

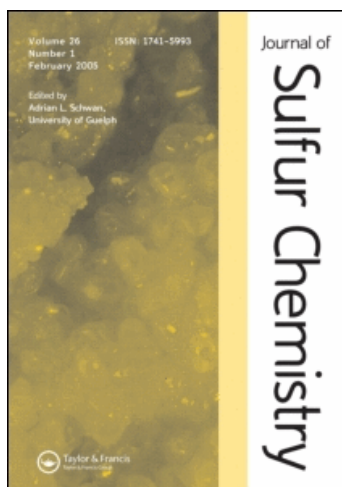
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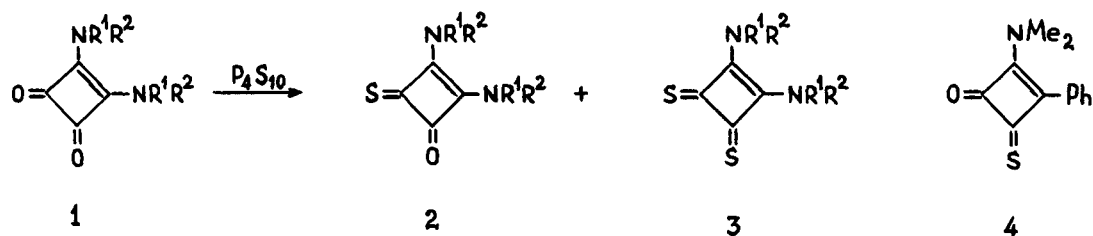
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APPENDIX ADDED IN PROOF

Since the preparation of this review two related reviews have appeared.^{1,2}

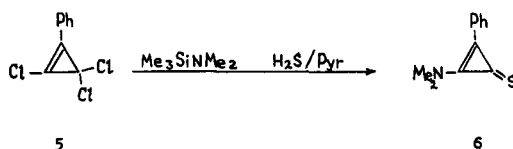
II.

Recently Seitz *et al.* have developed the rather interesting chemistry of thio analogs of squaric acid and its amides. Thus, the N,N,N',N'-tetrasubstituted enamino thioketones 2 and 3 were prepared by reaction of the squaric acid 1,2-diamides 1 with P₄S₁₀ in 1,2-dimethoxyethane or dichloromethane.^{3,4} 3-(N,N-Dimethylamino)-2-oxo-4-phenyl-1-cyclobutenethione 4 was obtained under analogous conditions.⁵



Boron sulfide and silicon disulfide⁶ have been successfully used for the preparation of N-methylthioacridone and other non-enolizable thioketones from the corresponding ketones.

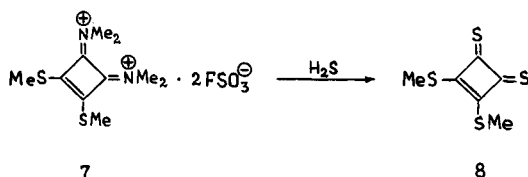
The cyclopropene derivative 6 has been prepared by successive treatment of the trichloro derivative 5 with (N,N-dimethylamino)-trichlorosilane and hydrogen sulfide in pyridine.⁷

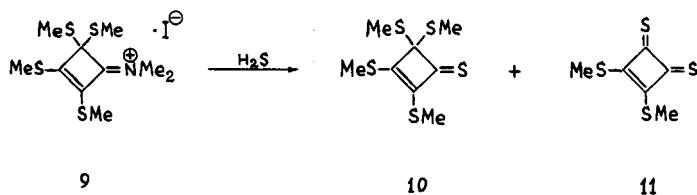


Several recent syntheses of enamino thioketones have been described.⁸⁻¹²

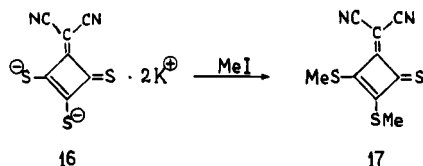
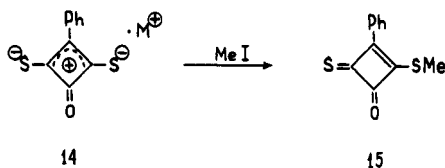
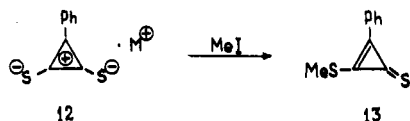
IV and VII.

Also cyclobutenes and cyclopropenes containing an RS—C=C—C=S group are known. Two synthetic approaches to these compounds have been described. The first one is based on the sulfurization of the immonium salts 7¹³ and 9¹⁴ leading to the β-alkylthiovinylene thioketones 8, 10, and 11, respectively.

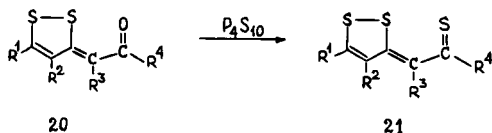
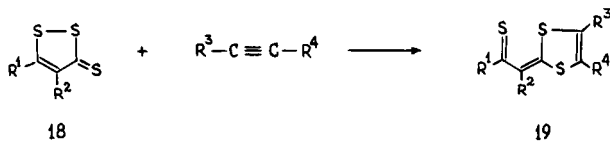




The second route involves the alkylation of the thiolates 12^7 and 14^5 as well as of 16^{15} with methyl iodide which leads to the formation of the unstable 13 and the relatively stable methylthio derivatives 15 and 17 , respectively. Compound 17 is also a rare representative of the β -cyanovinylene thioketones (cf. Section VII.).

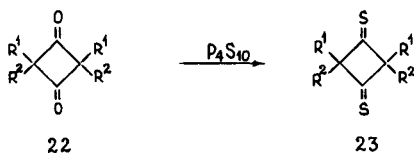


β -Alkylthiovinylene thioketones are structurally similar to the 1,2-dithiol-3-ylidene substituted thioketones 19 . These compounds are obtained by reaction of the corresponding 1,2-dithiol-3-thiones 18 with acetylenes.¹⁶⁻¹⁸ The synthesis of the analogous thioketones 21 is based upon the reaction of the ketones 20 with tetraphosphorus decasulfide.^{19,20}



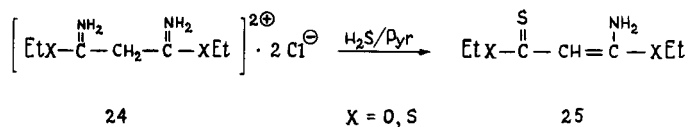
V.

Heating of the 2,2,4,4-tetrasubstituted 1,3-cyclobutanediones **22** with tetraphosphorus decasulfide in pyridine leads to a number of β -dithiodiketones **23**^{21,22} which are unable to enethiolize. The structures of these compounds have been studied by spectroscopic methods²⁴ and by X-ray diffraction.²⁵



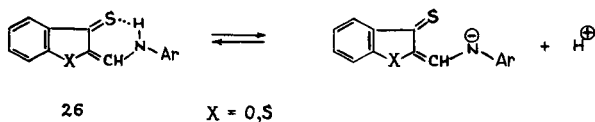
VIII.

The compounds **25** can be considered as both functionally substituted enamino thio-ketone analogs and β -alkoxy- or β -alkylthiovinylene thioketones. They have been prepared, together with other compounds, by treating the malonic acid derivatives **24** with hydrogen sulfide in pyridine.²⁶



IX.

The ionization constants of a number of heterocyclic enamino thioketones **26** have been determined by potentiometric titration in acetonitrile. The effect of the N-aryl group on the free energy of the deprotonation of these compounds has been quantitatively estimated by means of a correlation analysis.²⁷



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