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Turmeric Dyeing of Cotton Fabrics using Tie-Dyeing Techniques

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Abstract

Tie-dyeing is a resist-dyeing technique which for years has been used by SMEs to produce attractive textile fabrics. However, the technique is challenged by the use of synthetic dyes which are not environmentally friendly and the growing consumers' environmental awareness. Accordingly, researchers and industrialists are forced to explore the alternative natural dyes in response to the above challenges. Among the natural source of dyes available abundantly in Tanzania, but are yet to be fully exploited for applications via tie-dyeing technique are turmeric (*Curcuma longa L.*) roots commonly used for food coloring.

In this study turmeric roots were investigated for their potential as dyes for coloration of cotton fabrics through tie-dyeing technique. Turmeric dyes were extracted through boiling of powdered turmeric roots in water followed by filtering the mixture, and applying it to a pre-mordanted and tied fabric. For analysis purposes, un-tied (plain) cotton fabric was also dyed following the same procedure. The findings of this study suggests that, turmeric dyes can be applied on cotton using tie-dyeing technique and produce golden-yellow color on fabrics. Additionally, pre-mordanting of cotton increased the color depth of dyed materials while improving their fastness properties with respect to washing and both wet and dry rubbing.

Keywords: Natural dyes, turmeric roots, mordants, tie-dyeing, cotton fabric, fastness properties

Introduction

In most developing countries, including Tanzania, textile dyeing by SMEs is commonly done by resist-dyeing techniques to produce colored patterns on fabrics. The technique requires blocking some parts of the fabric to prevent dyes from going through it during the dyeing process (Choi and Powell 2008). Examples of the common resist dyeing techniques include tie-dyeing, hot wax resist technique, starch paste resist, tritik, and clamping methods. Tie-dyeing is a form of resist dyeing technique which involves tying or stitching fabric together so as to prevent the absorption of dye to a particular area on the fabric ties (Gürses et al. 2016). In tiedyeing process, colored patterns are produced by gathering together many small portions of the fabrics and tied-tightly with loops or threads before immersing the fabric in dyebath so as to maintain undyed parts to gives the design effects by replacing either pleats, stitches or ties (Gürses et al. 2016). The origin of tie-dye has been associated with China, Japan, some parts of Asia, Indonesia, India, and some African countries mainly in Ghana and Nigeria (Gausa and Abubakar 2015). In Tanzania, tie-dyeing and resist dyeing in general, started in 1980s, when the country's economy started to decline and its textile industry failed to produce adequate and good quality garments (Kinabo 2004). To date, a significant amount of people in the country continue to produce and sell these colorful fabrics with distinctive designs, thereby providing employment opportunities and income to the people.

The tie-dye production process usually involves the use of large quantities of chemically synthesized dyes which are reported to be unfriendly to the environment. The synthetic dyes are produced from nonrenewable petrochemical sources and tend to introduce to the natural environment, some severe environmental hazards, particularly from effluent discharges which contain toxic or carcinogenic amines (Uddin 2021; Naveed et al. 2022). Unfortunately, in various parts of Tanzania, the traditional tie-dyeing process is usually done at the backyards of the peoples' homes, and in a poorly controlled manner. The improper handling and disposal of synthetic dyes harmful and auxiliary chemicals could eventually affect the health conditions of the producers and their families. These chemicals can cause respiratory issues, skin irritation, fire hazards and allergic reactions (Bruce-Amartey and Acquaye 2022). These challenges associated with the tie-dye production set up, together with the recent global increase in environmental awareness and the currently strict environmental standards set by many countries to avoid the health hazards associated with the use of synthetic dyes (Iqbal and Ansari 2021) suggest the need to extensively investigate alternative sources of dyes for tie-dyed fabrics processing.

