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Reactivity And Selectivity in Nucleophilic Aromatic Substitution Reactions Using Sulfur-Based Nucleophiles

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Abstract:

A new category of nucleophilic aromatic substitutions, short for "conducted" SNAr instead of the more conventional "two-step" SNAr processes, has recently emerged as a result of advances in computational and experimental chemistry. In contrast to conventional SNAr reactions, which need substitutions that remove electrons, which greatly stimulate aromatic ring, concerted routes do not necessitate such groups. Organic chemistry revolves on substitution processes using aromatic rings. Some more processes that are regularly found include electrophilic aromatic substitution, SNAr nucleophilic aromatic substitution, SNArH, and vicarious nucleophilic, which are separate yet similar. The traditional stepwise method was considered the common mechanism for aromatic nucleophilic substitutions, whereas concerted nucleophilic substitutions were very unusual.

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Introduction:

Reagents that are capable of donating an electron pair in a reaction are referred to as nucleophiles, and this kind of reaction is known as nucleophilic. The term "nucleophile" comes from the fact that these molecules prefer to target areas of the substrate molecule with a low electron density, such as positive centers. Nucleophiles have an abundance of electrons. They might be neutral molecules with two free electron pairs or negative ions (such as carbanions). The conductance approach was used by Behera et al. to characterize the kinetics of the reaction between phenacyl bromide and phenacyl thiourea, with the aim of determining the processes. A study conducted by Acharya and colleagues examined the rate of reaction between alkyl iodides and phenacyl bromides in methanol containing 1, 4-dihydro-4, 4, 6-trimethyl-l-(substituted) pyrimidine-2-thiols.Prabhakaran and AnanthaKrishna Nadar have investigated the rate of reaction between phenacyl bromide and various 2-m-/p-substituted phenyl thiazolidines (SPT) in ethanol and at room temperature.

Rates of second order for the reactions of phenacyl bromide with several nucleophiles in acetone-water 60/40 solvent mixes at varying temperatures, including CO3 2-, HCO3 -, OH-, CH3COO-, N3 -, SO4 2-, phenoxide, imidazole, pyridine, aniline, and thiourea an affinity chromatographic technique for homogeneous purification of human coagulation factor utilizing monoclonal antibodies has been developed by the American Red Cross. In most cases, 2-Fluoro-2-methylpyridinium toluene-4-sulfonate is used to activate the resin supports. Researchers have used 2-fluoro-l-methylpyridinium toluene-4-sulfonate in their studies on photo-activated monoclonal antibody tagging. Research in this area includes chemically modifying E. coli RNA polymerase -subunits, studying lectin interactions with cell surfaces, and analyzing the RNA secondary structure of the Moloney Murine I eukemia virus packaging signal, among other things. applications of l-Cyclohexyl -3- (2-morpholinoethyl) cardodiimide metho-ptoluenesulfonate.

To identify the particular the liver’s plasma membrane uses 1-Anilino-8-naphthalenesulfonate, an organic anion binding protein. The 2-N-Acrylamino-6-naphthalenesulfonate demonstrates the specific quality. Use it as a probe for histone intrinsic and extrinsic fluorescence studies; it fluoresces moderately in water but strongly in organic solvents. Large doses of its sulfate salt, which forms a stable p-toluene sulfonate complex, when given to depressed patientsthe metabolite S-adenosyl-L-methionine (SAMe) seems to improve their mood. For the O-sulfation process, one uses l-Cyclohexyl-3-(2-morpholoethyl) carbodiimide p-toluenesulfonate. An essential component in the synthesis of many compounds, including thiophenols, sulfonic acid esters, sulfones, sulfonamides, sulfonohydrazides, and sulfonyl fluorides, is benzenesulfonic chloride, or BSC. The transformation of 2-lithio-l- (phenylsulfonyl) indole to 2-chloro-l- (phenyl sulfonyl) indole using benzenesulfonyl chloride proved that sulfonyl halides are a major source of Cl-.

