

# Review on Synthesis and Application of Azo Dyes Derived from Some Natural Phenols Like Carvacrol Eugenol and Thymol

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**Abstract:** Azobenzene motifs have a broad application across numerous scientific domains. Azo compounds are useful in the chemical industry because of their significant properties. Aromatic azo compounds have primarily been used as dyes since their discovery. The azo coupling reaction is the conventional process of creating azo compounds by combining diazonium salts with activated aromatic compounds such as arylamines and phenol. Reaction between a diazonium salt and another substance that contains an aromatic ring titled as a coupling agent. Combining benzene diazonium salt, alkaline phenol, and amines produced Yellow, red, and orange azo compounds. The synthesis and use of azo dyes derived from natural phenols, such as thymol, carvacrol, and eugenol, as coupling agents are summarized in this review. Thymol, eugenol, and carvacrol are some examples of naturally occurring ten-carbon phenols that occur in the essential oils of a variety of plants. These adaptable molecules find use in the agricultural, pharmaceutical, fragrance, cosmetic, and flavor industries as well as in many food products as helpful ingredients. They have numerous biological and pharmacological actions, including anti-inflammatory, antimicrobial, analgesic, anticancer, and antioxidant effects.

**Keywords:** *Thymol, Eugenol, Carvacrol, Synthesis, Azobenzene, Applications.*

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## I. INTRODUCTION

Any compound that has two carbon atoms connected to one or more azo groups (-N=N-) has a wide range of applications. Azo dyes account for roughly 50% of all dyes used in food and textile production [1]. Aromatic azo compounds are used in clothing, plastics, cosmetics, and food beverages as acid-base indicators, biological stains, and commercial colorants [2-4].

Larger-scale production of azo dyes is possible because the synthesis procedure is very straightforward and the raw materials are affordable and widely accessible. Because the reactions are typically conducted at lower temperatures and with water as the primary solvent, the environmental impact is minimized. Each of these elements helps to make azo dyes more inexpensively [5-8]. Azo dyes are heat stable, exhibit greater stability in comparison to natural dyes across the entire pH range of food, and do not deteriorate in the presence of light [9] or oxygen [10]. Tartrazine, yellow2G, sunset yellow, azorubin, and other azo dyes are used in food products due to their low toxicity,

decreased allergic reactions, and lack of hyperactivity effect [11]. Many azo-dyes, including methyl red, methyl orange, and Congo red, can be employed as acid-base indicators due to their ability to act as weak acids. Changes in the degree of electron delocalization result in variations in colour: greater delocalization causes the absorption maximum to shift to longer wavelengths and redder light absorption, whereas less delocalization causes the absorption maximum to shift to shorter wavelengths.

Changes can also be due to geometrical isomerism of the azo group. In non-textile applications, azo dyes have also been used in lasers, nonlinear optical systems [12], reprography [13], dye-sensitized solar cells [14], and metallochromic indicators [15].

A coupling agent is another chemical having an aromatic ring that reacts with a diazonium salt. The electrophilic diazonium salt reacts with the coupling agent's aromatic component creates a coloured azo compound precipitate right away. Because azo chemicals are stable, dyes don't fade. By joining an extended delocalized system,

the azo group stabilizes. The outcome of a coupling process between a coupling agent and a diazonium salt. Benzene diazonium salt and alkaline phenol and amines give yellow, red and orange azo compound. Because of its high demand, azodyes are typically made in industry using synthetic phenol or phenol derived from coal tar. Phenols are both synthesized industrially and produced by plants and microorganisms naturally [16].

Several natural phenol derivatives such as carvacrol [5-isopropyl-2-methylphenol] [17], thymol [5-methyl-2-(1-methylethyl) phenol] [18], and eugenol (2-methoxy-4-(2-propenyl) phenol) [19], according to reports, their structural counterparts exhibit antibacterial properties. Carvacrol and thymol are frequently utilized as antioxidants and preservatives in the food industry and cosmetics [20]. Throughout their range, thymus species are utilized in cosmetics and fragrances, as well as as fragrant and medicinal plants.

Most aspects of their medicinal use are related to the essential oil which contains various levels of thymol and/or carvacrol and phenolic derivatives possess potent and broad-ranging antibacterial properties [21-24].

Eugenol (4-allyl-1-hydroxy-2-methoxybenzene) is naturally occurring in many angiospermic plants with a pleasant scent and spicy taste with anti-convulsive, anti-microbial, anti-inflammatory and anti-oxidative properties. Used for a natural food flavour and as a preservative [25-29].

According the common uses of these natural phenols, we focused in this review to investigate our interest in creating azo dyes with these phenol moieties led us to consider creating a new line of azo dyes using these natural phenols in order to find synthetic methods for creating new, selective, and less harmful azo dyes. The synthesis, structural characteristics, uses, dyeing qualities, and antimicrobial assessment were gathered here. These novel azo dyes, which use azo linkages to combine various substituted aromatic amines with natural phenolic moiety.

## II. NATURAL PHENOLS

Phenols are abundant in nature, particularly in the kingdom of plants. Thymol, eugenol and carvacrol are some examples of naturally occurring phenols with a ten-carbon unit that can be found in the essential oils of a variety of plants. These adaptable molecules find use in the agricultural, pharmaceutical, fragrance, cosmetic, and flavour industries as well as in many food products as helpful ingredients. They have numerous biological and pharmacological actions, including anti-inflammatory, antimicrobial, analgesic, anticancer, and antioxidant effects.

### A. Thymol Occurrence and its Applications

Thymol, its isomer of carvacrol, also referred to as 2-isopropyl-5-methylphenol (**Figure 1**), is a phenolic compound that is found in the essential oils of *Thymus vulgaris* (Lamiaceae) and is a member of the phenolic monoterpene family. It is currently used as a strong food antioxidant [30, 31]. It is derived from the thyme and mint oils, and research has shown that it has both antibacterial and antiseptic qualities [32]. Terpene type substances, such as thymol's monoterpene derivatives, have reportedly been shown to have anti-tumor properties [33]. Thymol is still soluble in some alcohols and organic solvents, despite being less soluble in water at neutral pH [34]. Its active ingredient in several products, as well as a food preservative, antimicrobial, antispasmodic, antioxidant, and anti-inflammatory properties have been noted [35, 36]. It is a compound that is derived from *p*-cymene and is known to have antiseptic and antimicrobial properties [37, 38]. Thymol has been used for its anticancer properties, according to certain studies [39]. Numerous studies have verified the antioxidant properties of carvacrol and thymol, indicating that their addition as nutritive components could enhance the development of new functional foods [40]. The dental drug industry is drawn to thymol because of its ability to prevent caries and plaque [41].

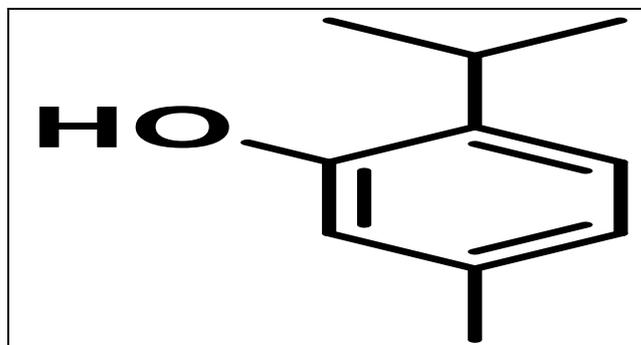


Fig 1: Structure of Thymol (2-Isopropyl-5-Methylphenol).

According to studies on environmental breakdown, particularly thymol and other hydrocarbon monoterpenes breakdown quickly in the environment (DT50 16 days in water, 5 days in soil). Because of this fast dissipation and low bound residues, thymol poses little risk to the environment, which supports its use as a pesticide agent. This is because thymol is a safe substitute for other, more persistent chemical pesticides that can be diluted in runoff and cause subsequent contamination [42].

### B. Carvacrol Occurrence and its Applications

Carvacrol  $C_6H_3(CH_3)(OH)C_3H_7$ , (**Figure 2**) is a phenolic monoterpene compound; 5-isopropyl-2-methylphenol, (carvacrol), is a key component of essential oils of oregano, thyme, wild bergamot and pepperwort. Thyme subspecies essential oil has a carvacrol content of 5% to 75%, whilst *Satureja* (savory) subspecies have a level of 1% to 45%. Carvacrol content is high in *Origanum Majorana* (marjoram) and *Dittany of Crete*, at 50% and 60–80%, respectively. It has a characteristic pungent, warm odour of oregano [17, 43].

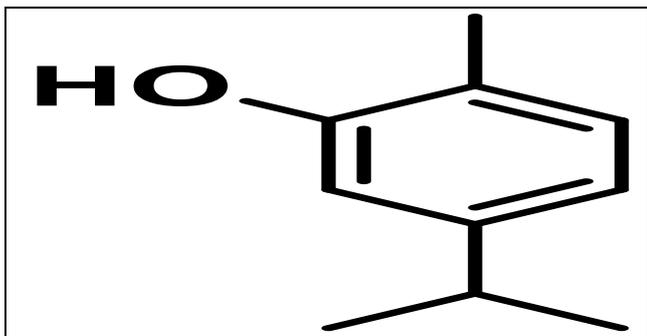


Fig 2: Structure of Carvacrol (5-isopropyl-2-methylphenol).

Its biological properties, including a broad range of antimicrobial activity, have garnered significant attention recently, making it the primary active ingredient responsible for the biological activity of essential oils and the isomer thymol (2-isopropyl-5-methylphenol) [44–45]. Many studies have reported the antioxidative, anti-inflammatory, antibacterial, antifungal, [46] anticarcinogenic, antidiabetic, antinociceptive, cardioprotective, and neuroprotective properties of carvacrol. These characteristics are ascribed to its hydrophilic qualities linked to the phenolic OH group and its hydrophobic qualities linked to the substituted aromatic ring [39, 47–55]. There are not many long-term genotoxic concerns associated with carvacrol. Carvacrol can be an effective antibacterial and antimicrobial agent due to its cytotoxic impact [56]. Its good pest-controlling potency has also been established, and the Food and Drug Administration (FDA) has permitted its use as an ingredient in food products [57–59].

### C. Eugenol Occurrence and its Applications

Eugenol (4-allyl-2-methoxyphenol) is a significant constituent of naturally occurring (Figure 3) phenylpropanoids [60], and it can be found in thyme and cinnamon as well as clove oil. A significant proportion of cloves contain eugenol, which is also found in basil, lemon,

cinnamon, nutmeg, and balm [61]. However, eugenol is primarily extracted from cloves, accounting for up to 80–90% of clove bud oil and 82–88% of clove leaf oil [62].

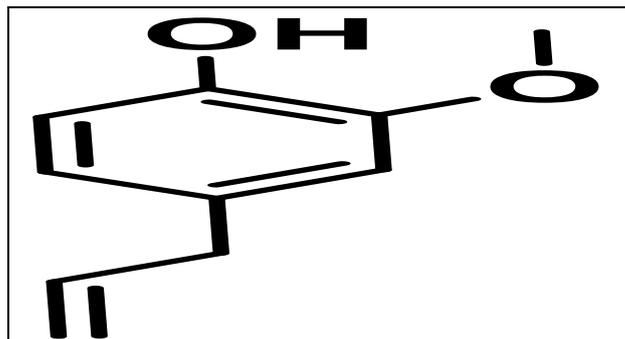


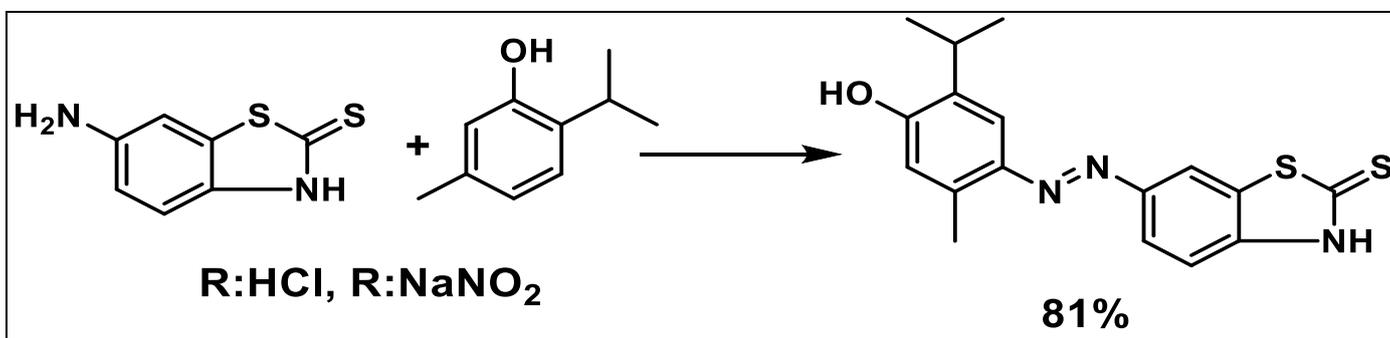
Fig 3: Structure of Eugenol (4-allyl-2-methoxyphenol).

This naturally occurring phenol has anti-inflammatory, anti-pyretic, anesthetic, anti-fungal, antibacterial, anti-cancer, antioxidant, and insect-repelling properties. Eugenol is widely used in many industries, including pharmaceuticals and cosmetics, because of its low toxicity, minimal side effects, and non-metabolized residue [63–64]. Due to its pleasing scent, it is utilized in perfumery, as a food flavoring, as an antiseptic and disinfectant in mouthwash [65, 66] for dental products, and in numerous other applications [67–69]. Its antibacterial and analgesic qualities make it useful in dentistry; when combined with zinc oxide, it creates cement for temporary tooth fillings [70].

