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A Critical Review on the Impact of Azo Textile Wastewater in wet Fabric Industries on the Receiving Water Bodies and Aquatic Living

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Abstract: The textile industry may constribute to high pollution in Malaysia. The wet fabric industry generated wastewater while solid waste is generated in the dry fabric industry. Azo is a common synthetic organic dye used and has a high toxicity compared with other dyes. Azo dyes their own cleavage that most of them very dangerous to aquatic living and water bodies. In this study, the objectives are reviewing azo dye families and its impact toward aquatics living and waterbodies and also to review the existing, efficient, economical, or advanced treatments for textile wastewater. To review the papers from the previous studies, there are 4 stages for collecting data related to the objectives which are identification, screening, eligibility and include. Also, the presentation of the extensive information on azo dye wastewater constituents, impact and suitable treatment. will become an interest of a this study. Based on the study, the azo dye has to produce colourless aromatic amines (AAs) through anaerobic azo bond-breaking. AAs come in various forms, from the simplest with a single amino group to the most complex with several aromatic rings and substituents. AAs are a significant category of environmental water contaminants as most of these compounds are carcinogenic and highly toxic. Based on European Commission and REACH Regulation produce databases list of AAs prohibited, which are 1A and 1B. Many prohibited AAs should category in 1A and 1B, but there is a lack of sufficient and comprehensive data because they import textiles from Asian countries. AAs and wet fabric processes emit extremely polluting effluent that, if not properly treated, will harm water bodies and aquatic life. There are several wastewater treatment options, each with its own set of benefits and drawbacks that should be considered in terms of efficiency, effectiveness, cost, and environmental impact. Biological wastewater treatment has certain advantages over other chemical and physical treatments, according to the study.

Keywords: Azo Dye, Textile Industry, Wastewater, Aromatic Amines, Toxicity

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1. Introduction

The textile industry uses dyes as the primary material to give colour to fabrics. It also helps design fabrics with beautiful colours. Dry industries produce solid wastes, while wet fabric industries produce liquid waste [1]. In the wet fabric industry, a significant volume of water, dyes, chemicals, and other products are used to finish wet cloth manufacturing. Water is primarily used for three purposes in wet textile manufacturing: a solvent for dyes and chemicals, a medium for transferring dyes and chemicals to the cloth, and a medium for washing and rinsingA common dyeing and finishing mill consumes about 150 m^3 of water on average for every tonne of textile processing.Over 80% of industrial wastewater is discharged through textile wet processing plants [2]. The textile sector is the major contributor of wastewater since its many wet treatment activities have increased water demand. Because sizing, scouring, bleaching, mercerization, colouring and finishing are numerous operations.

In addition, 17-20% of contamination is a result of dying treatment of the textile sector, according to the World Health Organization (WHO). Approximately 80% of the azo dyes have been utilised for colouring purposes, about 10-15% of the dyes are lost in the environment through effluent without adhering to the fiber.[3]Azo dyes are among the most commonly used synthetic dyes in the textile industry and have high water solubility. In addition, t azo dye has to produce colourless aromatic amines (AAs) produced via anaerobic azo bond-breaking [4]. AAs come in various forms, from the simplest with a single amino group to the most complex with several aromatic rings and substituents. AAs are a sizable category of environmental water contaminants due to the fact that the majority of these chemicals are carcinogenic and extremely poisonous. As a result, numerous government agencies have prohibited the use of azo dyes in textiles and foods.

2. Characteristics of Azo dye

From the azo dye databases, 896 azo dyes were identified with known chemical structures. 426 azo dyes, 48% have possible one or more controlled Aromatic amines (AAs). Where the remaining 470 azo dyes, 52% are converted 397 different non-regulated AAs compile a priority list with 15 non regulated AAs mainly suspected to be genotoxic and carcinogenic

According to [3,5,6], reductive cleavage according to EN 14362-1 (DIN, 2012) has found four possibly genotoxic and carcinogenic chemicals, 2,2'-dimethylbenzidine, 4-aminophenol, 4-ethoxyaniline and aniline from 153 samples of clothes were acquired from clothing stores in Switzerland.

Azo dyes are the most frequently used dye category in the textile industry. AAs can be released from azo dyes by dermal, systemic, or bacterial biotransformation. Azo dyes are composed of one or more azo bonds joining naphthyl or phenyl radicals that are typically replaced with one or more of the following functional groups: methyl ($-CH_3$), hydroxyl (-OH), nitro ($-NO_2$), sulfonic acid sodium salt (SO_3Na), chloro (-Cl), or amino (-Cl) ($-NH_2$). Azo dyes contribute between 50% and 70% of all dyes used in the textile industry [5].