For the live radical polymerization of acrylates, styrene(s), and methacrylate, arene sulfonyl halides serve as useful initiators. Sulfonyl transfer reagents include tosyl chloride, abbreviated as TsCl. The following processes utilize it: In order to create chlorides or intramolecular and intermolecular displacements, alcohols can be O-sulfonated. Vicinal diols can be used for epoxidation, 1, 3-diols for oxetane formation, carboxylic acids can be esterified or decarboxylated. Aliphatic amines can be N-sulfonated for protection, and C-sulfonated alkenes and silyl alkynes can be C-sulfonating. Urea can be dehydrated, and the formation of formamides and amides. One possible explanation for tosyl chloride’s reactivity is a polar effect. This occurs when the electron-withdrawing Cl atom creates an electron deficient center at the tetra coordinated sulfur atom that holds the halogen atom. As a result, a nucleophile may more easily approach this sulfur atom.

Literature Review:

Rohrbach, Simon et.al. (2019). Experimental and computational chemists have recently discovered a new alternative to the conventional two-step SNAr procedures, a family of nucleophilic aromatic substitutions known as concerted SNAr. There has been rapid growth in this class. The presence of electron-withdrawing substituents is required for traditional SNAr reactions to strongly activate the aromatic ring, concerted routes do not necessitate such activating groups.

Halldin Stenlid, Joakim et.al. (2017). Based on the molecular density contour at 0.004 a.u., the E(r) takes into account any virtual orbital situated below the limit of free electrons. There is a fair to outstanding correlation (R^2=0.98) between the descriptor values and the experimental reactivity data for six distinct series of electron-deficient arenes. Five- or six-atom rings with different fluorine, bromine, and hydride leaving group configurations are among them. In addition, they include homo- and heteroarenes. Furthermore, the solvent, temperature, and nucleophile are different throughout the sequence. The surface E(r) [ES(r)] performs better than transition-state computations in predicting reactivity for a coordinated SNAr reaction with a bromine nucleofug, gives much stronger correlations than LUMO energies, and is typically more reliable than the molecule electrostatic potential. With the use of ES(r), it is feasible to accurately anticipate isomeric distributions and to determine the electrophilic sites on a molecule. Reactivity predictions for the important class of nucleophilic aromatic substitution processes are provided by the descriptor in a timely and precise manner. due to the computationally cheap computations of ES(r). There are potential uses in fields such as toxicology, drug discovery, and synthesis.

Liljenberg, Magnus et.al. (2012). In kinetically regulated “nucleophilic aromatic substitution” processes, we have explored computationally efficient and practically applicable approaches for quantitatively predicting the Regio isomer distribution. One approach utilizes density-functional theory calculations to determine the intermediates of the isomeric Ͻ-complex and their respective stabilities. We show that for neutral nucleophiles the leaving group is HF and for anionic nucleophiles the leaving group is F (-), quantitative applications of this method’s predictions are possible. Due to challenges in identifying pertinent F-complex structures, the ’-complex method was ineffective whether the leaving group was Cl/HCl or Br/HBr, independent of the anionicity or neutrality of the nucleophile. By using such transition state structures and assuming a concerted substitution process, we were able to derive quantitatively useful results. Our findings align with previous theoretical investigations, which have shown stable σ-complexes in some circumstances and suggested that others go through a coordinated replacement process.

Sample, Harry et.al. (2020). For more than 150 years, people have understood and made extensive use of the nucleophilic substitution of aromatic moieties (SNAr) to functionalize (hetero)aromatic systems. There are now a number of recognized “types” of SNAr reactions, with significant applications in the field of porphyrinuria macrocycles. We describe in this section the SNAr responses of seven different kinds of porphyrinurias that vary in the amount and kind of pyrrole units: subporphyrins, norcorroles, corroles, porphyrins, azuliporphyrins, N-confused porphyrins, and phthalocyanines. We examine the different types of nucleophiles and the specific sites of substitution (α, β, or meso) for each one. Along with this, we assess this pathway as a synthetic method for producing unsymmetrical porphyrinoids. No matter the nucleophile, distinct patterns emerge for all porphyrinoid types. Nucleophilic substitution on porphyrinoids is often discovered to be an economical method that may produce intricate substituent patterns. This process can be carried out in non-anhydrous solvents using simple porphyrinoids that are readily available.