## III. AZO DYES SYNTHESIS AND APPLICATIONS

### A. Thymol based Azo dyes Synthesis and Application

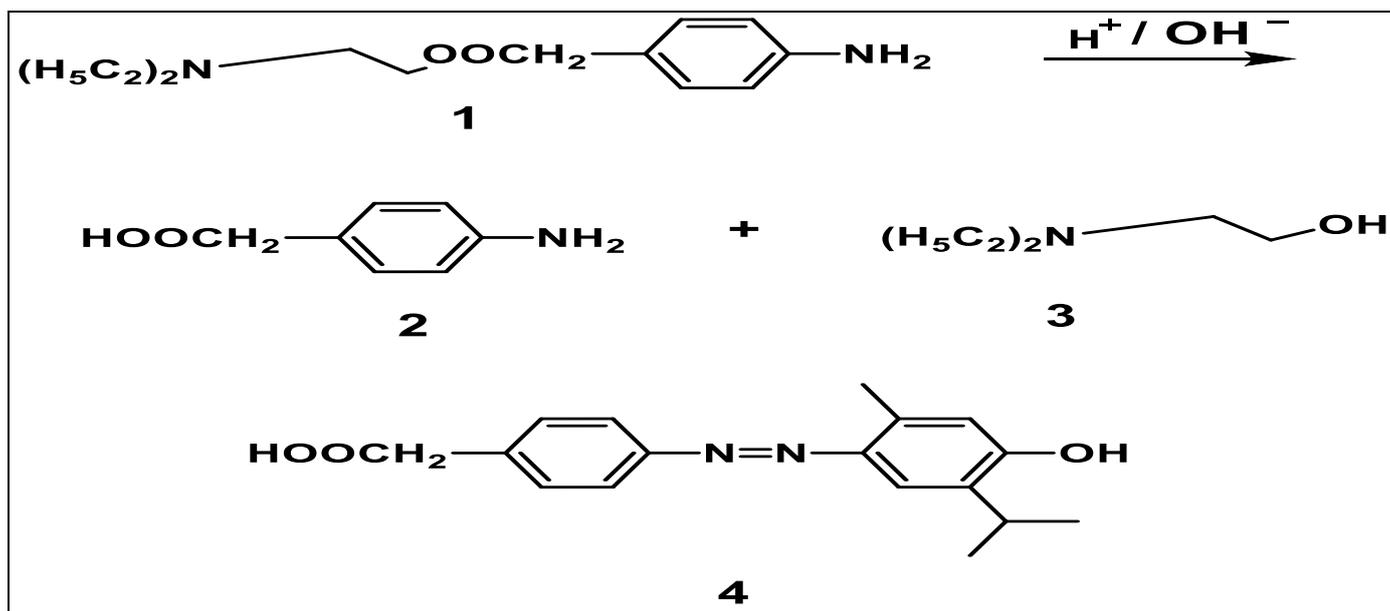
Lipthay et al [71] synthesized 11 new 6-(4-hydroxy-5-isopropyl-2-methylphenylazo)-2-substituted mercaptobenzothiazoles [Scheme 1] and studied their antiprotozoal activity.



Scheme 1: Synthesis of azo dyes from Thymol with heterocyclic moiety.

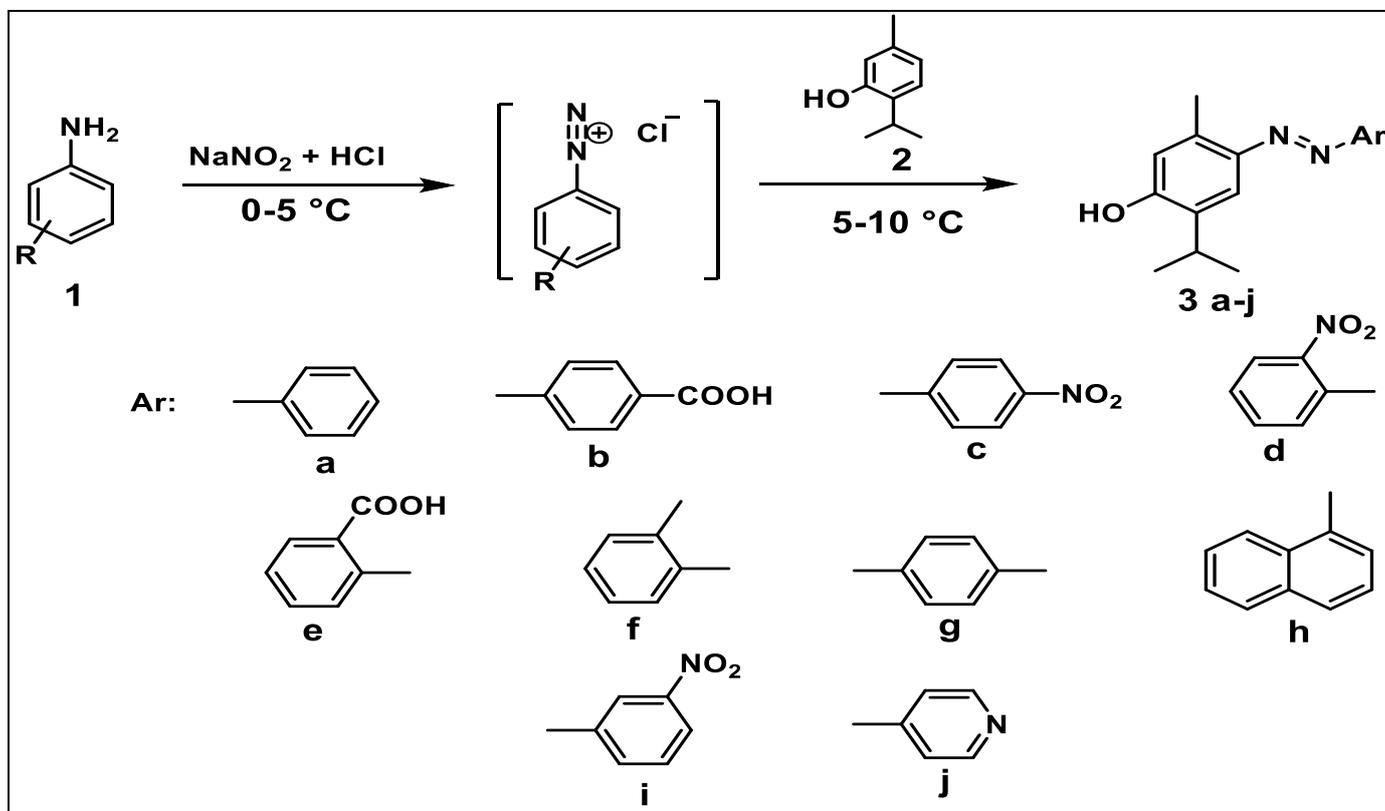
Busing et al [72] synthesized azo dye from 4-aminobenzoic acid coupling with thymol [Scheme 2] to generate evidence that 4-aminobenzoic acid is a byproduct of procaine degradation. To resolve the question of how the decomposition of procaine can be quantitatively recorded and followed analytically. At that time specified TLC method was impractical for quantification purposes. So,

quantification method which is based on a photometric measurement of the azo dye of 4-aminobenzoic acid with thymol is much more suitable. In actual quantification which is formed after separation of procaine by extraction with chloroform by diazotization of the 4-aminobenzoic acid residual in the aqueous layer and coupling with thymol.



Scheme 2: Azo dye from 4-aminobenzoic acid and Thymol.

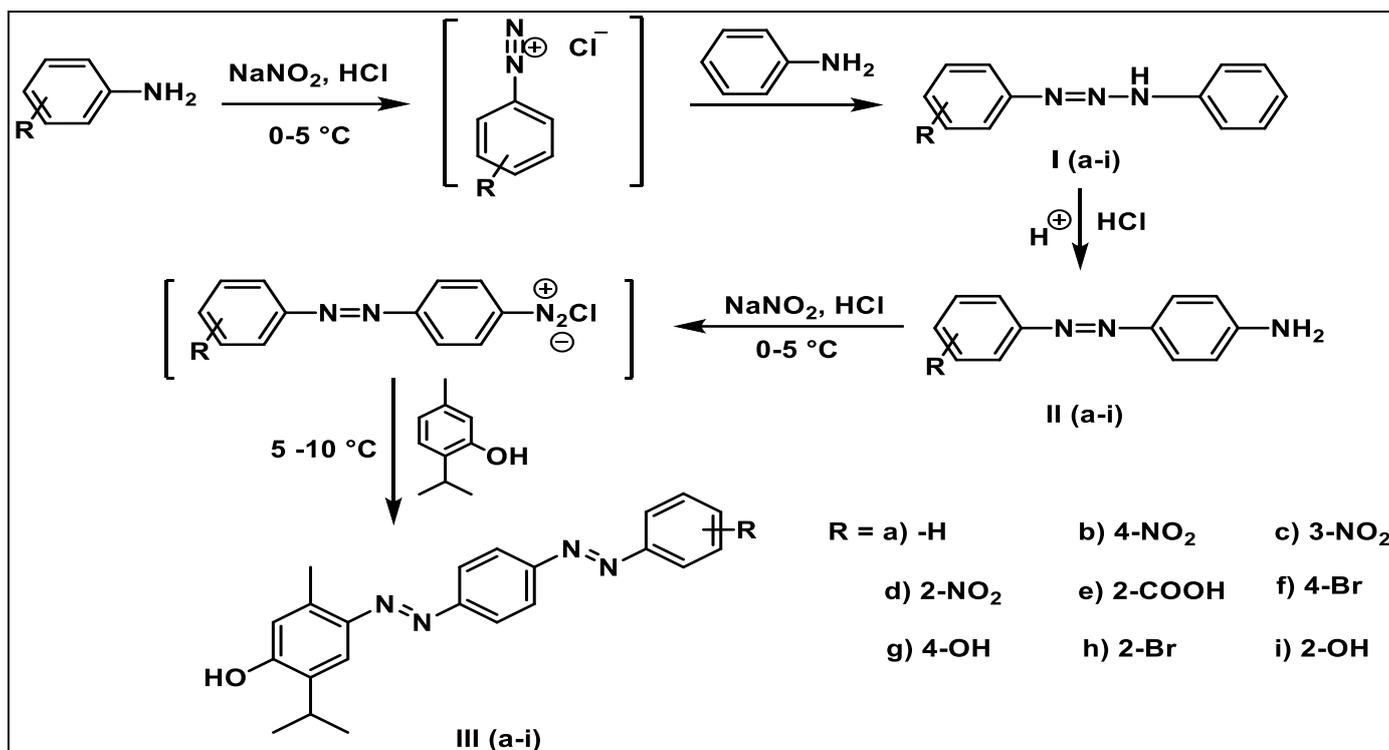
Koshti et al [73] synthesized azo compounds containing thymol moiety to study medicinal properties of thymol [Scheme 3] and characterized them by <sup>1</sup>HNMR.



Scheme 3: Azo Compounds Containing Thymol Moiety.

Piste et al. [74] used a straightforward diazotization and coupling approach to synthesize and demonstrate the antibacterial activity of p-amino azo benzene with thymol moiety from various substituted aromatic amines [Scheme

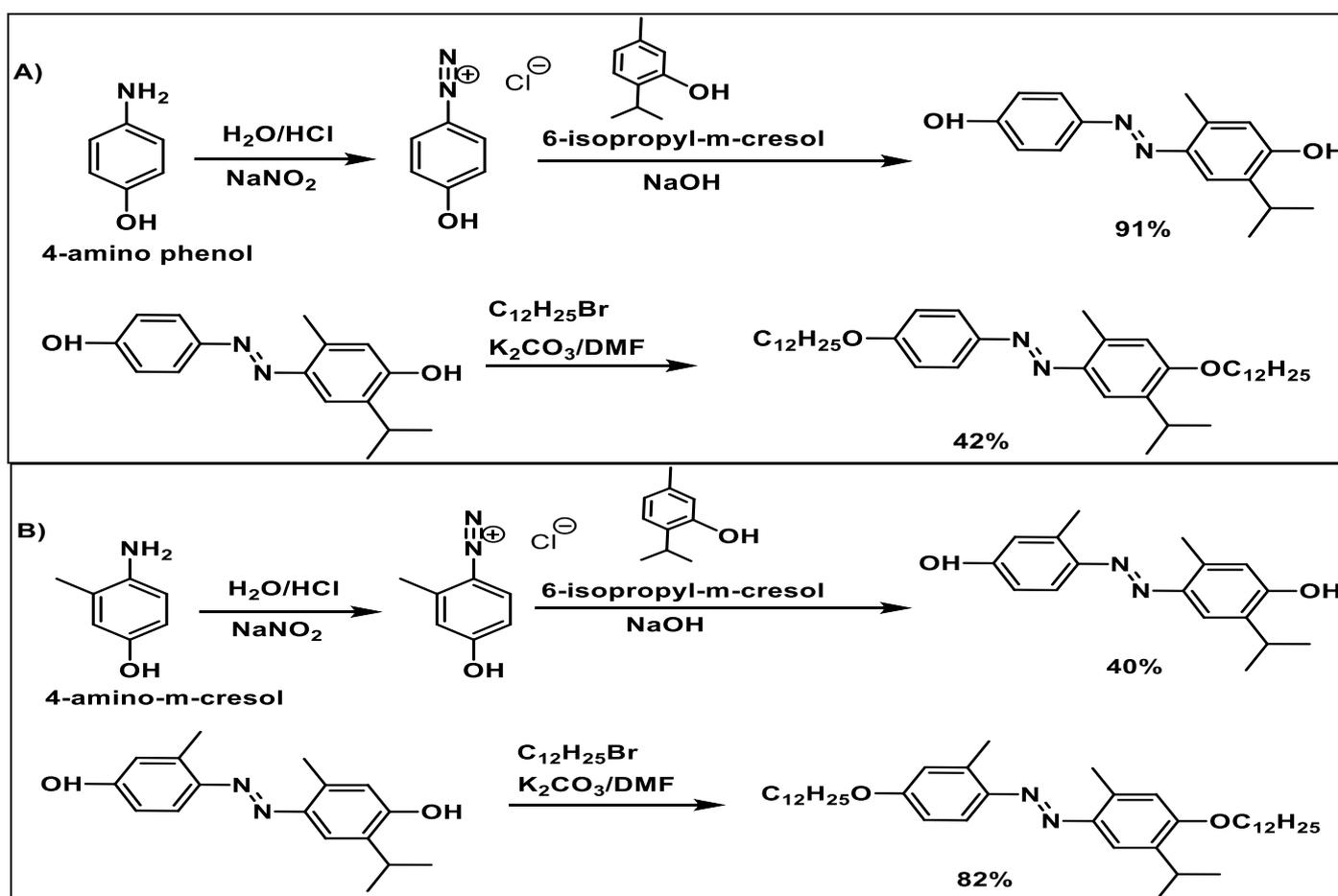
4]. In vitro tests of the synthesized compounds' antibacterial efficacy against *B. subtilis*, *S. aureus*, and *E. coli* revealed that some of them exhibited strong biological activity.

