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DECOLORIZATION OF DYES FROM TEXTILE WASTEWATER USING BIOCHAR: A REVIEW

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Abstract

The textile industry is one of the largest in many low and middle-income countries, especially in Asia, second only to agriculture. Textile wastewater is discharged into the environment due to the lack of affordable and sustainable solutions to adsorb or remove the dye from the water. Biochar is generated by pyrolysis of organic material from plant waste in low-oxygen conditions, and is considered carbon-negative. Biochar for dye adsorption in textile wastewater effluent was proven to be highly effective. However, adsorption efficiency varies with experimental parameters, therefore there is a gap in application especially in small dye houses. Efforts should be made to find innovative and affordable solution to make the textile industry more sustainable, by developing methods for collection and reuse, recycle and upcycle of textile waste, by reducing the consumption of water, energy and chemicals and by developing methods for treatment of the textile wastewater.

Keywords

textile dye effluent; biochar; wastewater; sustainability; contamination; sorption

Introduction

Climate change, population growth, rising standards of living and uneven distribution of water are the main causes for competition over water resources, water scarcity, poor water quality and variability of hydrological events. Water is the core of sustainable development and unfortunately, water is not equally available and in many areas around the world clean water is out of reach [1]. The environmental stress on water bodies is evident in terms of not only quantity, but also quality. Globalization of industrialization has resulted in high pollution of water resources worldwide. The major industries responsible for pollution are the dyeing industries, paper industries, tanneries, metal-plating industry, mining operations, fertilizer industries, agricultural waste and pesticides. The demand for water in the industrial sector is expected to increase by 283% during the first half of the 21st century [2], resulting in increasing industrial wastewater discharge.

Discussion

The textile industry is one of the largest in many low and middle-income countries, especially in Asia [3]. The textile industry is one of the cornerstones in economies of many countries [4]. For example, the Indian textile industry according to the India Brand Equity Foundation (IBEF), it is the second largest industry, after agriculture, providing employment to over 45 million people directly and 60 million people indirectly, and it contributes 14% of the Indian total industrial production [5]. Humans are aesthetically interested in dyed textile, therefore the use of dyes in textile will not be abandoned. Different types of dyes are used in a variety of industries including the food industry, textile, tanneries, plastics and pharmaceuticals. Many products in those industries contain

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several dyes from different chemical classes resulting in a complex wastewater [6]. Industrial dyes, in particular used in the textile industry, have complex molecular structures, synthetic in origin and recalcitrant [7].

Most of the chemicals are added in the dyeing process where a color is added to the dye baths, the fabric is immersed in the dye baths until the dye is fixed. In some cases, there is a need to add salt to the bath in addition to the color in the coloring processes to increases the affinity of the color to the fabric [8]. Over 7×10^5 tons of synthetic dye are produced annually, with 10–15% of it not ending up in the final product [4] thus eventually further contaminating the environment.

Therefore the effluent also contains a large amount of recalcitrant unfixed dyes (as acid dyes, basic dyes, sulfur dyes, chrome dyes, optical/fluorescent brightener and azoic dyes) as the dyes are not totally fixed to the fiber of different textiles during the dyeing process (fibers as wool and nylon, cotton and viscose, polyester and acrylic). The textile industry requires a large amount of water for the production process, and is also one of the major producers of wastewater that can have carcinogenic and mutagenic compounds [9]. Wastewater from the textile industry is frequently discharged directly into lakes and rivers without any proper treatment and often these water sources are used by locals domestically [10,11]. Since the dyeing process uses a significant amount of water, recycling used in the dyeing process can conserve water, however it requires treatment whether recycled or discharged to the environment. Reference values for water reuse in textile industry, included COD between 60-80 mg/L, conductivity of 1000 µS/cm and dissolved solids up to 500 mg/L [11]. In addition to dyes, the textile industry's wastewater contains also salts, acids and alkalis, oxidizers and recyclers, heavy metals, lubricating oils and fibers [11].