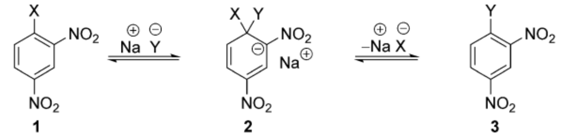
Crampton, Michael. (2015). We examine the data supporting the bimolecular SNAr mechanism and talk about how various leaving groups and activating groups in the arene ring affect reactivity. Base catalysis methods involving amine nucleophiles are examined, and reactivity variations according to the attacking nucleophile are reported. In addition, the Smiles rearrangement and other intramolecular rearrangements are considered, as are solvent effects. Formation of comparatively stable σ-adducts or Meisenheimer adducts may occur as a consequence of interactions between nucleophiles and electron-deficient arenes. The complexes are supported by spectroscopic and crystallographic data, and the diverse arenes and nucleophiles that contribute to their formation are addressed. It is suggested that adducts may be produced from very electrophilic substances like 4, 6-dinitrobenzofuroxan. We take into account the arenediazonium ion breakdown processes that are heterolytic and homolytic. Synthetic uses of nucleophilic replacements often include displacements of leaving groups, such as halogens and nitro groups. Intramolecular reactions involving sulfur, nitrogen, carbon, and oxygen nucleophiles are covered.

Aromatic Substitution Reactions

Organic chemistry revolves on substitution processes using aromatic rings. Some more processes that are regularly found include electrophilic aromatic substitution, SNAr nucleophilic aromatic substitution, SNArH, and vicarious nucleophilic, which are separate yet similar. SN1 reactions, benzyne intermediate substitutions, radical mechanisms (e.g., direct metalations, sigma tropic rearrangements, and electron transfer based SRN1 reactions and base-promoted homolytic aromatic substitution (BHAS) couplings and so on. Due to the sheer breadth of these chemical disciplines, only a few of seminal reviews or up-to-date sources can do them justice. The realization that coordinated SNAr reactions, rather than the usual two-step reactions, may account for a large portion of these processes has led to a surge of interest in SNAr reactions among these other kinds of reactions.

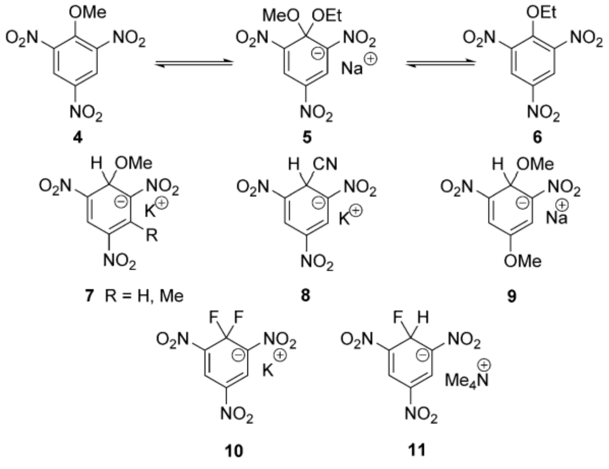
Atomic Substitution in A Classical Nucleophelial Model

Researchers have been looking on nucleophilic aromatic substitutions since the 1870s. The typically acknowledged process, shown in Scheme 1 for dinitroarene 1, included a two-



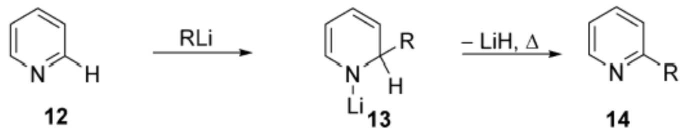
Scheme 1. Classical two-step mechanism for SNAr reactions.

In this stage of the process, which included a Meisenheimer intermediate, the arene becomes significantly more active for substitution when one or more electron-withdrawing substituents, preferably nitro, are present in the ortho or para positions to stabilize the resonance.”Concerted reactions are the exception rather than the rule” and “there is little doubt that most of the activated SNAr substitutions must proceed through the early-recognized addition-elimination mechanism” were written in Terriers’ 2013 top-notch book on SNAr reactions. Isolation of intermediates led to arguments in support of a two-stage replacement. According to Bunnett and Zahler’s 1951 assessment, some reactions resulted in the formation of isolated intermediate adducts (Scheme 2). Important research.