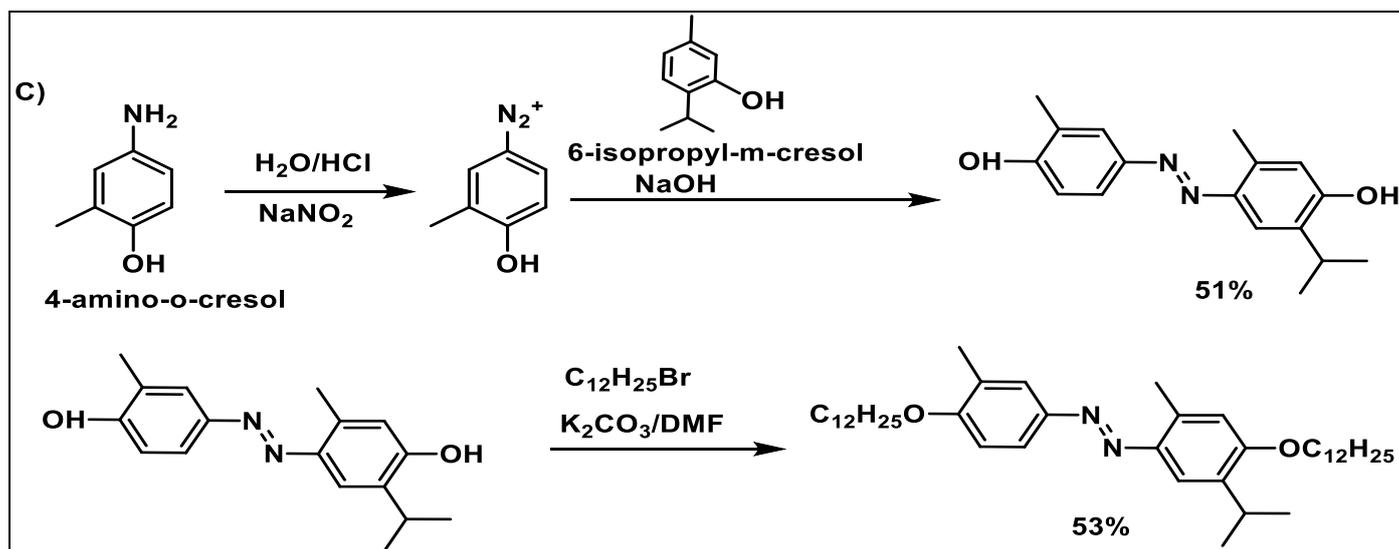


Scheme 4: p-Amino azo Benzene with Thymol moiety from different Substituted Aromatic Amines

Photosensitive azo benzene derivatives using thymol [Scheme 5] have been prepared by Norikane et al [75]

which were capable of undergoing reversible solid-liquid phase transition by cis-trans photo isomerization.

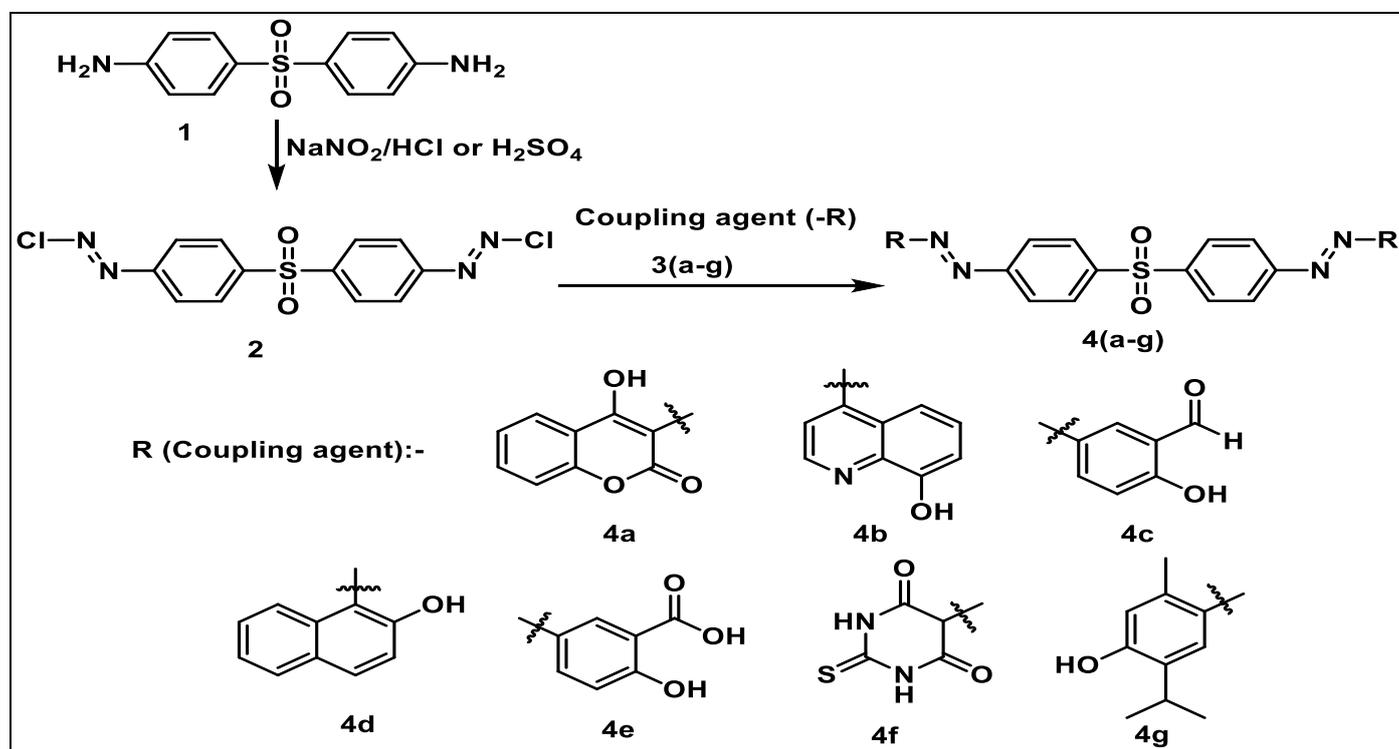




Scheme 5: Photosensitive azo Benzene Derivatives using Thymol.

By coupling diazotized dapsone with various nucleophiles under mild conditions with nitrosyl chloride present [Scheme 6], Sahoo et al. [76] created a series of seven bis diazo compounds derived from dapsone. Their in-vitro antimicrobial activity was assessed using ampicillin

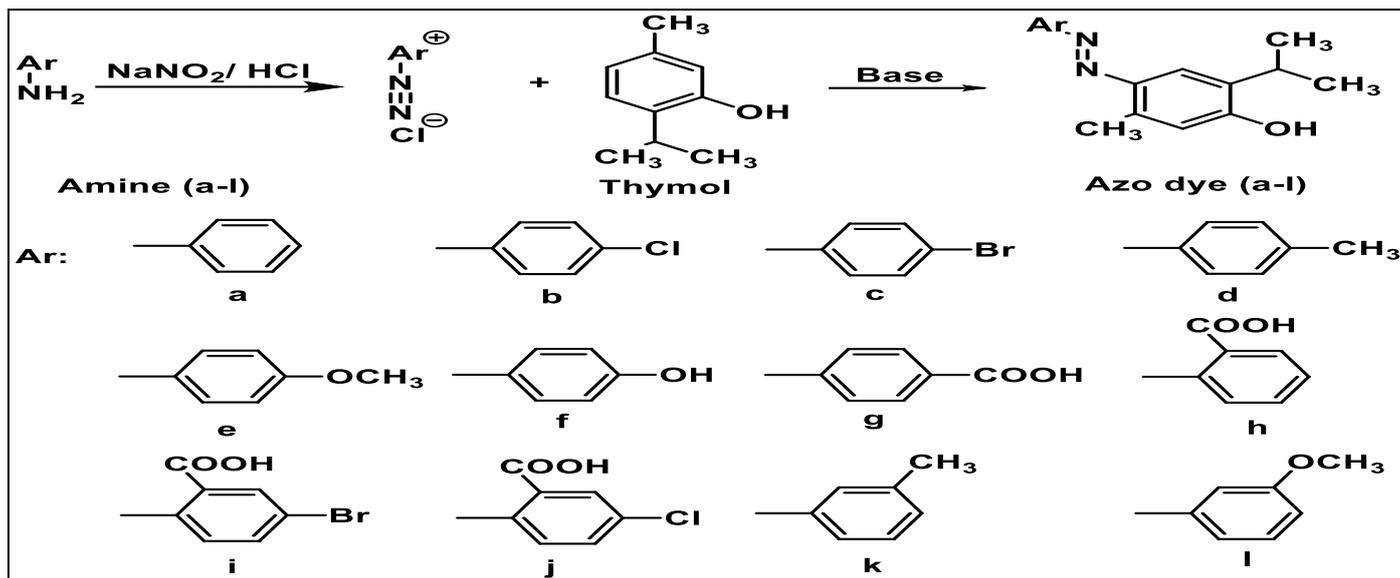
and fluconazole as reference antibiotics (RA). These nucleophiles include  $\beta$ -naphthol, 4-hydroxycoumarin, thiobarbituric acid, salicylic acid, salicylaldehyde, 8-hydroxyquinoline, and thymol.



Scheme 6: Coupling of Diazotized Dapsone with Different Nucleophiles in the Presence of Nitrosyl Chloride

In the presence of 10% sodium hydroxide, Padhy et al. coupled the diazonium salt of aniline derivatives with thymol to create a series of azoaryl substituted derivatives [Scheme 7] [77]. The agar-well diffusion method was used to assess antimicrobial activity in vitro against pathogenic bacteria that produce ESBL, antifungal-resistant fungi, and

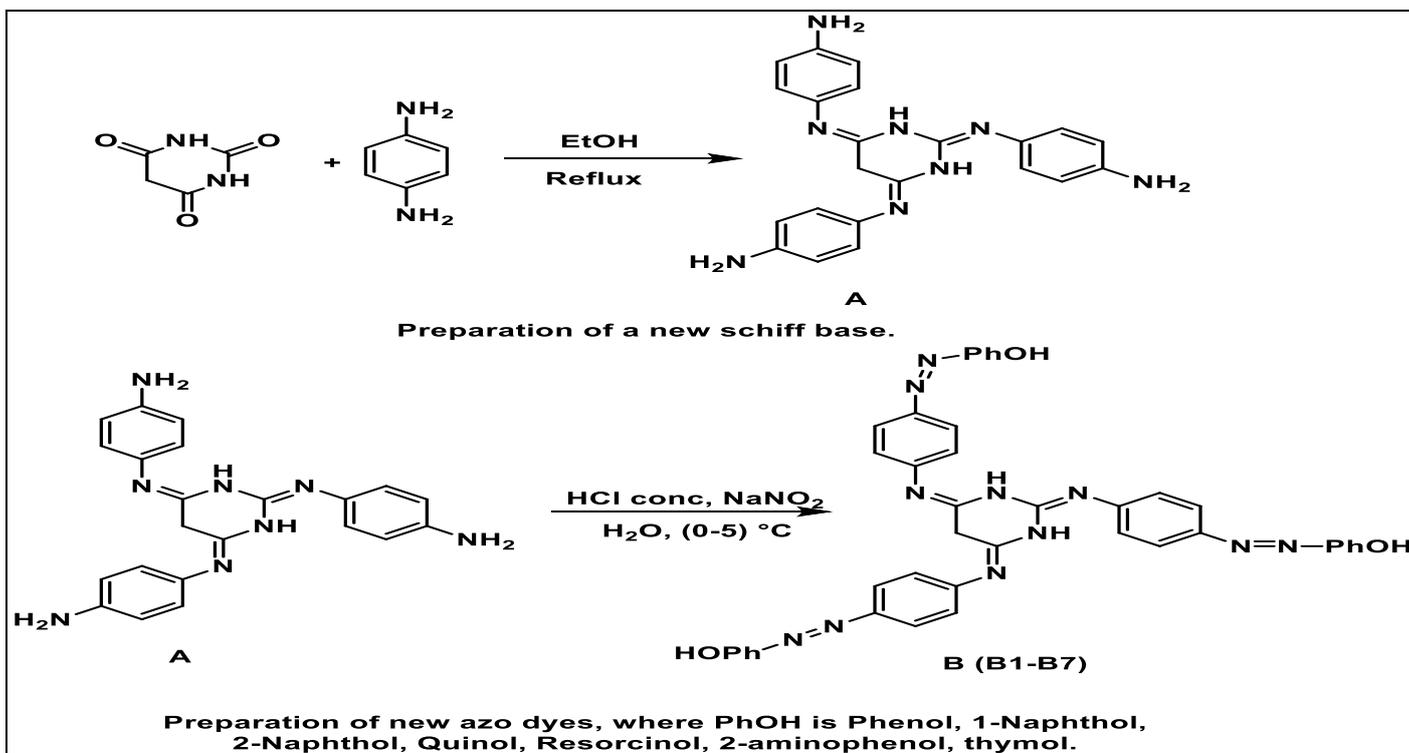
isolated MRSA. Furthermore, the synthetic derivatives' drug-likeness characteristics were assessed using bioinformatic methods like Lipinski rules of five, PASS prediction, and molecular docking, in addition to LD50 values and toxicological nature determination.



Scheme 7: Azoaryl Substituted Derivatives of Diazonium Salt of Aniline Derivatives with Thymol

The formation of seven novel azo-barbiturate dyes from Schiff bases and repellency bases by the double-reaction of repellent bases with various phenolic compounds is part of the research conducted by Kazim et al. [78]. The Azuna reaction, one of the key organic reactions, was used to create the novel azotocins with several phenolic derivatives using different phenols like

phenol, 1-naphthol, 2-naphthol, quinol, resorcinol, 2-aminophenol, and thymol [Scheme 8] and characterized by the classical and modern techniques. It is expected that the derived dyes could have industrial applications as dyes, various medicines, cosmetics, colored reagents, and analytical chemistry guides.