Natural dyes have recently become of an interest to many researchers and textile dyers as an alternative to the chemically synthesized dyes (Samanta and Agarwal 2009; Shahid et al. 2013) and extensive researches is currently being conducted globally. These dyes produce unique colors with smooth, soft and harmonizing shades compared to synthetic dyes (Samanta and

Agarwal 2009: Hosen et al. 2021). Furthermore, they also offer good UV protection and antimicrobial properties. Natural dyes are extracted from renewable sources, are biodegradable and are therefore more environmentally friendly than synthetic dyes (Saxena and Raja 2014). They can be extracted from various parts of plants, animals and microorganisms, with plant parts such as leaves, barks, flowers, roots and fruits being common. Natural dyes are generally applied on materials using exhaust dyeing technique where both yarns and fabrics can dyed. Other unconventional dyeing be techniques such as ultrasonic dveing (Ma et al. 2020; Baig et al. 2021) and microwaveassisted dyeing (Adeel et al. 2018; Naveed et 2022) have also been reported. al. Nonetheless, natural dyes have no affinity for most textile fibers requiring an additional mordant such as ferric sulphate, copper sulphate and potassium aluminum sulphate to connect the functional groups of dye molecules onto the fiber, making the fabric resistant to fading (Ma et al. 2020).

Turmeric (*Curcuma longa L.*) is a flowering plant that has been used as a source of natural colorant, condiment and aromatic stimulant since ancient time (Umbreen et al. 2008). Turmeric rhizomes are ultimately the sources of phenolic compounds called curcuminoids (Bhatti et al. 2010). However, three curcuminoids. namely; curcumin. demethoxycurcumin, and bisdemethoxycurcumin are most common (Umbreen et al. 2008) with Curcumin, also known as C. I. Natural Yellow 3 (C. I. 75300) being the most active of the curcuminoids (Bhatti et al. 2010). Curcumin (1,7-bis (4hydroxy-3-methoxyphenyl)-1, 6 heptadiene-3, 5-dione) belongs to the diaroyl-methane group called diferuloyle-methane and is responsible for the brilliant yellow natural dye that is capable of direct dyeing of natural fibers such as cotton, silk and wool (Bhatti et al. 2010; Maulik et al. 2014). Curcumin exists in keto-enol form (Figure 1) which enables it to form inter- and intramolecular hydrogen bonds with respective substrates (Han and Yang 2005; El-Shishtawy et al. 2009; Reddy et al. 2013).



Figure 1: Structural Representation of Curcumin in Keto and enol tautomeric forms

Turmeric plants are widely grown in various parts of Tanzania for their roots which are used for food flavoring and coloring but are yet to be exploited for their textile dyeing potential. This is probably due to existing little knowledge on the overall aspect of natural dyeing of textiles among Tanzania's textile dyers and the lack of standardized natural dyeing procedures. The current study investigated the possibility of applying dyes extracted from the Tanzanian grown turmeric roots in coloration of cotton fabrics through the tie-dye technique. Two types of mordants, namely copper sulphate and iron sulphate, were applied to premordant the cotton fabrics, at relatively low mordant concentrations to ensure the dyes adhere to the textile substrate. To ensure the stability of dyed materials during subsequent processing and usage was established by assessing their color fastness properties. For analysis purposes, plain (non-tied) cotton fabrics were also dyed with the extracted turmeric dyes using the same procedure.

Materials and Methods Materials

Turmeric roots were obtained from a local market in Dar es Salaam, while 100% bleached cotton fabric was obtained from 21st Century Textile Mills in Morogoro, Tanzania. Mordants used for this study, copper sulphate (CuSO₄) and iron sulphate (FeSO₄) were purchased from chemical suppliers in Tanzania. All chemicals were of laboratory grade obtained from different suppliers in and outside Tanzania.

Methods

Extraction of Dyes from Turmeric Roots

Dyes were extracted from fresh turmeric roots by using water extraction method described by Umbreen and colleagues (2008). Prior to extraction, turmeric roots were washed with tap water to remove soil and dirt, peeled, cut into small pieces and dried in open air under sunlight for 7 days to reduce the water content and make them resistant to rotting. The dried turmeric root pieces were then pulverized using a heavy-duty food blender to obtain a turmeric rhizome yellow powder. A total of 10 g of this powder was weighted by using analytical weigh balance and then added into a glass beaker containing distilled water using a material-to-liquor ratio of 1:10. The mixture was stirred overnight at room temperature and then subjected to further stirring for 1 hour at 90 °C. The mixture was then allowed to cool at room temperature and filtered accurately to separate the solid matter from the liquid where non-dye materials and fine impurities were removed by filtration using a Whatman filter paper no 1. The obtained filtrate had a yellowish color and was then used as the natural turmeric dye. Figure 2 summarizes the process of extracting dyes from turmeric roots.