Scheme 2. Some known Meisenheimer intermediates.

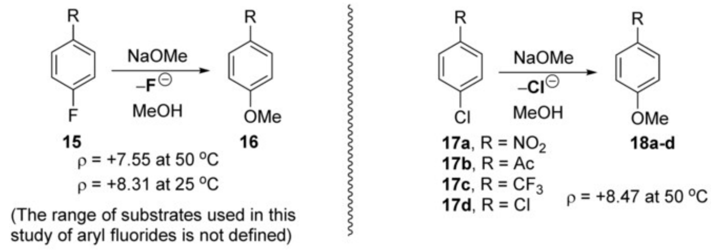
Undertook these procedures; To isolate a shared intermediate, Meisenheimer combined methyl ether 4 with sodium oxoet and ethyl ether with sodium molten melt. This intermediate was decomposed into a mixture of the parent ethers when the reaction was acidified. This kind of adduct step, also known as Meisenheimer intermediates, are extensively studied and used in organic chemistry. An essential and prevalent process in medicinal chemistry and agrochemistry is the nucleophilic aromatic substitution of pyridines, pyrimidines, and related heterocycles. Where excellent leaving groups are present, we have isolated intermediates from these replacements before; but, when bad leaving groups are at work, we are acquainted with the process. Isolatable intermediates at the other end of this spectrum include salts formed when organolithium compounds are added to pyridines; for example, a chemical that, when heated, yields the substituted pyridine while removing LiH (Scheme 3). Producing and



Scheme 3. Organolithium additions to pyridine, and re-aromatization.

The results of the Hammett experiments, which show that the rate-determining step is linked with substantial positive ρ values, provide evidence that the substitution processes are nucleophilic. When comparing ρ readings, keep in mind that they change depending on the experiment's temperature.

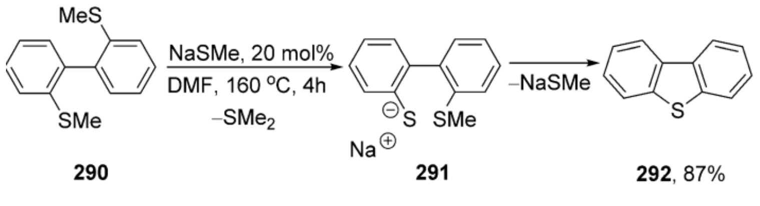
The rate-determining phase is characterized by a significant accumulation of negative charges, as seen in Miller's examples (Scheme 4). It is acknowledged that several Although the cases below in Scheme 4 have very high ρ values, SNAr responses still have values between +3 and +5.Examining the substrates that Miller selected provides insightful information. There were four instances of his substrate series 17 a-d, with R=NO2, Ac, CF3, and Cl. It is obvious that CF3 and Cl cannot delocalize a negative charge via resonance, in contrast to NO2 and Ac, but they may nonetheless contribute inductive stabilization to varying degrees. Miller's Hammett analysis revealed a strong relationship between the four substrates and σ* for these substituents, indicating that they share a similar mechanism.



Scheme 4Miller's studies of Hammett correlations.

Sulfur Nucleophiles

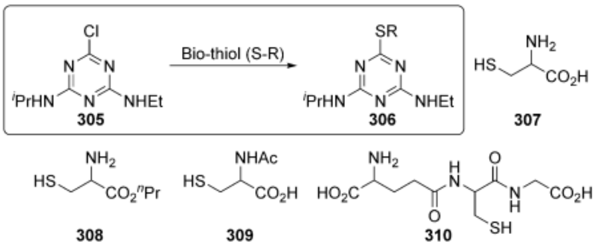
A lot of recent research has focused on sulfur nucleophiles as well. An interesting consequence to the Tobisu, Chatani, et al. recently reported a reaction of 2, 2'-bis(methythio)-1, 1'- biaryls 290 (Scheme 5) with catalytic amounts of methanethiolate salts in dimethylformamide (DMF) as a solvent. The reaction begins with the first step of produce an arenethiolate by demethylating the ArS-Me link.