Dyes cannot be removed through conventional treatment unit operations due to the complex characteristics of the wastewater as high solubility, non-degradable nature, diversity and often changing speciation in water. When industrial wastewater is discharged into natural water bodies it can result in hazardous effects on the living systems because of the carcinogenic, mutagenic, allergenic and toxic nature of dyes [6]. This is a paradox as current conventional and advanced methods for the removal or degradation of persistent and emerging textile contaminants are limited, since they often involve intensive capital, lack of adaptive technological tools, social barriers and emphasis on centralized systems. Consequently, to close the gap, there is a dire need for innovative solutions and for widespread decentralized systems in the textile industry suitable for rural areas and capital-challenged countries.

There are many treatment methods when dealing with textile wastewater. The different techniques can be based on physical (sedimentation, filtration, floatation, coagulation, reverse osmosis, solvent extraction, adsorption, incineration, and distillation), chemical (neutralization, reduction, oxidation, catalysis, ion exchange, electrolysis) and biological (stabilization, aerated lagoons, trickling filters, activated sludge, anaerobic digestion) [12]. Physical methods are very common methods in textile wastewater treatment due to their high color removal efficiency, especially adsorption, filtration and membrane filtration. Other treatment processes as reverse osmosis, nanofiltration and multiple effect evaporators are effective but expensive, while the common treatment methods are non-destructive, lower in cost, time-consuming and less efficient [13].

Adsorption by porous materials is one of the most promising and affordable techniques for the removal of dissolved pollutants, serving as an alternative to energy-intensive technologies [14]. Activated carbon is a widely used adsorbent, but biochar, which is inexpensive, abundant and may have comparable adsorption capacity, can be used as an affordable alternative [15-17]. However, the efficiency, local manufacturing, availability and costs must be examined. Biochar is the result of low-temperature pyrolysis of carbon-rich biomass (from agricultural and forestry wastes) under low-oxygen conditions [18]. As can be seen in Fig. 1, there is an increase in the number of publications using the keywords: dye sorption, textile wastewater and biochar (by google scholar).

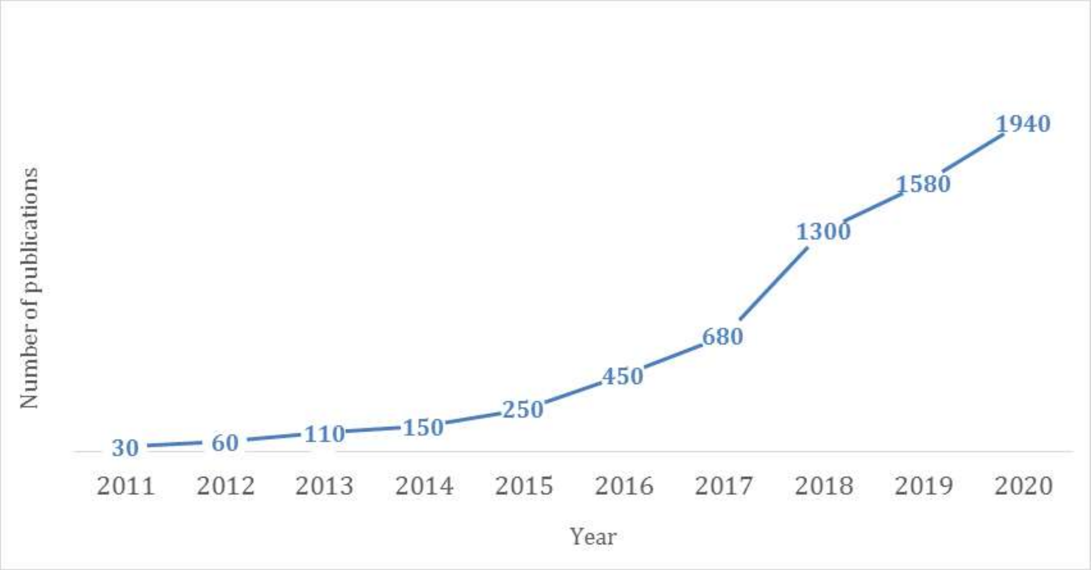

Fig. 1. Yearly number of publications using the keywords: dye sorption, textile wastewater and biochar *(by google scholar)*

The biochar created is a stable carbon black solid that is highly porous with large surface area (Fig. 2.). More than 70 percent of its composition is carbon [19]. The biochar's chemical composition varies with feedstocks used to make it and methods used to heat the biomass. The pyrolysis process varies as it can be done with different conditions as burning temperature and burning time, reactor volume, other gasses and materials in the reactor [20]. However, different thermochemical processes can be also used for biochar production [18]. Compared to activated carbon, biochar can be created from various types of biomass, requires less energy in the production process and consequently can provide a solution in the treatment of textile wastewater in poor income countries.