Scheme 8: Azo-Barbiturate Dyes Derived from Schiff Bases

In order to create 5-phenyl-1,3,4-thiadiazol-2-amine diazoamino derivatives [Scheme 9], Sivakumar and Geetha synthesized novel 1,3,4-thiadiazoles derivatives [79] by reacting different aromatic acids with thiosemicarbazide.

These derivatives were then tested for anti-tubercular activity using H37 RV strains and anti-cancer activity against lung cancer cell lines.

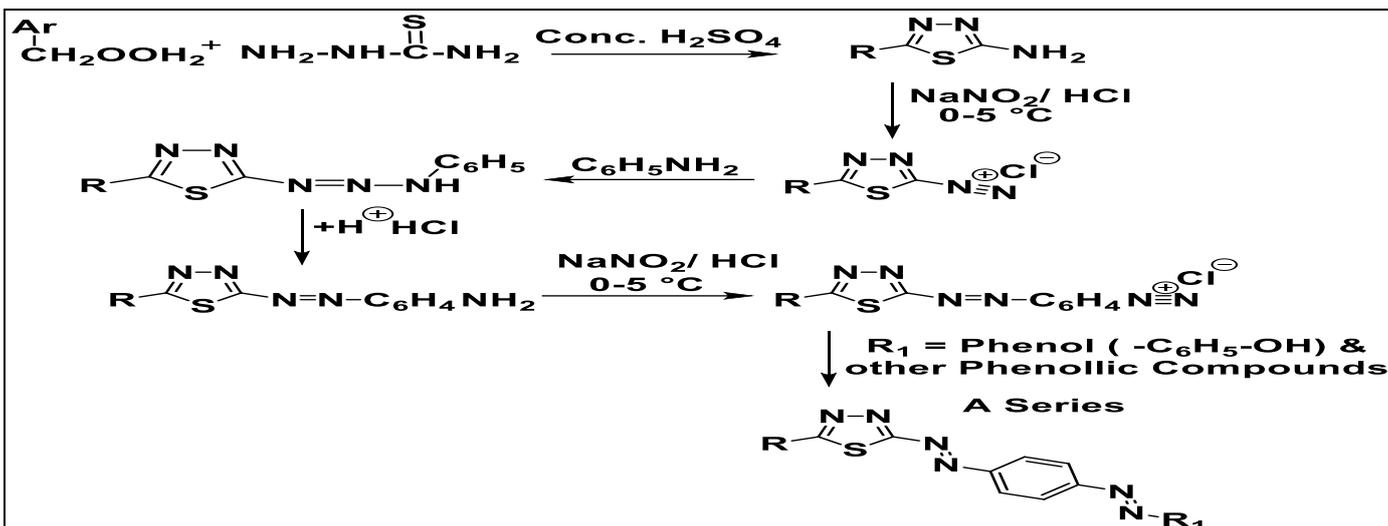


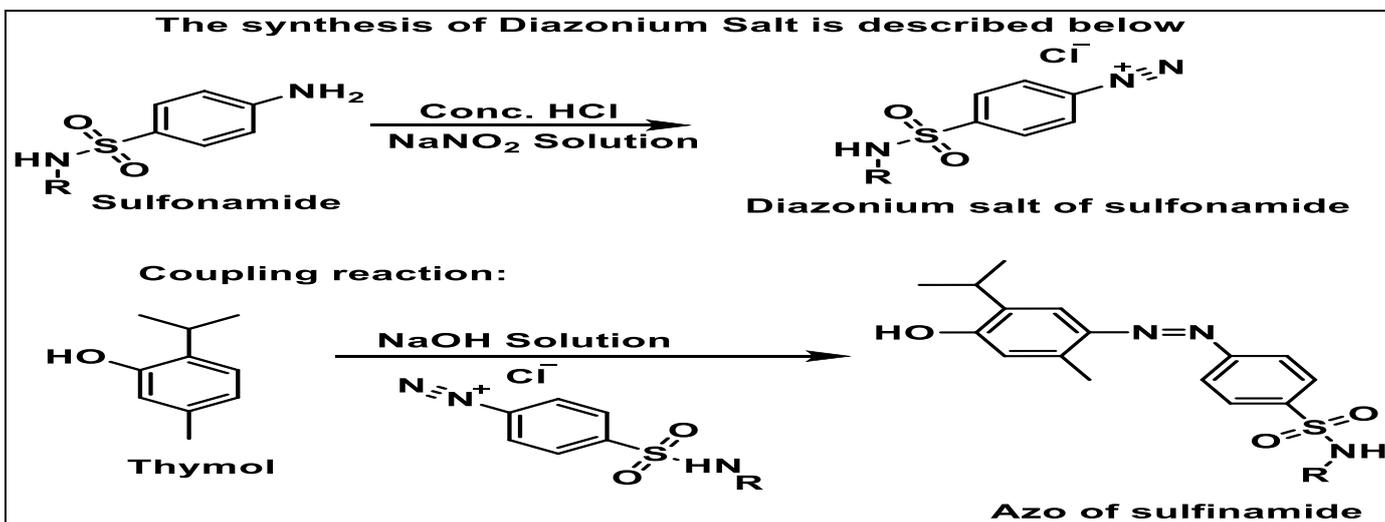
Table-1: 1,3,4-thiadiazole diazo derivatives of a series

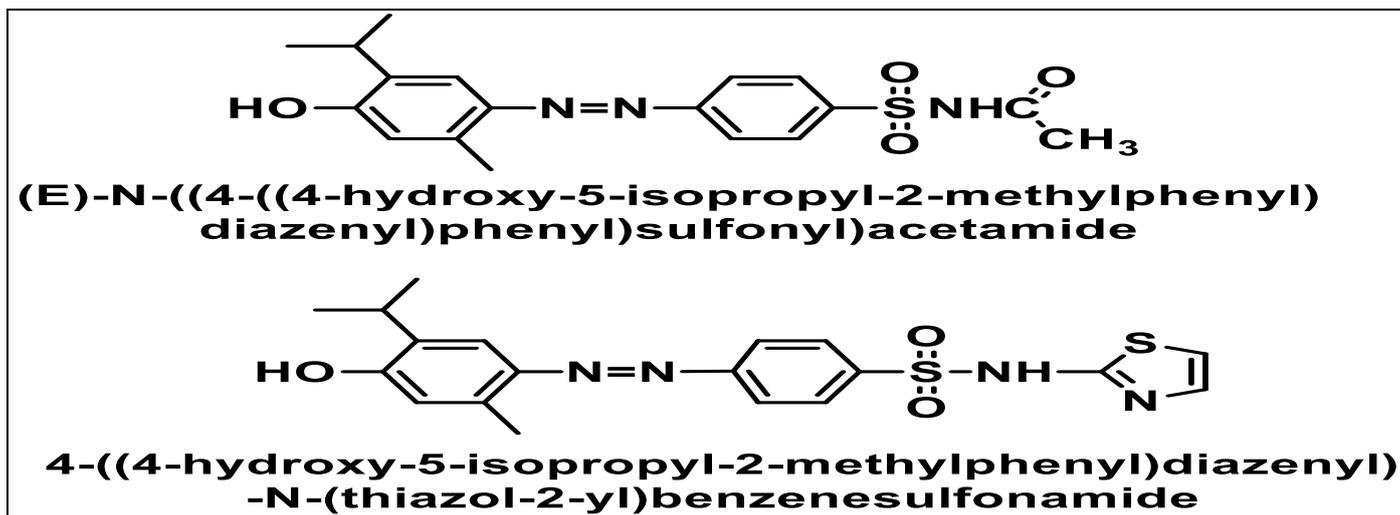
COMPOUND CODE	R	R1	
A Series with phenyl group (Benzoic acid)	Phenyl (C <sub>6</sub> H <sub>5</sub> -)	A1	4-Hydroxy Phenyl
		A3	4-Hydroxy-3-Methyl Phenyl
		A6	4-Hydroxy-2-Methyl Phenyl
		A7	3,5-Dimethyl-4-Hydroxy Phenyl
		A9	3-Isopropyl-4-Hydroxy-6-Methyl Phenyl
		A10	1-Naphthyl Phenyl
		A12	2-Naphthyl Phenyl
		A13	3-Isopropyl-4-Hydroxy-6-Methyl Cyclohexyl
		A14	2-Hydroxy-3-Methoxy-5-Vinyl Phenyl
		A15	3-Acetyl Phenyl
A series with p – Hydroxy Phenyl Group (p – Hydroxy Benzoic Acid)	p-Hydroxy Phenyl	A17	4-Hydroxy Phenyl
		A20	4-Hydroxy-3-Methyl Phenyl
		A23	4-Hydroxy-2 Methyl Phenyl
		A30	3-Isopropyl-4-Hydroxy-6-Methyl Phenyl
		A33	1-Naphthyl Phenyl

Scheme 9. 1,3,4-Thiadiazole Diazo Derivatives of Different Phenol.

With the aim to overcome the limitations of the sulphonamide antibiotics Koshti et al synthesized new azo compounds [Scheme 10] of thymol as prodrugs for sulfonamides and studied drug release by inoculating them with the bacteria *Pseudomonas aeruginosa*, which releases the parent chemical by secreting the azo reductase enzyme

[80]. The combination of thymol and sulphonamides as azo compounds has provided a novel tool for treating urinary tract infections as well as colon cancer. It may also find utility in a number of other sectors, such as pharmaceutical and medical chemistry.

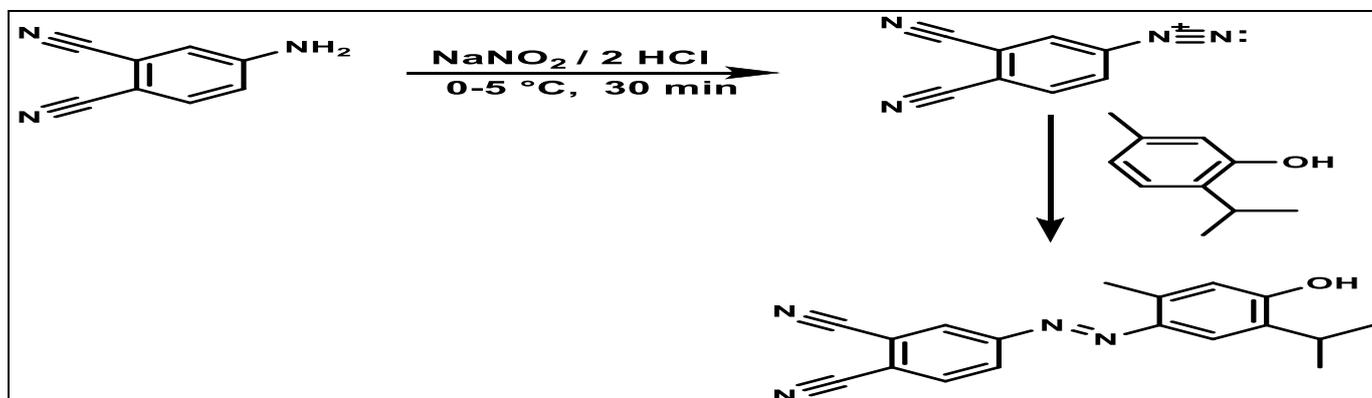




Scheme 10: Synthesis of Azo of Sulfinamide with Thymol

Rovshen [81], synthesized azo dye of thymol by making the diazonium salt of 4-amino phthalonitrile and coupled with thymol [Scheme 11]. The synthesis and characterisation of metal-free and metal phthalocyanines

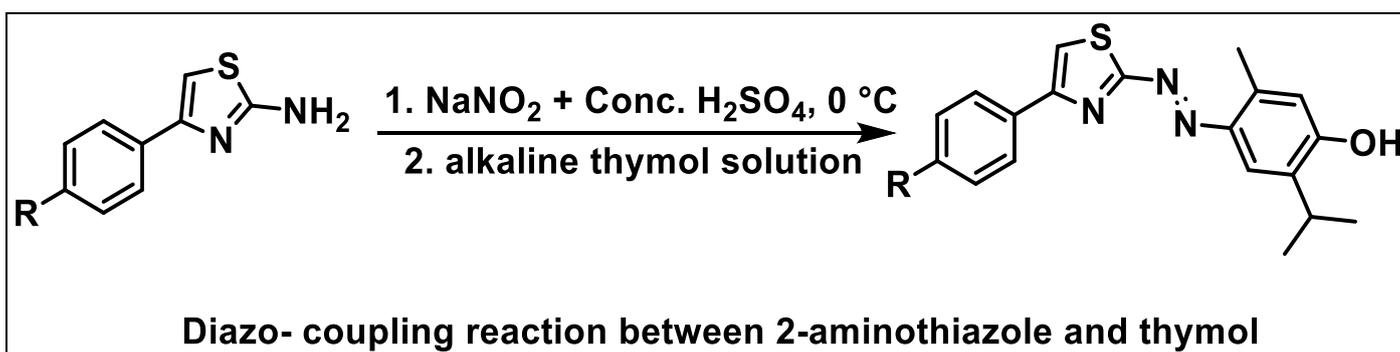
were carried out using these colours. For this synthesis, thymol moieties generated from antibacterial and antifungal terpenoid were employed.