Figure 2: Turmeric dye preparation process

Mordanting of Cotton Fabrics

Pre-mordanting of cotton fabrics was done using copper sulphate and iron sulphate following a method described by Umbreen and colleagues (2008). The two mordants were selected in order to establish the influence of mordant ion nature on the color properties of dyed cotton fabric while premordanting method was selected because it has been reported to offer high color strength and darker shades in natural dyes compared to other mordanting methods, namely meta and post-mordanting (Bhatti et al. 2010; Manhita et al. 2011).

During pre-mordanting, two 10 g of bleached cotton fabrics were placed in beakers containing mordant solutions of two different concentrations (2 g/L and 4 g/L) using a material to liquor ratio of 1:10. The treatment was done for 60 minutes at 60 $^{\circ}$ C followed by rinsing the fabrics thoroughly in tap water and drying on a flat surface at room temperature.

Tie-Dyeing of Cotton Fabrics

Prior to dyeing, fabric patterns were designed where different pieces of the cotton fabric were folded, wrapped, stitched and tied with the aid of threads, loops and needle (Figure 3) in order to create different designing effects on the fabric during subsequent dyeing.



Figure 3: Pattern designing by stitching, folding and looping method.

Designed fabrics were then dyed in an aqueous solution of the extracted turmeric dyes, where 10 g of the fabric was immersed in the turmeric dye solution using a goods-to-liquor ratio of 1:10. The dyeing process was carried out at 90 °C for 30 minutes using glass beakers and hot stirring plates. Figure 4 shows the dyeing profile for the mordanted cotton fabric. For comparison purposes, cotton fabrics with no mordant were also dyed using the same procedures. The dyed

cotton fabrics were then removed from the dyeing beaker, unthreaded followed by washing with tap water to remove excess dyes captured on the surface of the fabric until clear water was observed. The fabrics were air-dried at room temperature. To ensure proper assessment of the color properties obtained on turmeric-dyed fabric, the dye was also applied on plain fabric using the same dyeing parameters.



Figure 4: Dyeing Profile of turmeric dye on mordanted cotton fabric

Characterization of the extracted turmeric dye

The pH of the water-extracted turmeric dyes was established using a pH meter (HANNA HI-98107, HANNA Instruments, USA) while the absorbance of the dye was by UV-visible assessed using a spectrophotometer (SPECORD 210 PLUS, Analytik, Jena, Germany) with spectra values measured in a range of 400-700 nm wavelength. The Infrared spectra of the extracted turmeric dye were determined by using Bruker Alpha-P Fourier Transform Infrared Spectrometer (Bruker optic GmbH, 2011) equipped with a diamond Attenuated Total Reflectance (ATR) sampling module. Measurements were an average of 25 scans in the wavenumber range of 4000 cm⁻¹ to 400 cm^{-1} .

Color Measurements

The colorimetric properties of mordanted and un-mordanted turmeric-dyed cotton fabrics were assessed in terms of color coordinates using CIELAB color spaces as well as the color strength (K/S) of dyed materials. During measurement, each fabric was folded four times in order to achieve opacity and data were collected under illuminant D65 using a 10° standard observer and was the mean of four reflectance measurements.

Lightness (L*), red/green coordinate (a*), yellow/blue coordinate (b*), chroma (C*), hue angle (h*), and reflectance values were established using the Datacolor 200M portable benchtop spectrophotometer equipped with Datacolor TOOLS Plus v2.3 quality control software. The color difference (ΔE) was calculated from the CIE L* a* b* color space and was meant to show the difference in dye acceptance on the unmordanted and mordanted turmeric-dyed cotton.

K/S values were calculated from reflectance values using a Kubelka-Munk equation (Equation 1), which relates the reflectance of the dyed fabrics to its reflectance and scattering at a specific wavelength.

 $K/_{S} = \frac{(1-R)^2}{2R}$ (Equation 1)

Where, K is the coefficient of absorption, S is the coefficient of scatter, R is reflectance at maximum wavelength expressed as a proportional value.