Fig. 2. Biochar particles image (left) and scanning electron microscopy (right, taken at Tel-Aviv University)

The porosity and high surface area of biochar make it an excellent adsorbent of organic contaminants and heavy metals in wastewater [17]. This has led to a growing interest in using biochar for water treatment, although most research today focuses on its abilities for soil fertilization [15]. Adsorption of textile dyes was examined with various biochar types [15,21,22]. For each type of dye, the biochar type, process parameters (temperature, pH, agitation time) and wastewater quality influence the efficiency of the dye removal from the wastewater. Most studies have focused on the removal of pure, highly concentrated dye solutions that do not represent the actual effluent from real dye houses. Real effluent may contain lower dye concentrations and additional substances as previously mentioned, such as salts, detergents, solids, and fiber residuals, which have a tremendous effect on the biochar's sorption capacity.

There are numerous types of dyes and biochars – and therefore the biochar type and dose should match the dye type. The major anionic dyes are the acid and reactive dyes, and the most problematic ones are the brightly colored, water-soluble acid dyes [23]. Acid dyes are a sub-group of anionic dyes and are called so, because they are usually applied to the fibers in acid solutions [24]. Cationic dyes are dyes that can be dissociated into positively charged ions in the aqueous solution, and the cationic dye dyes the fiber through the binding of its cation ion [7,23]. Basic dye, are highly visible and have high brilliance and intensity of colors [25].

As mentioned above, the textile industry uses different colors, each with its own properties and characteristics. Moreover, the biochar itself can vary based on the initial biomass from which it is produced. In order to find the best biochar-dye combination, a summary of articles examining different combinations was made [26-43]. In these articles, the biochar efficiency in removing the dye from a mixture was calculated using mass balance equation for dye adsorption on biochar [37].

Table 1 summarizes different characteristics from 18 articles that present the use of biochar in treatment of textile wastewater [26-43]. The articles are catagorized to biochar properties, dye properties, experimental conditions, dye adsorption experiments compared to a common biochar and modified biochar. All the experiments presented were batch experiment in a controlled environment. From the table it is evident that there are some differences between the treatment requirements for cationic dyes and anionic dyes. The adsorption of the dye to the biochar depend on the characteristics of the solution, the type of the biochar and the conditions in which the experiment was performed.

One of the critical conditions is the solutions pH, where the adsorption of cationic dyes will be maximized when the pH of the solution is basic and anionic dyes in acidic pH, with preferred pH at 3-4. The pH of the solution has a significant effect on the interaction of the dyes with the biochar in the adsorption process. For example, the effects of the solutions pH were examined on the adsorption of three dyes, Methyl Orange, an anionic dye and Malachite green & Methylene blue, both cationic dyes and three types of biochar, each produced with a different solvent (acetone, ethanol and methanol) [30]. The adsorption of the cationic dye Malachite Green onto the biochar increased with the increase in pH. The solution pH influenced the adsorption through dissociation of functional groups on the active sites of the biochar. In another study, the adsorption of anionic dye Reactive Red 141 onto a Pecan Nutshell based biochar occured mostly in an acidic environment with a low pH [32]. Under acidic conditions, the biochar surface is positively charged whereas the Reactive Red 141 has several sulfonated groups, which are negatively charged that are attracted to the biochar surface, increasing the dye removal percentage.

The dye to biochar interaction depends on the temperature of the mixture, the interaction time, the steering speed and more. Almost all the experiments showed that a temperature of 25-30°C is the optimal temperature for the dye's adsorptions. A higher or lower temperature can affect the dye particles' velocity in the mixture and will reduce the adsorption [27]. This result is significant because it means the treatment will not require any major heating or cooling systems and the treatment can be effective in real wastewater. The interaction time is also a crucial parameter. With short interaction time, the dye will not be able to sufficiently interact with the biochar, while with long interaction time, the binding between the dye and the biochar could become loose, and the biochar can undergo modifications that can damage its functionality and even release material and pollutants to the water and cause more damage [27,30,34]. Each biochar-dye combination has its own optimal interaction time depending on the biochar and dye's structure and concentrations and on the other conditions regulating the environment. Mubarak et. al. showed an increase in removal effecincy with contact time of methylene blue (cationic) and orange-G (anionic) onto Empty fruit bunch based biochar [36]. Another factor that can affect the interaction is the rotation speed of the mixture. A high rotation speed can lead to a high shear stress that can break the bindings between the biochar and the dye and can even damage the biochar particles and thus lower its efficiency. The most common rotation speed as presented in Table 1, is ~130-150 rpm.