Scheme 11: 4-Aminophthalonitrile Diazonium Salt Synthesis, Followed by an azo Coupling Reaction between the Diazonium Salt and Thymol

Sarode et al. [82] created an azo dye using the diazo-coupling reaction [Scheme 12] between 2-aminothiazole and thymol; additionally, DFT calculations were used to validate the structures of the generated azo dyes. Additionally, specific heat capacity ( $C_p$ ) as a function of temperature for the produced azo dyes has been neatly calculated using thermal profiles (TGA-DSC). DSC analysis has been used to understand how electronic factors

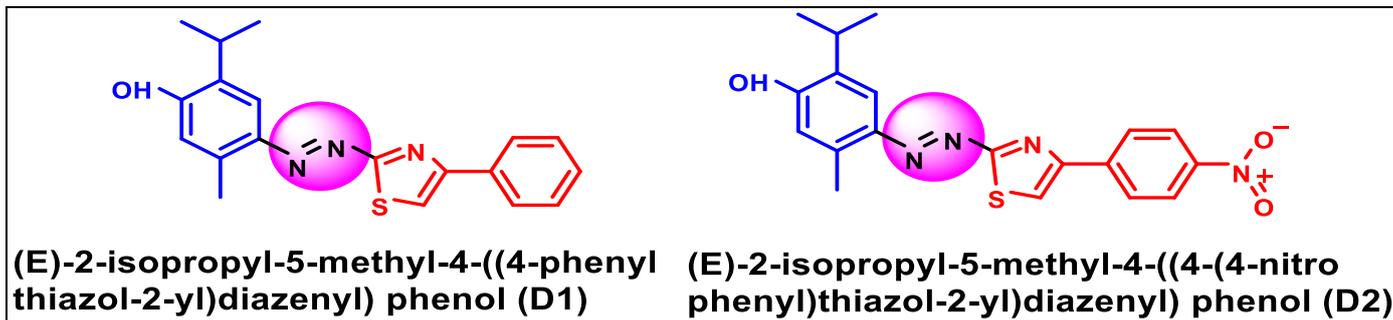
affect the melting temperature of the related azo dyes. For the first time, specific heat capacity statistics for the synthesized dyes as a function of temperature have been published. With the aid of thermal analysis, the electronic effects are used to determine the melting behaviour of the produced azo dyes. For scientists working on chemical modelling and docking studies, the specific heat capacity data can be useful.



Scheme 12: Formation of Azo Dye through a Diazo-Coupling Reaction between thymol and 2-Aminothiazole.

The azo dye (E)-2-isopropyl-5-methyl-4-((4-(4-nitrophenyl)thiazol-2-yl) diazenyl) phenol (D2) [Scheme 13] was produced by Jadhav et al. [83] and anchored to one-dimensional CdS nanowires (NWs) in thin film form with a high surface area. Through the creation of a

Cd(OH)<sub>2</sub> template and subsequent ion exchange conversion to CdS, CdS nanowires have been produced. In comparison to naked CdS (0.03%), the CdS-Azo dye-based solar cell's light-electricity conversion efficiency has demonstrated a stunning 5-fold increase (0.159%).

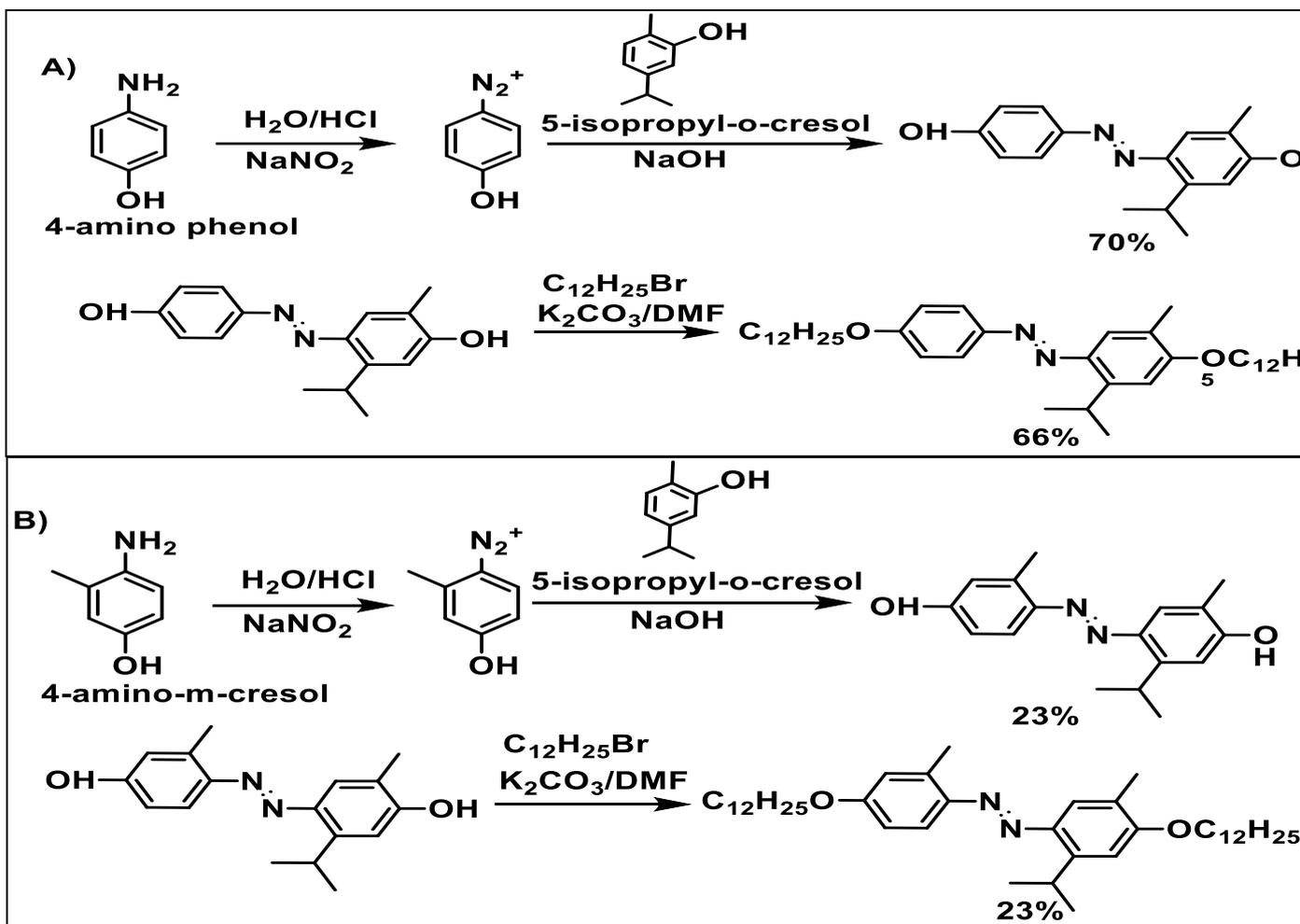


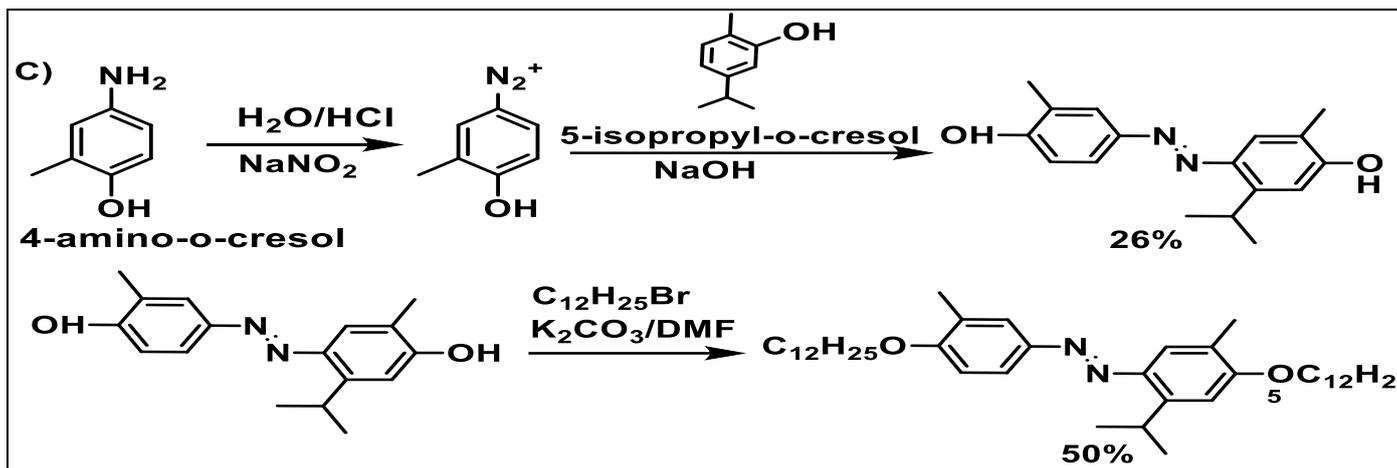
Scheme 13: Chemical Structures of the Prepared Dye (D1) and Dye (D2) Molecules.

### B. Carvacrol based Azo Dyes Synthesis and Applications

Norikane et al [75] prepared photosensitive azobenzene derivatives capable of reversible solid-liquid phase transition by cis-trans photo isomerization using carvacrol. 4-aminophenol diazonium salt is coupled with

carvacrol in basic conditions at room temperature by stirring for 16 hrs. The resulting dye is condensed with 1-bromo dodecane in presence of potassium carbonate at 80 °C [Scheme 14].

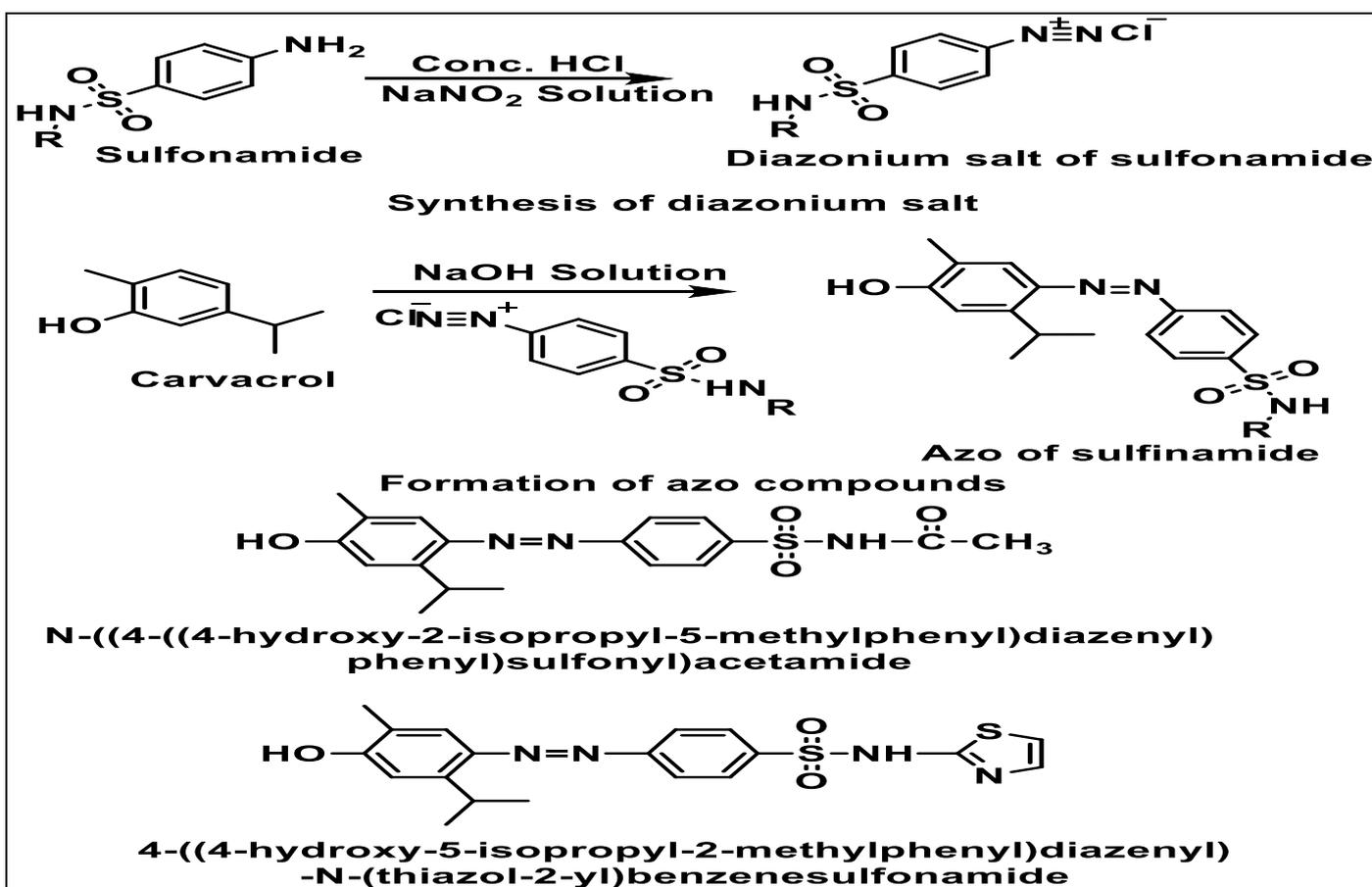




Scheme 14: Photosensitive azo benzene derivatives using Carvacrol.

Antimicrobial, antitumor, antimutagenic, antigenotoxic, analgesic, antispasmodic, anti-inflammatory, angiogenic, antiparasitic, antiplatelet, ache inhibitory, antielastase, insecticidal, antihepatotoxic, and hepatoprotective properties are among the remarkable biological activity exhibited by carvacrol and its derivatives. Therefore, Koshti et al. [84] investigated the prodrug approach study, particularly for antibacterial drugs that target the colon. Sulfonamides were chosen as an antibacterial agent for treatments that target the colon. The diazotization technique and coupling with carvacrol were

used to create the prodrugs of sulfacetamide and sulfathiazole [Scheme 15]. In vitro enzyme degradation employing the azoreductase enzyme, which is secreted by the pseudomonas aeruginosa bacteria, was used to conduct the drug release research of the parent molecule and the parent drug release was confirmed by the new spectral technique HPTLC. The combined formulation of sulfonamide and carvacrol as azo compounds can be developed as the new tool for colon targeting agents, urinary tract disorders, and for varieties of applications in pharmaceutical and medicinal fields.