Color Fastness Properties

The color fastness properties of turmericdyed cotton fabrics were established by assessing the washing and rubbing properties of the materials. The wash fastness was performed according to TZS 24:1979 standard, where dyed cotton fabrics were subjected to a bath containing 10 g/L of ECE Formulation Phosphate Reference detergent (B) for 2 hours under room temperature. Assessment of the color change on washed samples was evaluated using greyscale and graded 1 to 5, with 5 indicating excellent wash fastness (i.e. no color change) and 1 being poor wash fastness (i.e. high level of color change).

The rub fastness test was conducted according to TZS 138:1981 at Tanzania Bureau of Standards. Both wet and dry dyed cotton fabrics were rubbed against white rubbing cloth by using Crockmaster as the rubbing instrument. Greyscale was used to assess color staining on the rubbing cloth and was graded 1 to 5, with 1 indicating poor rub fastness (i.e. high level of staining) and 5 indicating excellent rub fastness (i.e. no staining at all).

Results and Discussions

This study investigated the applicability of dyes extracted from turmeric rhizomes on

cotton fabric using the tie-dyeing technique. Prior to dyeing, turmeric dyes were extracted from powdered turmeric roots by using water as a dye extraction medium. The extracted turmeric dye was then applied on premordanted, folded/tied cotton fabrics and on unfolded/untied fabrics for assessment purposes. The findings of the study are hereby presented and discussed.

Characterization of the Extracted Turmeric dyes

In this study, the pH value of 5.4 was obtained from the water-extracted turmeric dye indicating that the dye was weakly acidic. The acidic pH of turmeric dyes is important during the dyeing of materials, because at this pH (3-7), the curcumins are most stable and in keto-enol form (Wang et al. 1997), with the equilibrium favoring the enol form. The enol structure enables the curcumin present in turmeric dyes to form additional interand intra-molecular hydrogen bonds (El-Shishtawy et al. 2009; Ma et al. 2020). For that matter, the pH of turmeric dyes was not altered during the dyeing of the cotton fabrics.

The UV-Vis absorption spectra for the water-extracted turmeric dyes was estimated using UV-Vis spectrophotometer and the result is shown in Figure 5. It is clear from the Figure 5 that the maximum absorption peak of curcumin is at the 425 nm wavelength in the visible region (400-700 nm) and the color of the extract was yellow. Kim and colleagues (2013) have also reported the maximum absorption peak of curcumin to be 425 nm and assigned the band to low energy π - π^* excitation of the curcumin. The peak at this range has also been reported to be due to the presence of enol group in the curcumin molecule (Mondal et al. 2016).



Figure 5: UV-Vis Spectra of the turmeric dye extracted using water extraction method

The extracted turmeric dye was also characterized by using FTIR technique and its spectra properties are presented in Figure 6 with each peak indicating the presence of different functional groups in the analyzed turmeric extracts. The detailed description of the observed functional groups is also presented in **Table 1**. The IR spectra of the extracted dye showed a broad absorption peak at a range of $3000 - 3700 \text{ cm}^{-1}$ which is due to phenolic stretching vibration (Zhou and Tang 2016). The peak at 3371 cm^{-1}

vibration group in curcumin. Aromatic ring stretching of -C-H group was traced at 2959 cm⁻¹ while the peak at 1624 cm⁻¹ represents the -C=O stretching bond. Further peaks were observed at 1516 cm⁻¹, representing the -C=C band of benzene ring stretching vibration and at 1122 cm⁻¹, representing the -C-O stretching. The presence of phenolic groups in the extracted dye confirms the presence of the curcumin in the extracted dye, similar to the findings that have been reported in other studies (Rohman et al. 2015; Zhou and Tang 2016; Zhang et al. 2021).



Figure 6: FTIR Spectra of the dye extracted from powdered turmeric roots

Wavenumber (cm ⁻¹)	Possible Assigned Functional Group
3371	OH Stretching vibration
2959	C=H Aromatic stretching
1624	C=O Stretching
1516	C=C Aromatic stretching
1122	C-O Stretching

 Table 1: Detailed Functional Groups of the Extracted Turmeric dye

Source: (Rohman et al. 2015; Zhou and Tang 2016; Zhang et al. 2021)

Analysis of Shades attained on Tie-Dyed Cotton Fabrics using Extracted Turmeric Dyes

Figure 7 visually compares the shades attained on the turmeric dyed fabrics. It is clear from Figure 7 that, the turmeric dye was successfully applied on different tie-dyed fabrics resulting in a yellow-to-brownish color which has been reported to be due to curcumin, the pigment present in turmeric roots (Maulik et al. 2014). It can also be seen from Figure 7 that each design resulted in variation in terms of dye uptake which is due to different patterns that were initially imparted on the cotton fabric through the tying process.