Several studies attempted to increase an existing biochar activity by performing different modifications on it as Iron impregnation biochar, addition of cationic surfactant to the biochar surface, inserting magnetic formation to the biochar and more. These new characteristics provide the biochar with a stronger affinity and interaction between the biochar and the dye. One example of the benefits of these modifications used Rice Husk derived biochar mixed with a solution containing an aqueous phase reduction of ferrous iron ($FeSO_4 \times 7H_2O$) called nZVI, resulting in attachment of nanoscale zero-valent iron particles to the biochar creating a modified biochar (nZVI/BC) to adsorb the anionic dye Methyl Orange. In this experiment, three types of biochar various theoretical mass ratios of nZVI/BC at 1:3, 1:5, and 1:7 [28]. The nZVI/BC 1:5 adsorbed almost 100% of the dye in the mixture compared to the other modified biochars that peaked at 90% and the non-modified biochar that reached only 10%. Although there may be an advantage in using modified biochar, there might be a problem after extended usage. For example, high concentrations of oxygen can convert the Fe_0 molecules into ferrous or ferric oxide leading to a passivation layer forming on the nZVI surface.

Table 1. A summary of articles presenting biochar source, postproduction modifications, dye type, optimal conditions and experimental results

Biochar Source	Biochar preparation	Postproduction Modifications	Dye	Optimal Condition				Results	Reference
				pH	Temp (^oC)	RPM	Contact time (min)		
Spent mushroom substrate (SMS)	Pyrolysis at 450°C for 4 hours	200 cm^3 STP/min of N_2 was fed into the reactor, and steam was used as the activation agent at 800 °C for 2 h. Modification goals: Enhancement of the biochar's textural properties.	Congo Red (CR) (anionic) Crystal Violet (CV) (cationic)	CR:4 CV:6	30	150	CR:750 CV:1000	✓ Color and COD removal efficiencies up to 99.6% and 67.7% for CV. ✓ Color and COD removal efficiencies up to 10.3% and 23.7% for CR.	[41]
Rice husk (RHB) and Coir pith (CPB)	Pyrolysis at 700°C for 5 hours	RHB or CPB were added to a solution containing Fe $(NO_3)_3{\cdot}9H_2O$ dissolved in water. The mixture was oven dried at 105 °C for one day, followed by calcination at 500 °C for 4 h. Modification goals: Provide the biochar with oxidizing properties.	Acid Red 1 (AR1)	3	30-50	150	120	✓ Maximum dye removal for Fe-RHB was 97.6%. ✓ Maximum dye removal for Fe-CPB was 99.1%	[27]
Rice husk	Pyrolysis at 500°C	Biochar was mixed with HCl for demineralization then mixed with different mass ratios of nZVI.	Methyl orange (anionic)	4	25	-	15	✓ Maximum dye removal for nZVI at 5:1 was 98.5%. ✓ Removal capacity of 97.8,	[28]