3. Aromatic Amines (AAs)

Azo dyes are the most often used synthetic textile dyes. They are produced via anaerobic azo bond-breaking, followed by creating colourless AAs. AAs come in several forms, starting with the simplest amino group and the most complicated one with several aromatic rings and substituents. AAs range from the straightest aromatic structure connected to a single amino group up to complex structures with numerous aromatic rings. AAs are a sizable category of environmental water contaminants due to the fact that the majority of these chemicals are carcinogenic and extremely poisonous. As a result, numerous government agencies have prohibited using azo dyes in textiles and foods [5].

There is no definition of the classification of carcinogenic and/or mutagenic AAs produced in Europe; nonetheless, only 22 AAs have been banned in Europe [6]. Also, Azo dyes, as listed in annex xvii of the scope of regulation, emit one of the 22 known carcinogenic AAs and are thus restricted in several nations.The European Commission has conducted a fast-track consultation on the possibility of restricting the use of more hazardous compounds Carcinogenic, Mutagenic and Reprotoxic (CMR) 1A and 1B stated in table 4.1 in textile goods and consumer clothes. Furthermore, prohibited AAs listed in Categories 1A and 1B do not follow the existing REACH regulation. Because in Europe, the majority of textile dyes sold are made in Asia, for example, China, India, Bangladesh, Indonesia. Clothing textiles sold in Europe are primarily coloured in those Asian countries before being imported. There is no such comprehensive genotoxicity dataset available for most AAs constituting azo dye cleavage products in textiles. The data requirements are based on the fact that production levels are predominantly modest in Europe, and hence data on mutagenicity are either unavailable or insufficient [6].

AAs constitute a sizable group of environmental water contaminants due to the fact that the majority of these chemicals are carcinogenic and exceedingly harmful. The below shows the unregulated aromatic amines (AAs) with their group of toxicity.

Table 1: List of the unregulated aromatic amines (AAs) according to the group A, B and C [7,8]

	Aromatic amines	**CASRN**	**Priority list**
1.	2,6-xylidine	87-62-7	A
2.	2,4-Xylidine	95-68-1	A
3.	2-Amino-5-nitrophenol	121-88-0	A
4.	2-Methoxy-4-nitroaniline	97-52-9	A
5.	2-Amino-4-nitroanisol	99-59-2	A
6.	2-Amino-4-nitrophenol	99-57-0	A
7.	2-Amino-5-nitrothiazol	121-66-4	A
8.	4-Nitroaniline	100-01-6	A
9.	2-Amino-6-methoxybenzothiazol	156-43-4	A
10.	4-Nitro-1,3-phenylenediamine	5131-58-8	A
11.	2-Amino-6-nitrobenzothiazol	6285-57-0	A
12.	3-Amino-5-nitro-2,1-benzisothiazol	14346-19-1	A
13.	3,4-Dichloroaniline	95-76-1	A, B
14.	4-Aminophenol	123-30-8	A, B
15.	2,5-Diaminotoluene	95-70-5	B
16.	4-Ethoxyaniline	156-43-4	B
17.	4-(N,N-Diethyl)-2-methyl-p-phenylenediamine	2051-79-8	B
18.	N,N-Dimethyl-1,4-phenylenediamine	99-98-9	B
19.	1,3-Phenylenediamine-4-sulfonic acid	88-63-1	B
20.	4-Aminodiphenylamine	101-54-2	B
21.	p-Phenylenediamine	106-50-3	B
22.	Sulfanilic acid	121-57-3	B
23.	1,4-Diamino-2-methoxybenzol	5307-02-8	B
24.	2-Amino-5-chloro-p-toluenesulfonic acid	88-53-9	C
25.	2-Chloroaniline	95-51-2	C
26.	p-Toluidine	106-49-0	C
27.	m-Phenylenediamine	108-45-2	C
28.	Aniline	62-53-3	C
29.	2,2' -Dimethylbenzidine	84-67-3	C

According to the table above, it can be seen that the AAs have their hazardous scoring, which are A, B, and C. Group A potentially content AAs that have carcinogenic and genotoxic, and Group B is

AAs which may cause sensitization by skin contact. In contrast, group C was detected by environment Canada, Health Canada (2012) and the Food Control Authority of the Canton Bern.

Carcinogenic, mutagenic, and reprotoxic are the effects that Aromatic Amines (AA) in azo dye create, not only because a chemical may exhibit all three types of dangers, but also due to parallels in classification and legal approach.

4. The implication of Azo dye

Most textile industries prefer to use Azo dye because the workability of the dye is proven. According to [9], azo dyes have become a widespread industrial dye due to their inexpensive cost, simplicity of manufacture, variety, colour intensity, and fastness. But they also one of the leading industries that created environmental pollution due to the careless discharging of untreated or inadequately treated wastewater into regularly used irrigation water [9]. When azo dyes encounter reductive cleavage, aromatic amines are formed, mutagenic, carcinogenic, genotoxic, and teratogenic [9]. The cleavage of azo dye can cause severe pollution since it can trigger the change of the genetic or properties of the water.