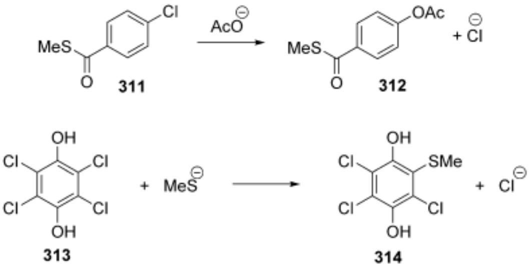
Scheme 5. cSNAr reactions in the formation of dibenzothiophenes.

Conducted theoretical and experimental investigations into the atrazine 305–biothiol 307–310 reaction and postulate that it happens on the cusp of two distinct reaction pathways: coordinated and stepwise.

Computing Despite a Brønsted plot showing b = + 0.5, which corresponds to a stepwise process via a Meisenheimer intermediate, the intrinsic reaction coordinate says that no Meisenheimer intermediate can be found. The authors speculate that this could be because chloride is being lost at a fast pace. Scientists have discovered that 4-chlorobenzoyl CoA model compound 311's chloride is nucleophilically aromatically displaced (Scheme 6). by means of the acetate ion. In the same investigation, thiomethanolate was used to replace chloride in tetrachlorohydroquinone by a nucleophilic aromatic substitution.



Scheme 6. There is a fine line between coordinated and sequential methods when it comes to thio-dehalogenation of atrazine 305.



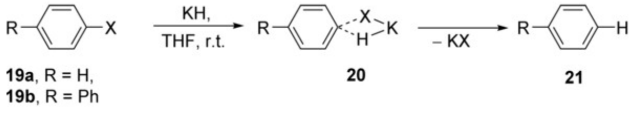
Scheme 7. Bio-inspired substitution reactions.

The semiempirical technique (PM3) to go forward in tandem. It is possible, as the authors note, to stabilize the building "negative charge" in the solution phase or on the enzyme during the transition stage. Therefore, a Meisenheimer intermediate may play a role in the reaction's stepwise pathway in the solution phase, as opposed to the gas phase, when the reaction follows a coordinated approach.

Concerted Nucleophilic Aromatic Substitution (Csnar)—Early Developments

Despite the widespread acceptance of two-stage SNAr reactions in the literature, more and more abnormalities began to surface, especially when considering substrates such as 17-d, which were studied by Miller., were reactive. therefore, alters how we see the frequency of cSNAr responses.

As an early example, consider the study that investigated the interaction of KH with aryl halides in a single 1980 publication. Under Scheme 8, the hydrodehalogenation of substrates 19 was carried out using tetrahydrofuran (THF) as the solvent. Although thorough mechanistic studies were not conducted to investigate the responses, the findings that were obtained provided valuable insights.



Scheme 8 Proposal for concerted S_NAr reactions

Nucleophilicity of Sulfur Compounds

We name thiols and mercaptans the sulfur analogues of alcohols, and we call sulfides the sulfur analogues of ethers. There are significant differences in the chemical behaviors of ethers and alcohols and thiols and sulfides. We predicted that thiols would be sturdier acids than comparable “alcohols” and “phenols”, and we are not surprised; hydrogen sulfide (H2S) is a ten-million-fold stronger acid compared to water. In the alkyl halide and tosylate SN2 reactions, thiolate conjugate base reactions have shown to be efficient nucleophiles, and they are easy to produce.

$$R\text{–}S^{(-)}\ Na^{(+)} + (CH_3)_2CH\text{–}Br \longrightarrow (CH_3)_2CH\text{–}S\text{–}R + Na^{(+)}\ Br^{(-)}$$

There are a lot of fascinating and practical electrophilic replacements of sulfur that are often not seen for oxygen, even though the number of replaceable hydrogen ions of ethers is significantly higher compared to sulfides. This is because sulfur has a significantly higher nucleophilicity than oxygen. For instance, “ternary sulfonium salts” are produced when sulfides react with alkyl halides, much as “quaternary ammonium salts” are formed when 3-amines are alkylated. The “oxonium salts of ethers” that are chemically similar do exist, however, they showhigh reactivity and need very harsh preparation methods. Despite sulfur’s limited positive charge and oxygen’s partial “negative formal” charge, sulfoxides sulfinate salts and sulfite anion may alkylate on sulfur.