		Modification goals: increase dye adsorption by transforming it to low molecular weight products through destruction of its N=N bonds.						✓ 306.7, 605.0, and 709.1 mg/g for initial concentrations of 60, 200, 400, and 600 mg/L, respectively.	
Bael shell (BS)	Pyrolysis at 500°C for 3 hours	The biochar did not undergo any special modifications	Patent blue (PB) (Anionic)	2.7	-	110	60	✓ Maximum dye removal was 74% (3.7 mg/g)	[21]
Carboxymethyl cellulose (CM) from raw chicken manure	Pyrolysis at 600°C for 2 hours	The biochar did not undergo any special modifications	Methyl orange (anionic)	6.5	25	150	30	Almost 100% dye removal	[29]
Sewage sludge (SS) with acetone as the solvent	Liquefaction at 260-280 °C	The biochar did not undergo any special modifications	Methyl orange (MO) (anionic) Malachite green (MG) (cationic) Methylene blue (MB) (cationic)	7	30	150	60	✓ Acetone based solution gave 53.12% removal. ✓ The bio-chars were only effective on cationic MG and MB with removal of 10–40 mg/g and 15–45 mg/g	[30]
Eichhornia crassipes-molasses	Pyrolysis at 400°C for 5 hours	The biochar did not undergo any special modifications	Methylene blue (MB) (cationic)	8	25	125	30	✓ Maximum adsorption capacity of 44.13 mg/g	[31]
Pecan nutshell	Pyrolysis at 800°C for 1 hour	The biochar did not undergo any special modifications	Reactive Red 141 (RR-141) (anionic)	2-3	25	250	80	✓ Increased initial dye concentration provided an increase adsorption from 40 to 130 mg/g ✓ 85% dye removal. 80% of saturation was attained within 10 min	[32]
Palm Kernel Shell (PKS)	Pyrolysis at 350oC in a rotary kiln for 20 min	The biochar did not undergo any special modifications	Crystal Violet (CV) (cationic)	6	25	100	30	✓ maximum adsorption capacity of 24.45 mg/g	[33]

Switchgrass Biochar modified by cationic surfactant (SB-TTAB)	Pyrolysis at 450ºC for 20 minutes	Biochar added to a solution of tetradecyltrimethyl ammonium bromide (TTAB) in ethanol. Modification goals: binding a cationic surfactant to the biochar to form micelle like structures which can solubilize dye within this structure and increase the biochar capabilities.	Reactive Red (RR-195A) (anionic)	5	25	150	40	✓ Dye removal from different contaminated solutions: conc.10/30/50 mg/L) ✓ Tap water %: 10 - 96.61 30 - 98.82 50 - 98.76 ✓ Raw water %: 10 - 98.56 30 - 97.79 50 - 99.26 ✓ Wastewater %: 10 - 100.00 30 - 94.83 50 - 94.24 ✓ Sea water %: 10 - 92.96 30 - 92.60 50 - 90.98	[34]
Kappaphy-cus alvarezii seaweed	Pyrolysis at 350ºC for 2 hours	The biochar did not undergo any special modifications	Reactive blue 4 (RB4) (anionic) Reactive orange 16 (RO16) (anionic)	2-3	30	180	60	✓ Around 90% of total reactive dye sorption occurred within the first 60 min of contact ✓ uptake of 0.324 mmol/g for RB4 ✓ uptake of 0.140 mmol/g for RO16	[35]
Empty fruit bunch (EFB)	microwave at 800W for 30 min.	EFB particles were treated chemically by ($FeCl_3$) before pyrolysis. Flow of nitrogen gas provided iron oxide magnetite formation to the chemical treated EFB. Modification goals: biochar with magnetic features enable the dye to be separated by magnetic separation techniques	Methylene blue (MB) (cationic) Orange-G (OG) (anionic)	MB: Both 2&10 OG:2	25	120	120	✓ Maximum adsorption capacity of 96.68% (31.25 mg/g) for MB ✓ Maximum adsorption capacity of 90.76% (32.36) mg/g for Orange-G	[36]
Empty fruit bunch (EFB)	Pyrolysis at 400ºC	The biochar did not undergo any special modifications	Methylene blue (MB) (cationic)	-	30	150	250	✓ Dye removal of 91%, 90%, 49% and 36% for 50, 100, 200 and 300 mg/L respectively ✓ The EFB biochar has a maximum sorption of 55.25 mg/g.	[37]