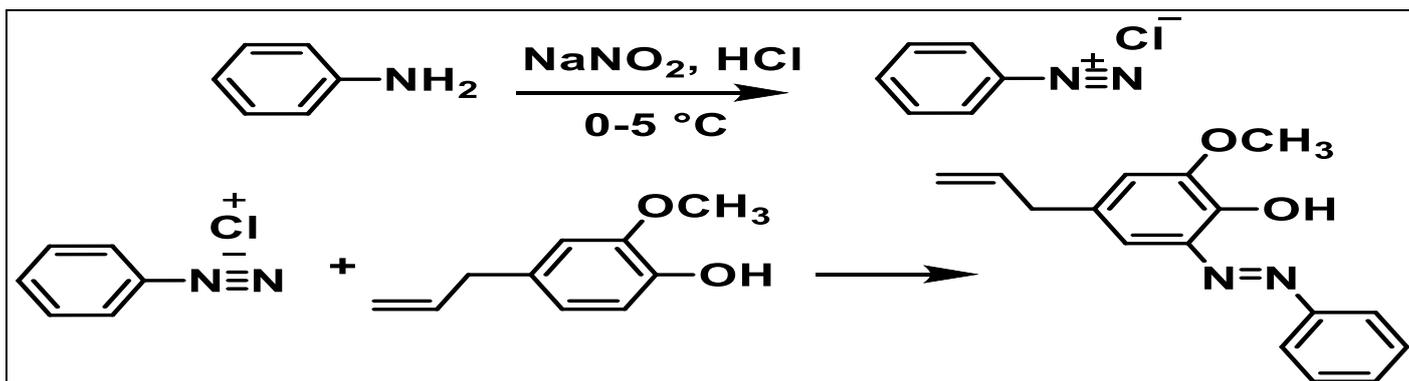


Scheme 15: Synthesis of Azo Dye of Sulfinamide with Carvacrol

### C. Eugenol based azo Dyes Synthesis and Applications

The syntheses and applications of Eugenol azo dyes are shortly summarized in the following segment. Islik et al [85] prepared 5-allyl-2-hydroxy-3-methoxyazobenzene by the reaction of 4-allyl-2-methoxyphenol and benzene

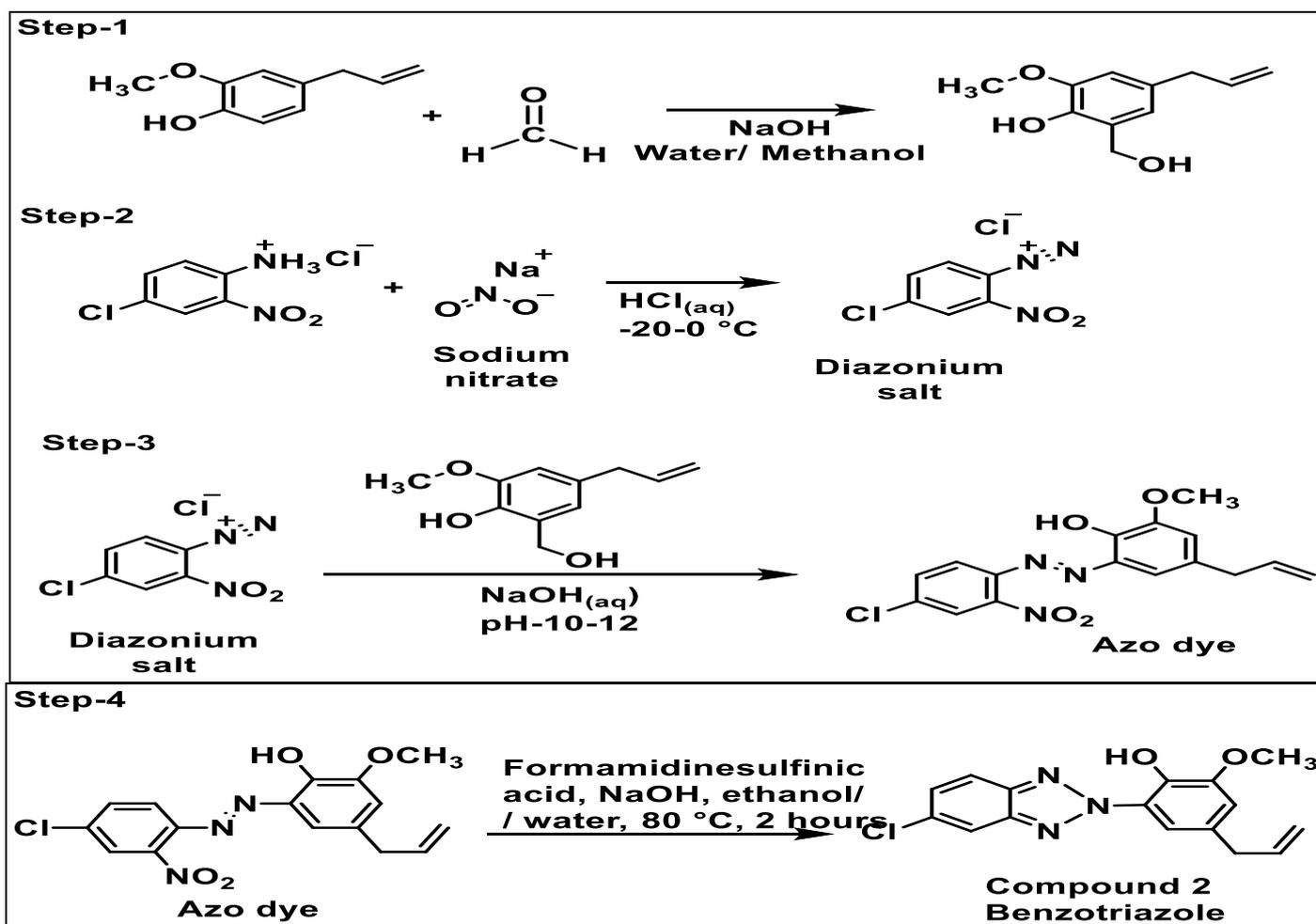
diazonium chloride [Scheme 16] and crystallized from Dimethylsulphoxide to yield pure dye crystal to its crystallographic study.

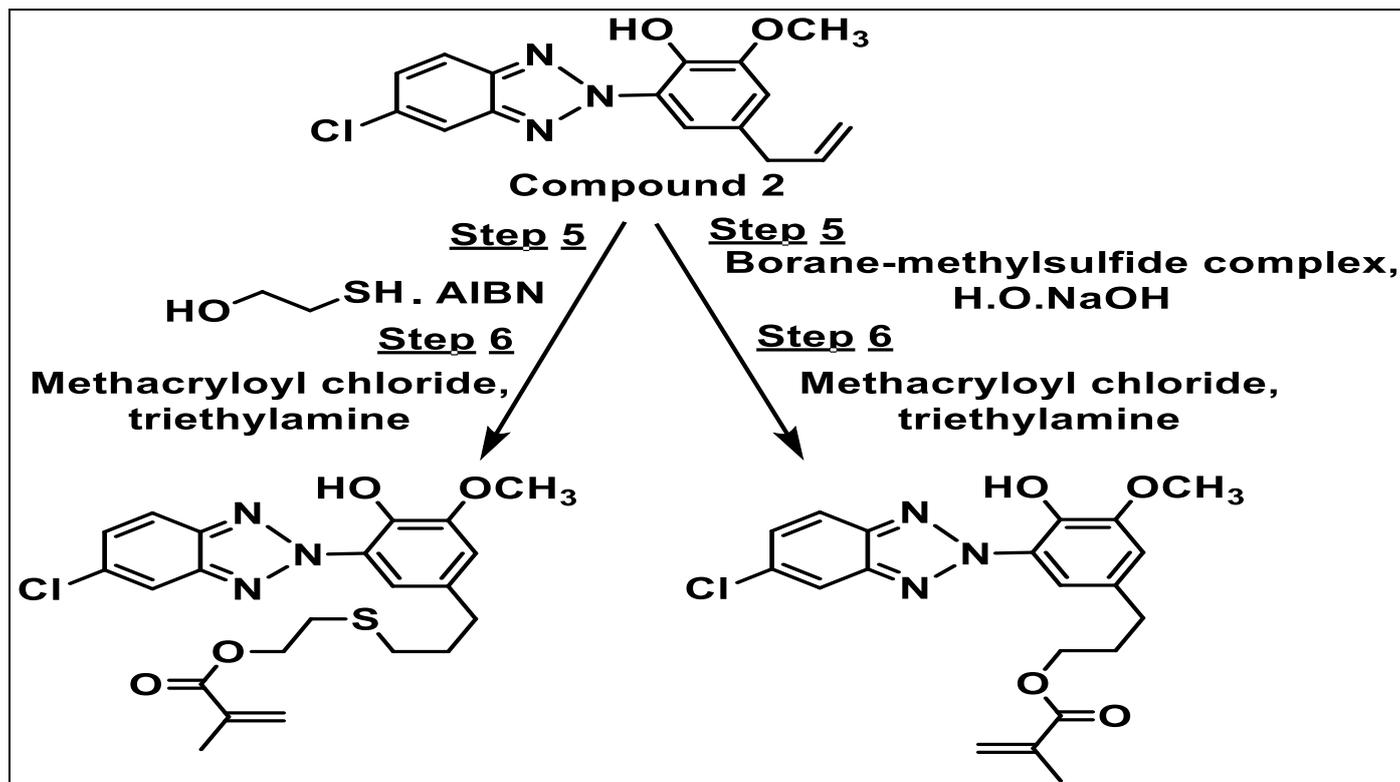


Scheme 16: 5-allyl-2-Hydroxy-3-Methoxyazobenzene from 4-allyl-2-Methoxyphenol and Benzene Diazonium Chloride.

Benzotriazole light-absorbing monomers [Scheme 17] are disclosed by Laredo et al [86] which absorb both ultraviolet light and a portion of visible light ('UV/Vis absorbers'). It is acceptable to utilize these absorbers in contact lenses and other ophthalmic lenses. In implantable lenses, such as intraocular lenses (IOLS), are especially

helpful. In addition to higher energy UVA photons between 400 and 320 nm, UVB rays between 320 and 280 nm, and UVC rays below 280 nm, the developed compounds also absorb light wavelengths between 400 and 450 nm. Because of their reactive groups, the absorbers can be covalently attached to the materials of the ocular lenses.

