **IV** (in which the P–O bond is adjacent to one P-O-P unit) is predicted to be 9 and 7 kcal mol^{-1}, respectively. These differences are much smaller than those predicted between isomer **III** and **V** of P_4O_4 underlining again the stability of the P_3 ring. For P_4O_6 the energy difference between the respective isomers (isomer **II** and **III**) is somewhat larger than for P_4O_5 (5 kcal mol^{-1}) and our computations predict the reverse ordering, that is the isomer in which the P–O bond is adjacent to three P-O-P units is more stable than the isomer in which one P–P bond is in the neighborhood of the terminal P–O bond. Besides structural information about larger compounds (i.e. P_mO_n with $m > 4$ and $n = 1 - \frac{3}{2}m$), the thermodynamic and the kinetic stability of the clusters are of great interest. The stability of the various compounds with respect to the most stable elementary form[13] are summarized in Figure 1. As expected, Figure 1 shows that the thermodynamic stability of phosphorus suboxides increases strongly along the series P_4O_n. This strong gain in energy is perhaps one of the main reasons for the difficulties in the attempts to prepare phosphorus suboxides. Besides the increasing thermodynamic stability along the P_4O_n series, we also expect that the polymerization to amorphous phosphorus oxides (i.e. to P_mO_n compounds with $n \gg 4$) is also thermodynamically favorable.

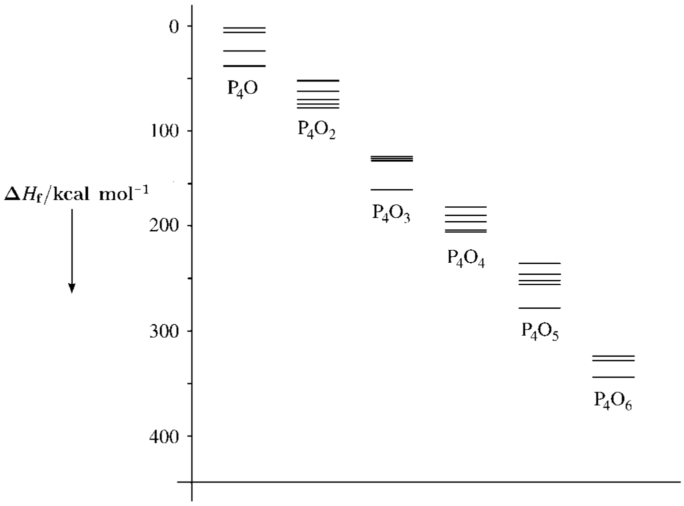


Figure 1. Heat of formation [kcal mol^{-1}] for the computed P_4O_n isomers.

In summary we have discussed energetically low-lying isomers of phosphorus suboxides of the summation formula P_4O_n ($n = 1-6$) and extracted structural factors which determine the relative energy of such compounds. With these data the nature and the stability of various possible bonding types (P–P bonds, P–O single and double bonds, P_3 rings) could be analyzed. In addition, we computed the heat of formation for the various clusters to obtain a key for the experimental difficulties in the preparation of phosphorus suboxides. The findings of the present work are helpful for further investigations of phosphorus suboxides, for example for the study of larger compounds (P_mO_n compounds with $m \gg 4$). The understanding of the chemistry of these compounds is important to gain insight into polymerization processes which hamper the synthesis of molecular phosphorus suboxides. A comparative study of phosphorus suboxides and phosphorus subsulfide is underway.

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[1] H. Novottnick, R. Blachnik, *Z. Anorg. Allg. Chem.* **1999**, *625*, 1966, and references therein.

[2] J. Pelouze, *Ann. Chim. Phys.* **1832**, *50*, 83.

[3] L. Andrews, R. Withnall, *J. Am. Chem. Soc.* **1988**, *110*, 5605.

[4] S. A. Jarret-Sprague, I. H. Hillier, I. R. Gould, *Chem. Phys.* **1990**, *140*, 27; L. Lohr, *J. Phys. Chem.* **1990**, *94*, 1807; L. Lohr, *J. Phys. Chem.* **1990**, *94*, 4832.

[5] For the simulated annealing procedure[14] we used the Monte Carlo method[15] combined with the PM3 approach.[16] Test calculations have shown that PM3 is able to predict reasonable estimates for the geometrical parameters also for P_4O_n isomers that possess unusual bonding.

[6] The DFT computations were performed with the B3LYP functional[17] in connection with an AO basis of double zeta plus (DZP) polarization quality.[18] The second step of our procedure is necessary because for phosphorus suboxides the PM3 method gives reasonable geometries but fails in the prediction of reliable heats of formation.

[7] For the refinement of the geometries an analytical gradient optimization were used. These computations were performed by employing the DFT approach described in reference [6]. Bond lengths obtained from second-order perturbation theory (second-order MBPT theory[19]) differ by less than 0.01 Å.