There is rising concern about wet fabric industry effluents and the possible disastrous effects of genotoxic and mutagenic on the aquatic ecosystem and humans due to contaminated water utilized for irrigation and recreational activities. For decades, various studies have concentrated on the toxicological properties and the mechanisms of azo dyes biotransformation, including some carcinogenic and mutagenesis effects. [10].

4.1 Water bodies

Table 2 shows that several researchers identified the primary characteristic of effluent wastewater according to the processes of the wet fabric industry. According to [3] The azo colouration has a long-term life effect, which is dangerous to azo colouring properties and elevates pH, BOD and COD in water bodies where it releases. When the water bodies loss amount of fresh oxygen, the toxicity parameters will be raised then it will caused respitory distress.

Table 2: The fundamental properties of textile wastewater based according to the processes [11,12]

	Processes	COD (mg/L)	BOD (mg/L)	TS (mg/L)	TSS (mg/L)	TDS (mg/L)	pH	Color (ADMI)
Carmen and Daniela (2012)	Desizing	4600–5900	1700–5200	-	-	-	-	
	Scouring	8000	100–2900	7600	-	-	10–13	694
	Bleaching	6700–13,500	100–1700	2300–14,400	-	4800–19,500	8.5–9.6	153
	Mercerising	1600	50–100	600–1900	-	4300–4600	5.5–9.5	-
	Dyeing	1100–4600	10–1800	500–14,100	-	50	5–10	1450–4750
Zhang et al. (2012)	Bleaching	528 ± 7.9	-	-	-	-	9.4	17
	Fibre scouring	-	-	-	-	-	7.3	21

	Rinsing	311 ± 2.1	-	-	-	-	8.6	7
	Soaping	578 ± 23.5	-	-	-	-	12	38
Savin and Butnaru (2008)	Burning	1512–7802	679-925	-	105–936		5–6.5	-
	Bleaching	1060–6556	80-520	-	56–147		7–12.4	-
	Dyeing	258–1970.6	70-300	-	72–956		6.3–10.7	-
	Dyeing Gauge	458–7561	230-410	-	175–325 -		6.5–12.1	-
	Dressing	825–1905	60-180	-			7–7.11	-

Table 4.2 shows that bleaching has a high value of COD and pH compared to other processes. As can be seen, good bleach requires a sufficient amount of peroxide to begin with. The bleaching rate is determined by the peroxide concentration in relation to the weight of the solution. The higher the solution concentration, the faster the bleaching; a high bleaching agent concentration contributes to a high COD value. Other than that, bleaching purpose in fabric industries is to remove colour, fat, wax for the following process and also the quality of fabric colour, so the high temperature needed to increase the degree of whiteness as well as so good fat removal at high temperatures such as 110°C [13].

Other than that, desizing/ Burning-off and scouring are the processes with a high value of BOD compared to the other processes, which are 1700-5200 mg/l,100-2900 mg/l and 679-925 mg/l. Effluent for desizing process get high range biological oxygen demand (BOD) and pH which are 300-450 ppm and 4-5 [1]. It is because to enzyme or dilute mineral, acid hydrolysis or oxidation is used to eliminate sizing chemical for the next process. According to Toprak & Anis, (2017) desizing will emission VOCs from glycol ethers, and the wastewater contaminant is BOD from sizes lubricants, biocides anti-static compound. It also has its alternative by replacing the chemical with amylases to improve the environment friendly.

Other than that, using hydrolysis or oxidation in desizing has 3 stage washing, namely, hot wash, warm rising and cold rising, is carried out with a liquid ratio of 20:1 at each stage. it has been reported that about 50% of the pollutant in textile industry originates from desizing operations [14].

Without proper treatment of azo dye effluent, it will affect water body and ecosystem due to vast amount of coloured and remain metals. It can be seen from the result where the dyeing process has a high value which is 1450–4750 ADMI.

4.2 Aquatic Living

The term "aquatic living" refers to any type or species of mammal, fish, amphibian, reptile, or other animals that live in freshwater or saltwater. Furthermore, an unusually high temperature (slightly above 50°C) was detected in textile industry effluents released in Narayanganj, Bangladesh. According to [15], the national and international permitted limit for discharged effluents is 40°C, as is the highest temperature measured in textile industry effluents globally. A wastewater temperature of 77°C is expected to significantly negatively influence the animals, plants, soils, and wetlands in the surrounding area.