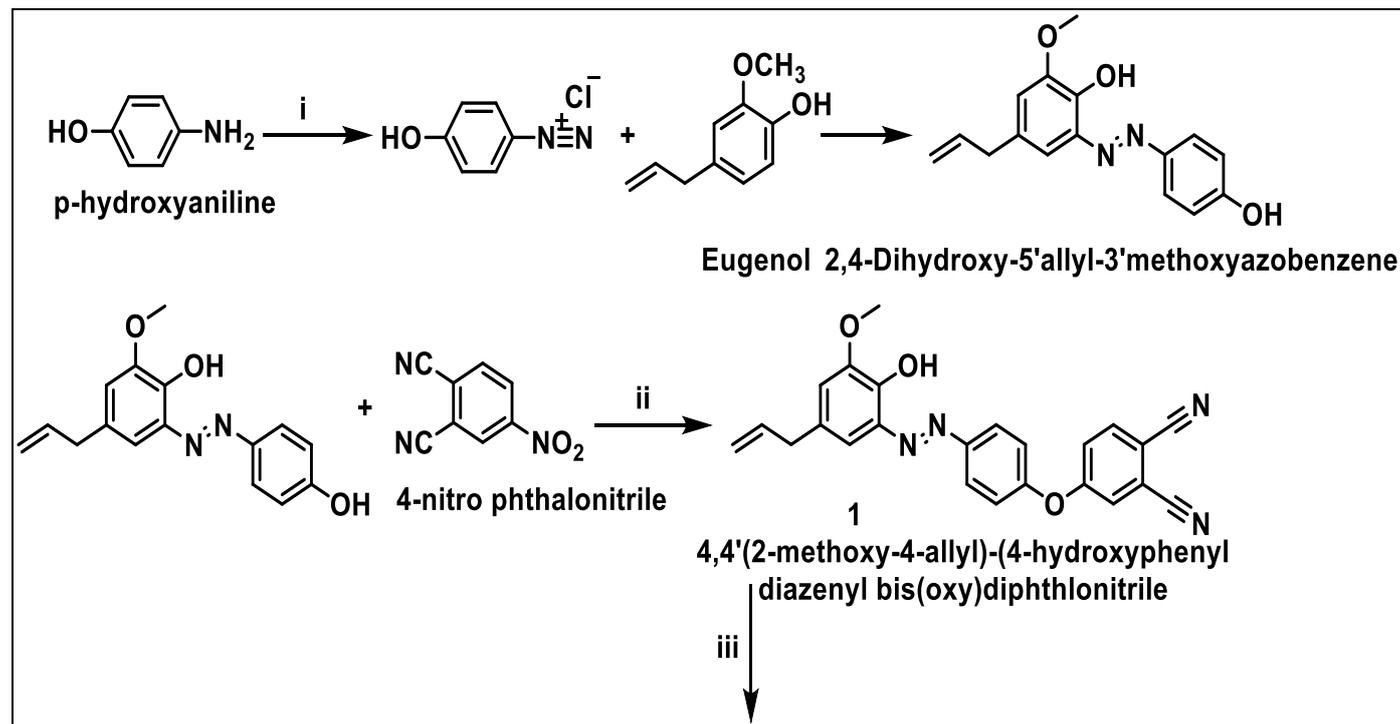


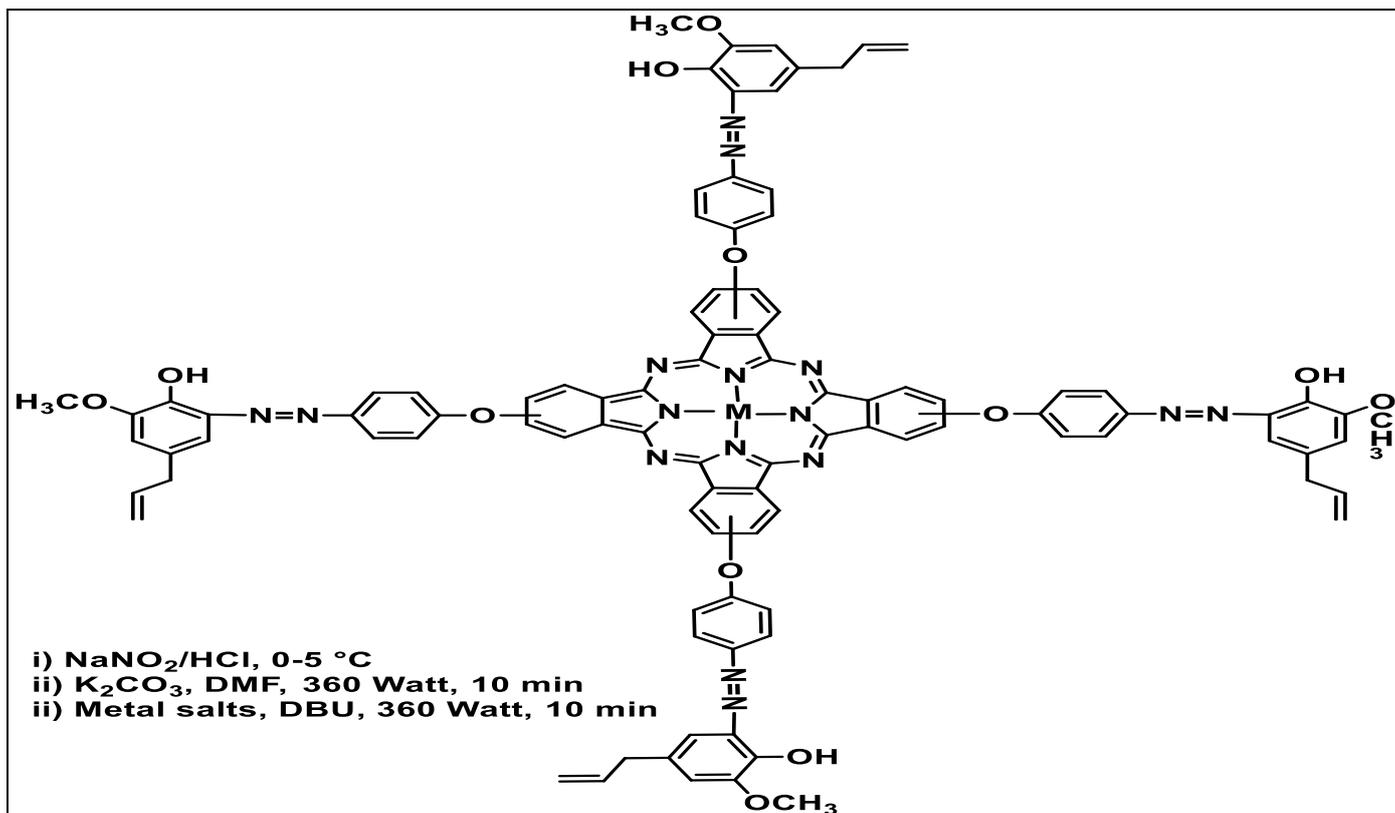


Scheme 17: Synthesis of Benzotriazole Light-Absorbing Monomers from azo dye of Eugenol and their Further Modification

The new metal phthalocyanines (Co, Ni, Cu, Zn) were substituted with an azo compound that contained a eugenol moiety [Scheme 18], and Kantar et al. [87] employed a microwave-assisted synthesis method of eugenol azo dye. 4-nitro 1, 2-dicyanobenzene is utilized in base-catalysed nucleophilic aromatic displacement to create

monosubstituted phthalonitrile derivatives. First, p-hydroxyaniline was treated with eugenol (4-allyl-2-methoxyphenol) to create 4,2'-dihydroxy-5'-allyl-3'-methoxyazobenzene, which is phthalocyanines substituted with an azo compound containing a eugenol moiety.



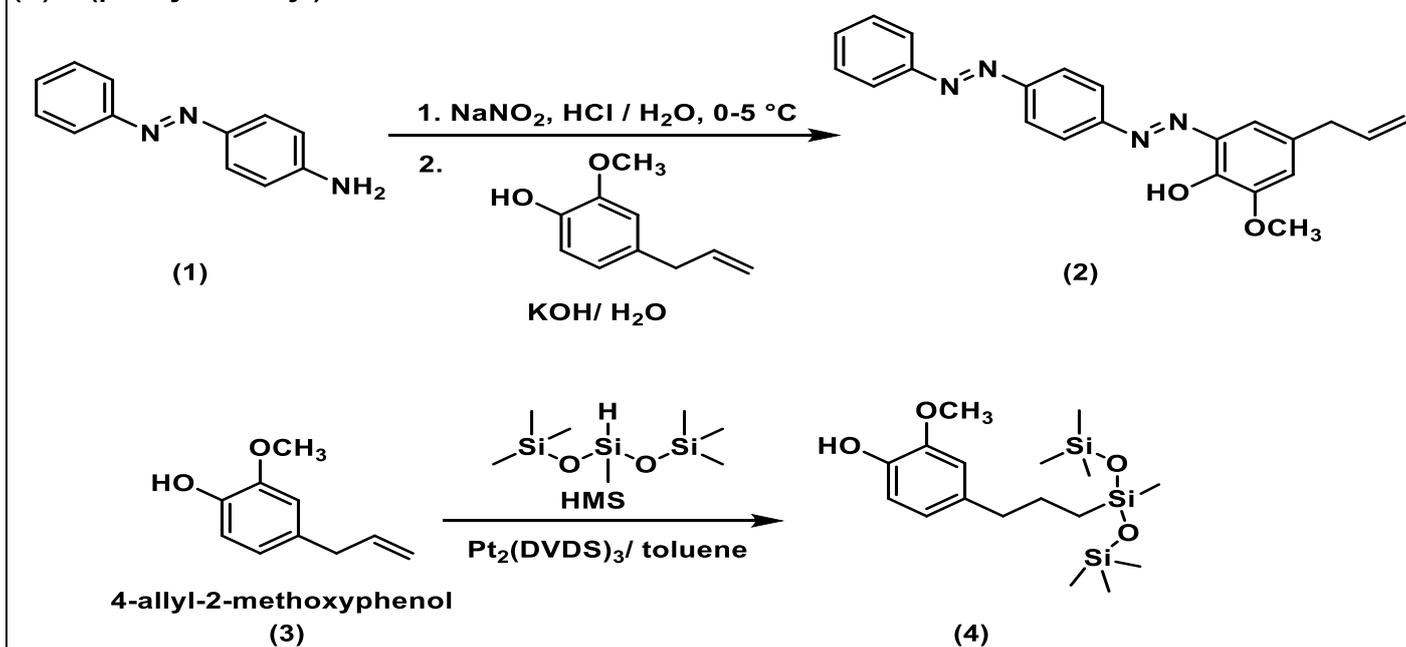


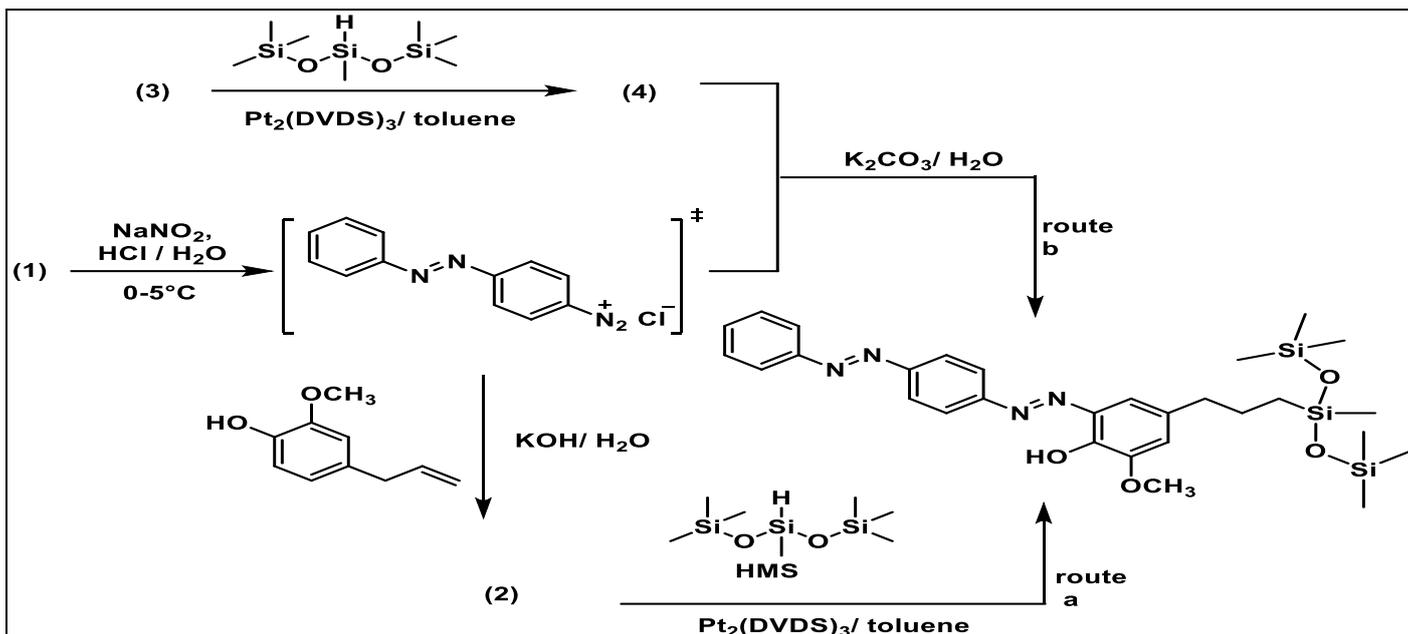
Scheme 18: New Metal Phthalocyanines (Co, Ni, Cu, and Zn) Substituted with azo Compound Containing Eugenol Moiety.

Using hydrosilylation and azo coupling processes as an example of the model heptamethyltrisiloxane [Scheme 19], Drozdove et al. [88] demonstrated two straightforward techniques for introducing an azo dye into a siloxane matrix. The siloxane derivative has been discovered to crystallize when the bisazoeugenol fragment is added to the

heptamethyltrisiloxane molecule. Polarization optical microscopy (POM), differential scanning calorimetry (DSC), and nuclear magnetic resonance (NMR) spectroscopy techniques were used to investigate the produced chemical.

#### (E)-4-(phenyldiazenyl)aniline

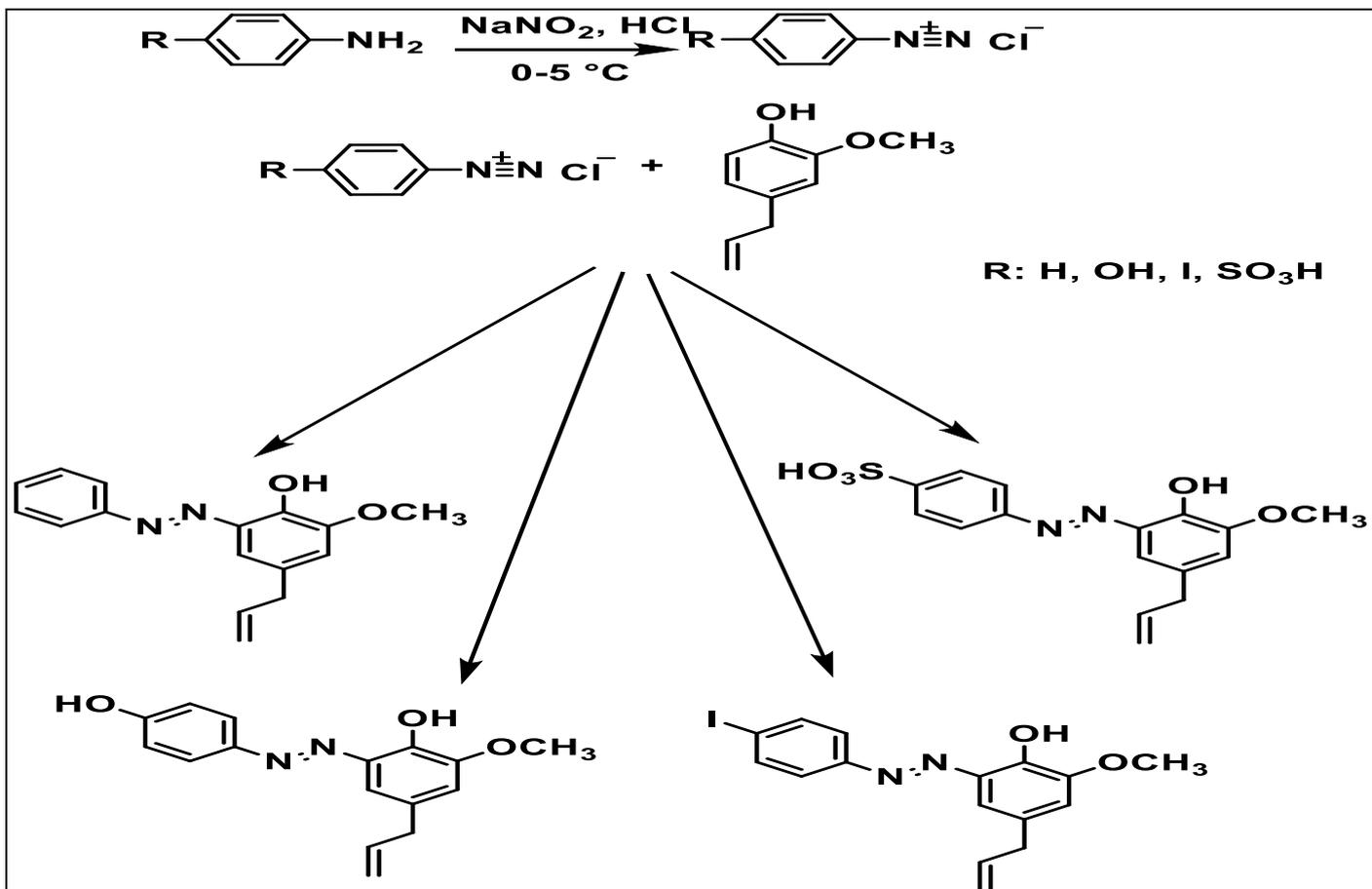




Scheme 19: Hydrosilylation and azo Coupling Procedures are used to Introduce an azo dye into a siloxane matrix.