States of Oxidation Compounds of Sulfur

There are only two possible oxidation states for oxygen in organic molecules: -1 for peroxides and -2 for all other compounds. Comparatively, the oxidation states of sulfur ranging from -2 to +6 has been represented in the table below. (orange represents certain simple inorganic compounds).

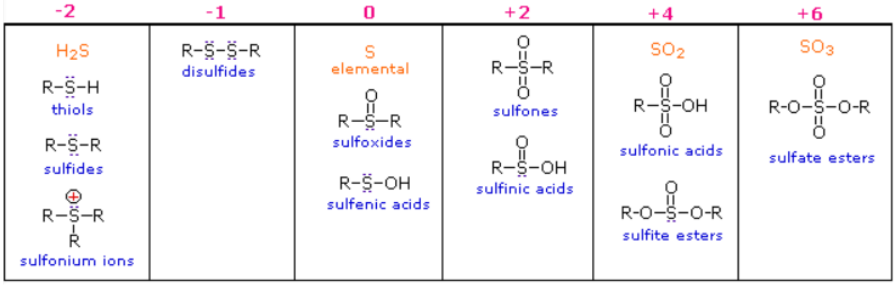


Figure 1. Sulfur Oxidation States in Organic Compounds

To depict sulfur atoms in the presented compounds Lewis-structures can be a useful tool. Higher oxidation states mostly exhibit “formal charge separation” with the constrained formulae – valence shell electron octets, as mentioned before. The double bonding that increases sulfur’s valence octet in these formulations neutralizes this charge gap. True, S=O double bonds do not include the typical ρ and π-orbitals which is quite common in carbon double bonds. Sulfur’s five vacant 3d-orbitals for p-d bonding, which is analogous to p-p (π) bonding, since it is a third-row element. Sulfur compounds, such as sulfoxides, may add two electrons to eight valence electrons in the outermost shell, whereas sulfones can add four. Because of sulfur, pairing of non-bonding electron takes place occupying one corner of a tetrahedron with sulfur in the core, sulfoxides have a permanent pyramidal form.

Thus, sulfoxides are chiral if they include two distinct alkyl or aryl groups as their substituents. It is possible to separate enantiomeric sulfoxides, and they are stable. When compared to alcohols, thiols’ oxidation chemistry is quite different. When 1° and 2°-alcohols are oxidized to produce ketones and aldehydes, it alters carbon’s oxidation state, but oxygen remains unchanged. Thiols and other sulfur compounds undergo oxidation, which changes sulfur’s oxidation state instead of carbon. The following examples show a few sulfur oxidations that are characteristic of the reaction. Thiols are changed to disulfides in the first scenario by moderate oxidation. Typically, it is not seen that alcohols undergo a comparable oxidation to peroxides. The causes of this deviation in conduct are easily discernible. In peroxides, the strength of the O-O bond is over double that of the S-S bond, and the strength of the O-H bond is almost 25 kcal/mole more than that of an S-H molecule. Therefore, disulfide production is thermodynamically preferred to peroxide creation.

Conclusion

The traditional stepwise method was considered the common mechanism for aromatic nucleophilic substitutions, whereas concerted nucleophilic substitutions were very unusual, according to the review. Computational methods that elucidate the mechanics have aided investigations. Substrate nucleophile and leaving group, as well as environmental factors, have a significant impact on whether reactions are concerted or stepwise, and some substitutions may seem as either depending on the circumstances. Reagents that are capable of donating an electron pair in a reaction are referred to as nucleophiles, and this kind of reaction is known as nucleophilic. Organic chemistry revolves on substitution processes using aromatic rings. Some more processes that are regularly found include electrophilic aromatic substitution, SNAr nucleophilic aromatic substitution, SNArH, and vicarious nucleophilic, which are separate yet similar.

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