Figure 7: Visual appearance of colored patterns developed after tie-dyeing

Furthermore, mordanting of cotton fabric before subsequent tying and dyeing led to different shades compared to cotton fabrics dyed without being mordanted (Table 2), with darker shades observed on premordanted cotton fabric. In addition, different mordants also resulted into different shades. For instance, pre-mordanting with copper sulphate resulted in golden-yellow shade on cotton fabric while pre-mordanting with iron sulphate produced golden-brown shade. To ensure proper color assessment of the turmeric-dyed fabric, the dye was applied on plain fabric by using the same dyeing parameters and the results are presented in Table 3. It can be seen from the table that stronger yellow shades were observed on cotton fabrics pre-mordanted with copper sulphate and iron sulphate mordants compared to the pale shade obtained on unmordanted cotton. The shades were especially deeper when higher concentration of copper sulphate mordants was used which was then followed by the fabric premordanted with lower concentrations of iron sulphate. Similar findings have been reported

in previous literature where different types of mordants such as alum, iron sulphate and copper sulphate have been shown to produce different shades with different levels of depth when applied on textile materials (Manhita et al. 2011; Ghoreishian et al. 2013; Mulec and Gorjanc 2015).

DESIG N No.	Mordant Used	Un-Mordanted	2 g/L Mordant	4 g/L Mordant
1	CuSO ₄			
	FeSO ₄			
2	CuSO ₄	A STATE		
	FeSO ₄			No. of Contraction

Table 2: Comparison of Mordanted and Un-mordanted tie-dyed cotton fabric

Morda	Undyed	Un-mordanted	2 g/L Mordant	4 g/L Mordant
nt	Cotton			
CuSO ₄ FeSO ₄				
			民有法治	

 Table 3: Visual comparison of color developed on un-mordanted and mordanted turmeric-dyed cotton fabrics

Color properties of turmeric dyed cotton materials

The colorimetric coordinates as measured by colorimetry for turmeric dyed cotton fabrics are shown in Table 4. The results indicate that the Lightness (L*) values of the dyed cotton fabric decreased from 88.1 on un-mordanted cotton to 79.5 on the cotton material pre-mordanted cotton fabric with 4 g/L of iron sulphate. The decrease in lightness values of the pre-mordanted cotton could be due to deepening of the shades compared to the un-mordanted cotton fabrics. The great reduction in lightness of the iron sulphate mordanted cotton fabrics agrees with the findings reported on the darkening of colors upon mordanting with iron sulphate (Zarkogianni et al. 2011; Mulec and Gorjanc 2015).

Sample	L*	a*	b*	C*	h
Un-mordanted	88.08	-2.35	29.34	29.43	94.58
2 g/L CuSO ₄	83.51	-1.62	44.77	44.8	92.07
4 g/L CuSO ₄	80.95	-0.91	48.78	48.79	91.07
2 g/L FeSO ₄	81.82	0.63	40.82	40.83	89.11
4 g/L FeSO ₄	79.48	2.59	35.48	35.57	85.83

Table 4 Effect of Mordant on Colorimetric parameters of turmeric dyed cotton

Pre-mordanting of cotton materials also resulted to shifting in a* and b* values indicating the change in tone of the dyed cotton. The values of a* shifted from greenish on un-mordanted cotton fabric to redness when both copper sulphate and iron

sulphate mordants were used, with a big shift observed on using higher concentration of iron sulphate. Similarly, b* values shifted from blueness to yellowness, with a greater shift observed when higher concentration of copper sulphate mordant was used. The chromatic (C^*) and hue angle (h) of the turmeric-dyed cotton fabrics were also affected by the presence of mordants whereas chroma values increased indicating saturation or colorfulness while hue value decreased indicating a slight shift of shade from yellowness to redness. A significant shift in hue angle and chromatic value was observed on fabrics mordanted with iron sulphate, which agrees with the findings previously reported on the color-darkening tendency of iron sulphate when used as a mordant (Zarkogianni et al. 2011). Results of the color

difference (Figure 8) also show that premordanting of the cotton fabrics resulted in significant color changes with ΔE values ranging from 11 - 21, indicating that the obtained color change was visible. A higher color difference was observed when copper sulphate mordant was used which clearly indicated the superiority of Cu ions in the affinity of the cotton fibers as compared to Fe ions (Manhita et al. 2011).