CMR stands for corgenixc, mutagenic, and reproductonic, all of which have a critical effect on aquatic life's reproduction by affecting the hormone system of hormone-secreting glands that can move throughout the body and affect numerous cells, tissues, and organs. Numerous physiological functions are regulated by the endocrine system, including reproduction, metabolism, sleep, growth, the stress response, and the immune system. Water pollution developed which caused breathing distress to aquatic creatures. Inhalation of fumes or ingestion of contaminated seafood can result in a variety of adverse health effects, from dizziness and nausea to some types of cancer and central nervous system problems.

Moreover, when the dye is combined with water, the effectiveness of light penetration into the water decreases, affecting the entire aquatic ecology. Cleavage of azo dye dissolve into water and are consumed by aquatic living. Then, the aquatic living comumed by human, resulting in hypertension, sporadic disorder, cramping, and other long-term negative effects [3].

5. Treatments for Azo wastewater

Physicochemical processes (for example, coagulation/flocculation) and biodegradation are the primary treatment procedures for textile dyeing effluent [15]. The removal range of AAs which is 35% to 100% in azo dye wastewater through anaerobic-aerobic reactor system [16]. However, AAs cannot be completely removed from wastewater due to the absorption of AAs by physical and chemical processes in sediment and soil.[16]. Based on findings, Table 3 shows the treatments used in treating azo dye wastewater.

Table 3: Effectiveness of different method for azo dye treatments

Method	Finding	References
Anoxic-aerobic MBR	• The proposed technique highly removed AAs with a single aromatic circle (over 80%), while most AAs with two aromatic rings were removed moderately (35%–75%). • Achieve high COD, colour, and AAs removal rates. • MBR was ineffective except for the removal of triclosan is 99 %. • Removal of ECs is 70% to 80% • up to 97 % of arenol.	[5,17]
Wetland system, TEWS Method: Tri-phasic engineering free-floating wetland system (Macrophytes)	• The cumulative of colour removal is 76% for 3 tanks • 87% of COD removal efficiency for three Tanks varied between 190 and120 mg of COD/L, • The pH range of influent and effluent between 7 and 8 in each tank. • The overall dye removal is 76%	[17,18]
Al-Amrani et al. (2014) Method: anoxic/aerobic	• High removal of dyes and COD	[12]
Bacteria degradation	• The mixture of aerobic and anaerobic bacteria is more effective and efficient in degrading the azo dye than one single bacteria.	[3,19]

Method: Microbial enzymes	• Using microbial enzymes, the degradation of chemical bonds is faster because many enzymes attack chemical bonds at the same time in a different way.

The membrane bioreactor (MBR) is a device that combines membrane technology with biological treatment. According to Albahnasawi et al. (2020), MBR has been utilised to treat industrial wastewater in a variety of purposes. For example, high-carbon and nitrogen, colour removal and complete solid removal, which results in high-quality treated waste water for reuse are important advantages of using MBR for textile effluent treatment. In addition, treatment at higher temperatures in an MBR is incredibly efficient, which might be enhanced and applied commercially [5]

According to Albahnasawi et al. (2020), the practicality of an anoxic-aerobic MBR process for textile wastewater treatment removes a high concentration of COD, colour, and AAs. The percentage removal and fate of AAs were determined in real-world textile wastewater treatment applications utilising an advanced anoxic-aerobic MBR. The redox environment has been identified as a significant factor determining dye and AAs' biodegradability. Furthermore, the physical and chemical features of AAs had a significant impact on their removal efficiency. For example, EWG such as -Cl and azo bonds reduces removal efficiency, but the presence of EDG such as -NH2 in the AAs structure promotes removal efficiency.

According to Ahmed et al. (2017a), MBR improves the removal efficiencies of ECs. MBR can effectively remove a wide variety of ECs, including those that are resistant to activated sludge and built wetland procedures. MBR has a greater capacity for removing EDCs from wastewater. MBR is more successful than built wetlands at removing EDCs, PCPs, and beta-blockers.

6. Conclusion

Azo dyes are the synthetic textile colours most widely used. They contributed to between 50% and 70% of all dyes used in the textile industry and produced via anaerobic azo bond breaking, which created a colourless aromatic amine. Azo dyes emit AAs that content hazardous carcinogenic and prohibited in some countries. The European Commission has conducted a fast-track consultation on the possibility of restricting the use of additional hazardous compounds CMR 1A and 1B. However, the wet fabric sector had processes prior to employing azo dyes that affected water bodies and ecosystems, including sizing and desizing, scouring, bleaching, mercerizing, dyeing and finishing.

Apart from that, most of the journals screened used biological treatment as a low-pollutant treatment method, utilizing natural or organic agents to treat azo wastewater with the least amount of environmental pollution. Although bacteria and enzymes are relatively expensive, they can help reduce pollution. Therefore, microbial degradation of Azo dye has garnered considerable attention due to its low input, cost-effectiveness, and environmental safety.

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