By coupling reactions of various amines with eugenol [Scheme 20], Kantar et al. [89] created azo dyes that contained eugenol. Three in vitro tests, including the 2,2-diphenyl-1-picrylhydrazyl (DPPH) radical scavenging assay, the 2,2'-azinobis (3-ethylbenzothiazoline-6-sulfonic

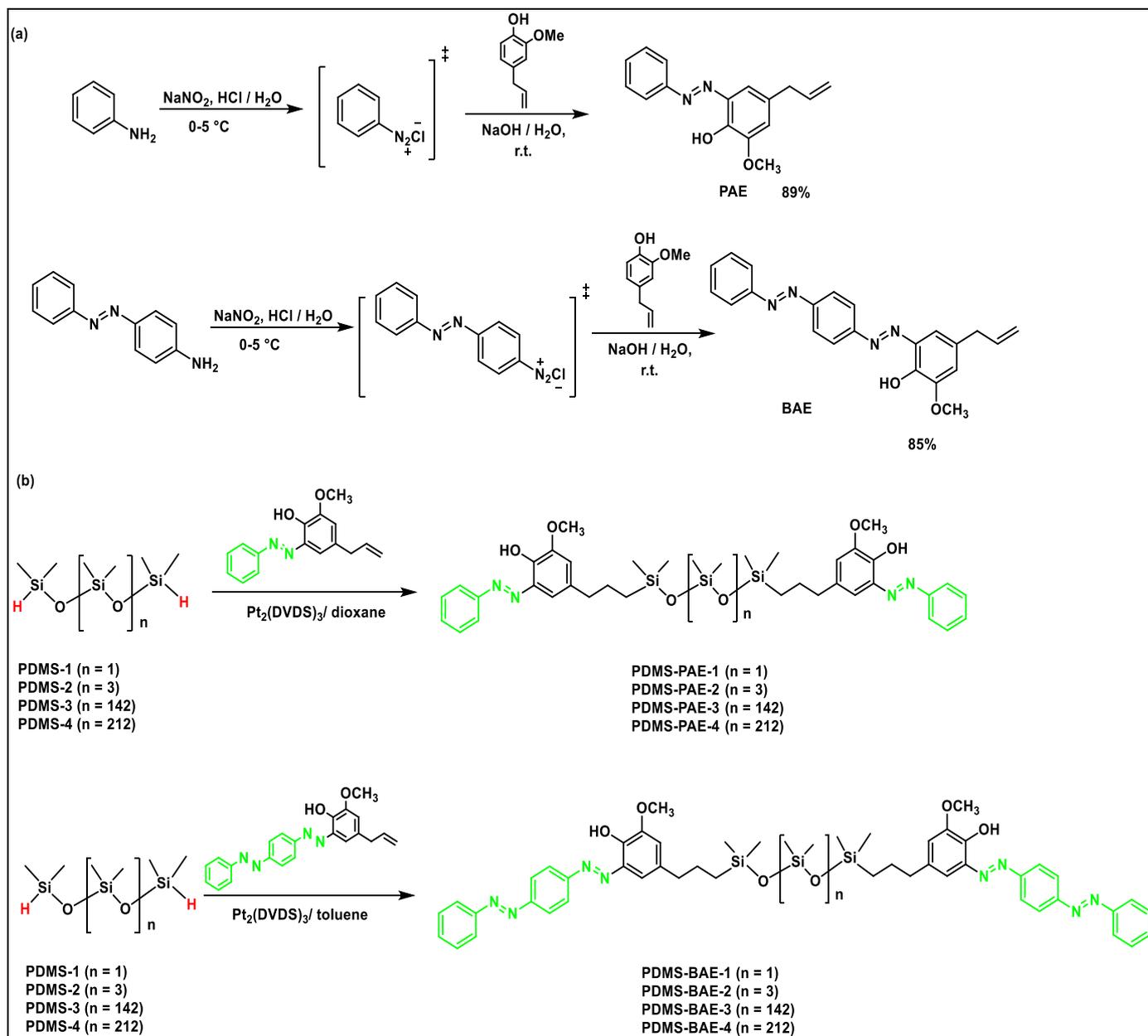
acid) (ABTS) assay, and CUPric Reducing Antioxidant Capacity (CUPRAC), were used to evaluate the dyes' antioxidant qualities. All compounds have also been evaluated for inhibitory effect against urease and Helicobacter pylori growth in vitro.



Scheme 20: Azo dyes containing Eugenol by coupling reactions of different amines with eugenol.

Based on the commercially available natural phenol eugenol, Ryzhkov et al. [90] created two azo dyes: bis (phenylazo) eugenol (BAE) and phenylazoeugenol (AE). [Scheme 21]. Trisiloxane, pentasiloxane, and polysiloxane are siloxanes with varying chain lengths that react to further hydrosilylate these dyes. Through the use of NMR spectroscopy, GPC, TGA, and DSC, these changed dyes were described. Using the beginning of decomposition (Td 5%) was reported through TGA method to increase with increasing length of siloxane spacers and melting points decrease as the length of the siloxane spacer increases.

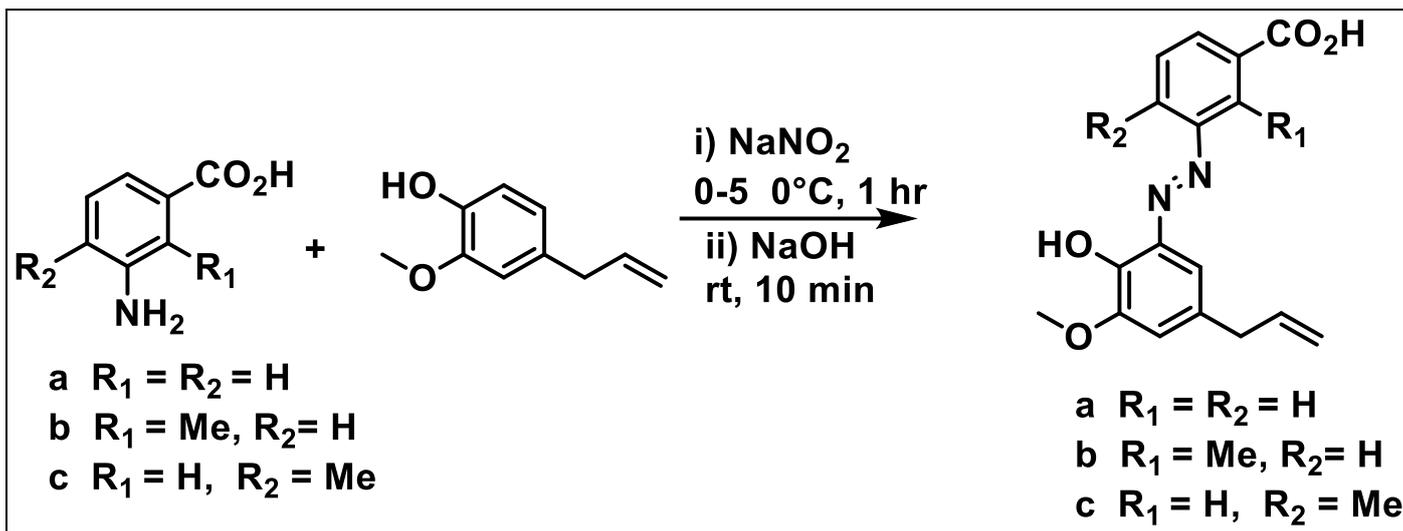
Investigations into the optical characteristics of the resultant dyes revealed that high-molecular coloured siloxanes blend well with organosilicon liquids and retain their vibrant colour even when only 1% dye is added. It is anticipated that these dyes will show great promise in creating coloured damping fluids that will enable the detection of Polymethyl siloxane (PMS) liquid leaks in machinery and the prompt resolution of the issue.



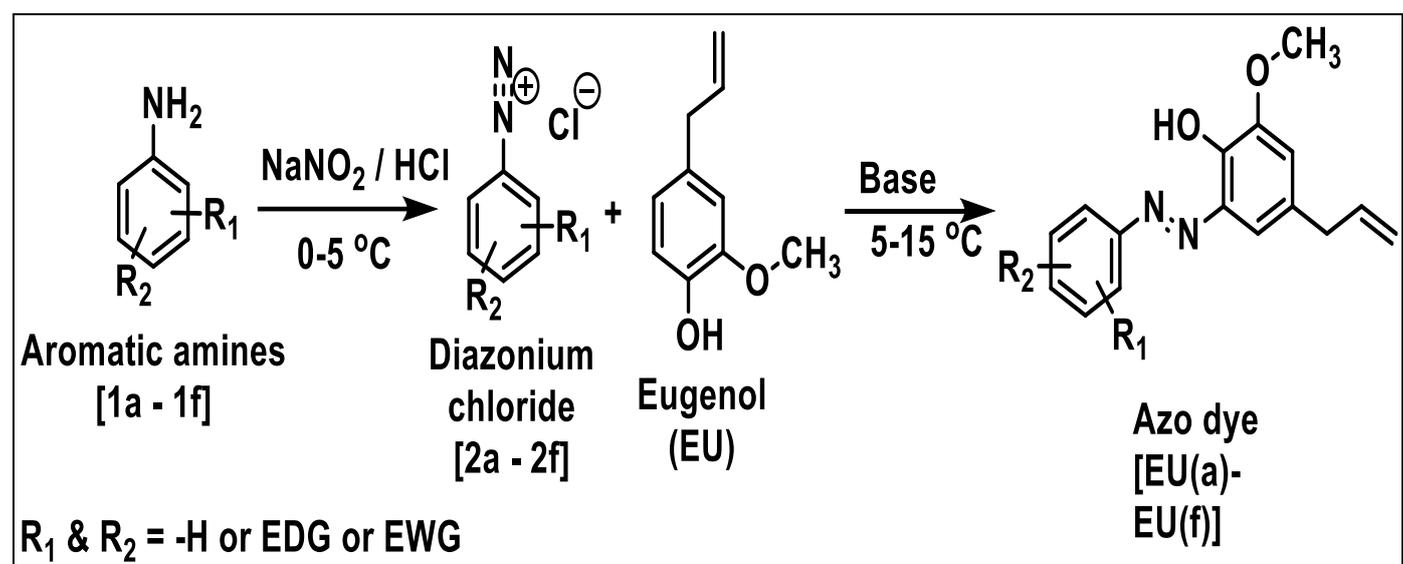
Scheme 21: Synthesis of Phenylazoeugenol (AE) and Bis (Phenylazo) Eugenol (BAE) from Commercial Eugenol.

Coelho et al [91] have synthesized azo dyes containing eugenol, by combining eugenol with diazonium salt reactions of various amines based on 3-aminobenzoic acid [Scheme 22]. In order to ascertain the colour fastness and staining under home washing circumstances, these

novel azo dyes were thoroughly characterized and applied to polyamide fabric in initial reactive dyeing tests. The findings demonstrated these dyes' potential for use in the textile industry.



Scheme 22: Coupling reactions between eugenol and diazonium salts of various amines based on 3-aminobenzoic acid



Comp. Code	$R_1$	$R_2$
EU(a)	4-NO <sub>2</sub>	H
EU(b)	H	H
EU(c)	4-CONH <sub>2</sub>	H
EU(d)	3-CH <sub>3</sub>	6-OCH <sub>3</sub>
EU(e)	4-Cl	H
EU(f)	2-CH <sub>3</sub>	H

Scheme 23. Synthesis of azo dyes from eugenol and aromatic amines.

Where **EDG** = Electron Donating Group, **EWG** = Electron Withdrawing Group

Santosh V. Dandge et al [92], synthesized five novel azo dyes from eugenol, a naturally occurring molecule, by combining it with diazo of various aromatic amines in a high yield by the conventional method. These aryl-azo-eugenol derivatives were verified their structures using mass spectroscopy, FT-IR, 1H NMR, and 13C NMR. Using the micro broth dilution method, the in vitro antibacterial activity of all recently synthesized azo dyes of eugenol was assessed. The results show that the eugenol-derived azo dyes exhibit a range of inhibitory effects on bacterial and fungal strain growth. Furthermore, all compounds were tested against the human breast cancer cell lines MDA-MB-

241 to determine their anticancer potential; of them, compound EU(d) exhibits exceptional anticancer activity.

#### IV. SUMMARY OF SYNTHESIS AND APPLICATIONS

From the above collected data from references, we segregated the Thymol, Carvacrol and Eugenol azo dyes on their different application basis and summarized in the following **Table 1** for the ease of future synthesis and its applications study.

Table 1: Segregation of Thymol, Carvacrol and Eugenol azo dyes on application basis.

No	Type of application	Thymol azo dyes	Carvacrol azo dyes	Eugenol azo dyes
1	Only Synthesis and Characterization like TGA DSC and crystallographic study	Scheme 2 [72], Scheme 3 [73], Scheme 8 [78], Scheme 12 [82]	NA	Scheme 16 [85]
2	Synthesis and biological activities, antioxidant activities	Scheme 1 [71], Scheme 4 [74], Scheme 6 [76], Scheme 7 [77]	NA	Scheme 20 [89]
3	Synthesis and new metal phthalocyanines of (Co, Ni, Cu, Zn) by microwave	Scheme 11 [81],	NA	Scheme 18 [87]
4	Synthesis and medicinal, antitubercular and anticancer activities	Scheme 9 [79], Scheme 10 [80]	Scheme 15[84],	Scheme 23 [92]
5	Synthesis and Photosensitive and solar cell application	Scheme 5 [75], Scheme 13 [83]	Scheme 14[75],	NA
6	Synthesis and used in ophthalmic lenses	NA	NA	Scheme 17 [86]
7	Synthesis and dyeing properties	NA	NA	Scheme 22 [91]
8	Synthesis and hydroxysilation	NA	NA	Scheme 19 [88], Scheme 21 [90]

## V. CONCLUSION

Natural phenolic Terpenes are comparatively less toxic, biodegradable, no residual effects as well as applications of azo dyes of natural phenol derivatives as colorant are not being tested/reported so far. As the natural substrates' thymol, carvacrol & eugenol can be used to synthesize new azo dyes by reacting with various amines as reported in literature, to utilize resulting dyes in different applications. Because of the broad range of applications of azo compounds, they are widely used and accepted. They are utilized in some chemical reactions and the manufacture of specific compounds, as well as as dyes to colour the topic. Azo dyes are colorants used to give textiles, food, cosmetics, and other things a pop of colour. Their prospective uses have been demonstrated because, in addition to colouring, they are used as indicators, catalysts, electronic and optical storage devices, and even in the pharmaceutical industry.

Because of the importance of azo compounds, we have undertaken the survey of azo dyes from naturally occurring compounds containing phenolic moiety. The available data of the structural modifications of Thymol, Carvacrol and Eugenol will be helpful in the future research to give direction for the creation and modification of novel analogs based on naturally occurring phenolic monoterpenoids so as to have improved biological activities.

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