Figure 8: Color difference (ΔE) between mordanted and un-mordanted turmeric dyed cotton

The color strength (K/S) of the turmeric dyed cotton (Figure 9) increased significantly when both copper sulphate and iron sulphate mordants were applied to cotton fabric prior to dyeing of the material. The highest K/Svalue was however observed on using higher concentration of copper sulphate where a K/Svalue of 1.97 was registered compared to K/Sof 0.5 obtained on un-mordanted cotton fabric. The enhanced K/S values on the premordanted cotton indicate an increase in dye uptake and subsequent fixation of the dye on fabric which is due to the metal mordants forming insoluble metal complex with the reactive groups of the dye molecules and hydroxyl groups present in cotton fabric. Both copper and iron are well known in making complicated coordination complexes (Zarkogianni et al. 2011; Prabhu and Teli 2014) resulting into higher color strength values compared to the un-mordanted cotton. The possible chemical reaction involved in turmeric dyeing of cotton in the presence of mordant is shown in Figure 10.







Turmeric dye

Cotton fibre, Mordant, Turmeric dye



Color Fastness Properties of Dyed Cotton Fabrics

Color fastness properties of the turmeric dyed cotton fabrics were evaluated and the results are shown in Table 5, where both washing and rubbing fastness properties are presented. Color fastness to washing was done to evaluate the ability of the dyed materials to withstand frequent changes while color fastness to rubbing was done to determine the amount of color transferred from the surface of the dyed material to other surfaces by rubbing. It can be seen from Table 5 that, even without mordanting the wash fastness properties of turmeric dyed cotton fabric were observed to be good to excellent with grey scale color change ratings of 4 and 5, respectively, indicating little or no significant color change. The little or no significant color change is thought to be due to the stability of the curcumin structure and the presence of van der Waals forces (Reddy et al. 2013; Lykidou et al. 2021).

Table 5: Color Fastness Properties of Turmeric dyed Cotton F	abrics
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	(Color Change)	Dry	Wet
Unmordanted	4	5	4/5
2 g/L FeSO ₄	4	4/5	4
4 g/L FeSO ₄	4	4/5	4
2 g/L CuSO ₄	5	4/5	4
4 g/L CuSO ₄	4	4/5	4

The results for both wet and dry rubbing fastness properties were observed to be good to very good with grey scale rating of 4 and 4-5, respectively, indicating no substantial staining of adjacent material even without the use of mordant. The stability of turmeric dyes on cotton even when un-mordanted could be due to the stability of the two benzene rings present in curcumin molecules (Bhatti et al. 2010). These compounds attach firmly on cotton giving it maximum resistance to detaching.

Conclusion

In this study, dyes from turmeric roots were successfully extracted from turmeric roots by using aqueous extraction method and were applied on cotton fabric using the tiedyeing method. For assessment purposes, the extracted dyes were also applied on plain cotton fabrics. Both tie-dyed and plain-dyed cotton fabrics appeared to have a yellow color, with the lightness of the fabrics decreasing on pre-mordanted fabrics indicating higher dye uptake and deepening of the color on dyed materials. The turmericdyed cotton fabric also exhibited good to excellent fastness properties on both mordanted and un-mordanted cotton fabric.

It can therefore be concluded that dyes extracted from turmeric roots can be applied on cotton fabric using the tie-dyeing method in the presence or absence of mordants and still produce good colors that will not bleed or stain adjacent materials during use. Turmeric dyes should therefore be considered as an eco-friendly source of natural yellow dye for tie-dyeing of textile materials, a technique that is mostly performed by smallscale (cottage) industries. Nevertheless, research is still needed to establish the use of natural mordants so that the potential risks introduced by synthetic and metallic

compounds are significantly reduced in the whole process.

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Conflicts of interest

The authors declare no conflict of interest regarding preparation and publication of this manuscript.

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