Spirulina platensis algae	Pyrolysis at 450°C for 2 hours	The biochar did not undergo any special modifications	Congo red dye (CR) (anionic)	2-7	30	120	15	✓ 75-80% dye removal ✓ For different initial dye concentrations.: 30,50,70,90,200mg/l	[38]
Chicken bones (CBB) after modification-MCBB	Pyrolysis at 500°C for 2 hours	Powdered CBB subjected to co-precipitation with a mixture of Fe^{3+} and Fe^{2+} salts. The CBB was added into a solution containing $FeSO_4 \times 7H_2O$ and $FeCl_3 \times 6H_2O$. Modification goals: biochar with magnetic properties for rapid sorption and a convenient recovery.	Rhodamine-B (RB) (basic dye)	10	50	150	120	✓ 88.5% dye removal for 40mgL-1. (36.2 mg/g) after 120 minutes. ✓ Approximately 96.5 mg/g of RB was adsorbed at pH 10 within 180 min and reduced to 68.5 mg/g in the presence of 0.5 g NaCl.	[39]
Pulp and paper sludge (PPS)	Pyrolysis at 108°C for 2 hours	PPS soaked in a $FeCl_3.6H_2O$ solution and dried before being pyrolyzed. Modification goals: Reduce porosity and decrease in pore volume as a result of nanoparticle impregnation that will lead to a rapid dye diffusion into the active sites when the particles organically detach from the biochar into the solution	Methylene blue (MB) (cationic)	12	-	-	40	✓ Impregnating PPS with Fe_2O_3 increased maximum adsorption capacity of the adsorbent by more than 50% saturation points for BC (97%) and NC (98%) both occurred at 5 g/L adsorbent ✓ The maximum adsorption capacities calculated 33 and 50 mg/g for BC and NC	[40]
Korean cabbage (KC)	Pyrolysis at 500°C for 1 hour	The biochar did not undergo any special modifications	Congo red (CR) (anionic) crystal violet (CV) (cationic)	CV:11 CR:7	30	150	1400	✓ maximum adsorption values: CR on KC: 95.81mg/g CV on KC: 1304mg/g	[41]
Sugarcane bagasse (SCB)	Pyrolysis at 400,600,800 °C for 1 hour. Best results for the 800 °C biochar	The biochar did not undergo any special modifications	malachite green (MG) (anionic)	7.5	60	10,000	51.89	✓ 100% removal of dye (conc.:500 mg/L) for SCB prepared at 800°C	[42,43]

Impact

The sustainability of the textile industry should be addressed across all sectors from fashion designers, manufacturers, product developers and the consumers. The process residuals as waste and wastewater generated requires innovative and affordable technologies and processes for collection and reuse, recycle and upcycle of textile waste (clothing and other textiles), for reducing the consumption of water, energy and chemicals and for treatment of the textile wastewater both in treatment plants and in small dye house industries. The environmental law for the dyeing effluent is in many cases very stringent and this necessitates the need for efficient treatment methods that must follow Zero Liquid Discharge (ZLD) either in common or in non-common treatment plants; however in practice due to the treatment costs it is not always practiced. In addition, even when plants are set for the ZLD, they are still generating hundred tons of hazardous solid waste per day as sludge (residual dyes and waste salts). Efforts should be made also on recovery of the dyes, and other organics from the wastewater before their discharge on to the soil and water bodies, in addition to efficient water treatment by combining novel hybrid membranes and nanotechnologies.

Tamil Nadu is in southern part of India and is engaged in textile processes especially the cotton textile industry. Real textile wastewater effluent from dye houses located in Coimbatore, Tamil Nadu, southern India, was examined for acid-dye removal from wastewater generated when dyeing silk filaments for production of soft silk sarees. The used dye solution from the dye houses is often discharged to the drainage or into the environment due to lack of affordable solutions. These dyes can potentially cause serious environmental damage and health. In our study, optimal conditions were demonstrated for filtration followed by high dye adsorption onto pine derived biochar (both in batch and column studies), and recommendations were suggested for reuse of the water back to the dye houses and for recovery of the biochar post use.

Conclusions

Different types of biochar were effective in adsorption of dyes from the textile wastewater effluent. Parameters that affect the process are temperature, rotation and mixing speed of the biochar with the dye in batch tests, and reaction time. Another important parameter is the pH of the biochar-dye suspension. Basic environment (pH higher than 7) was proven to be ideal for cationic dyes, where acidic environment (pH below 7) was proven to be ideal for anionic and acid dyes, with the optimal pH being between 3-4. Different post-production modifications to the biochar can increase the efficiency of the adsorption process and thus improve the entire treatment process; however, the long-term use and reuse of the modified biochar should be monitored.

Conflict of interest ′

There are no conflicts to declare’.

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