[8] To test the accuracy of the DFT approach we performed CCSD(T) computations in conjunction with the DZP AO basis for selected systems. In addition we checked the basis set dependency of the B3LYP functional. Both computations point to an error bar of about 3 $\text{kcal}\,\text{mol}^{-1}$ for the relative energies of the various isomers. Furthermore the DFT approach always predicted the same energetical ordering as the more sophisticated CCSD(T) method. In addition we found that the DFT calculations predict the correct energetical ordering for the isomers of P_4S_6 and P_4O_6.

[9] The explicit geometrical data can be obtained from the authors.

[10] M. Haeser, O. Treutler, *J. Chem. Phys.* **1995**, *102*, 3703.

[11] The energy difference between both isomers increases to about 8 $\text{kcal}\,\text{mol}^{-1}$ if the more sophisticated CCSD(T) method is employed. Using a more flexible AO basis (triple zeta quality plus two polarization functions) combined with the DFT approach gave an energy difference of about 6 $\text{kcal}\,\text{mol}^{-1}$.

[12] J. Clade, F. Frick, M. Jansen, *Adv. Inorg. Chem.* **1994**, *41*, 327.

[13] The heats of formation were obtained from the reaction energy of the reaction $P_4+\frac{n}{2}O_2\rightarrow P_4O_n$. All computations were performed on the level described above.[6] The influence of the nuclear motion and temperature effects were incorporated in the standard approach.[19]

[14] *Adaption of Simulated Annealing to Chemical Optimization Problems* (Ed.: J. H. Kalivas), Elsevier, **1995**.

[15] J. P. Valleau, S. G. Whittington in *Modern Theoretical Chemistry, Vol. 5* (Ed.: B. J. Berne), Plenum, New York, **1977**.

[16] J. J. P. Stewart, *QCPE Bull.* **1985**, *5*, 2823.

[17] A. D. Becke, *J. Chem. Phys.* **1993**, *98*, 5648; C. Lee, W. Yang, R. G. Parr, *Phys. Rev. B* **1988**, *37*, 785.

[18] H. Huzinaga, *Approximate Atomic Wave Functions, Vol. 1+2*, University of Alberta, **1971**; A. D. McLean, G. S. Chandler, *J. Chem. Phys.* **1980**, *72*, 5639.

[19] F. Jensen, *Introduction to Computational Chemistry*, Wiley, Chichester, **1999**; J. B. Foresman, A. Frisch, *Exploring Chemistry with Electronic Structure Methods*, Gaussian Inc., Pittsburgh, PA, **1993**.

Alkylating Polymers: Resin-Released Carbenium Ions as Versatile Reactive Intermediates in Polymer-Assisted Solution-Phase Synthesis**

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Dedicated to Professor Ivar Ugi
on the occasion of his 70th birthday

Although combinatorial chemistry was dominated by solid-phase synthesis during its first decade,[1, 2] the solution-phase synthesis remains to show indisputable advantages, for example, in respect to the versatility of reactions applicable and the accumulated knowledge of synthetic protocols. Ideally, a synthetic strategy should be able to combine these merits with the advantages of solid-phase synthesis protocols, such as the possibility to use reagents in high excess, to remove them by filtration, and to employ automated multiple synthesizers. This combination can be realized in polymer-assisted solution-phase (PASP) synthesis either by using scavenger resins or by the implementation of polymeric reagents.[3–5] Especially desirable is the substitution of chemicals which are highly toxic, explosive, or difficult to handle during preparation, reactions, work-up, waste disposal, and in large amounts. Many alkylating agents, such as diazoalkanes, sulfate esters, sulfonate esters, and alkyl halogens belong to this group of hazardous compounds.[6] Solid-supported sulfonate esters have been employed in alkylations of amines and thiols at elevated temperatures under basic conditions.[7]

Elemental nitrogen is an excellent leaving group in alkylations. Therefore we focused our efforts on introducing a novel concept of alkylating polymers based on the release of carbenium ions and nitrogen from precursors bound to insoluble polystyrene gels. The alkylating species are generated from solid-phase-bound 3-alkyl-1-aryltriazenes under acidic conditions and we demonstrate their use as very reactive, mild, and versatile alkylation reagents.[8] The chemistry and applications of triazenes have a long history dating back to the 19th century.[9, 10] Their alkyl derivatives are stable entities unless treated with acid.[11–14] They fragment into radicals when moderately heated,[15] under irradiation[15] or by one-electron oxidation.[16]

Electron-rich triazenes have higher reactivity,[17] and therefore *para*-alkoxy-substituted anilines were selected as efficient starting materials. Solid-supported triazenes have been

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