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Effect of Natural Fibers on Some Thermal and Physical Properties of Denture Base Materials

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Abstract

In this research, composite material consist of PMMA and MMA as a matrix materials was prepared and reinforced by natural fibers (sisal fibers) in different concentrations (5, 10%wt). The conventional processing technique for complete dentures was followed to prepare the composite specimens. FTIR test was carried out to reinforcement material (sisal fibers) before and after salinization to determine, whether or not there is chemical bond between reinforcements materials and saline coupling agent. Physical tests such as thermal conductivity, water sorption and solubility were performed on all specimens. The results refer to a highly significant differences in: thermal conductivity, water sorption and solubility of reinforced specimens compared with pure specimens. Increasing the fiber concentration showed a slight decrease in the thermal conductivity of PMMA specimens reinforced with sisal fibers, and increase both water sorption and solubility of composite specimens. FTIR results showed a new absorption band was developed after sialne treatment.

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1. Introduction

The artistic, physical, and mechanical characteristics of composites, as well as their clinical behavior, have an influence on their structure [1]. Polymethymethacrylate (PMMA) resin, commonly utilized in denture base. It has numerous advantageous such as ease of application and repair, inexpensive, acceptance by majority of the patients, oral cavity stability and aesthetical characteristics, but it has several drawbacks. Many efforts have been made to enhance its mechanical and physical properties, these efforts includes Denture materials can be strengthened by adding filler materials, changing the chemistry of PMMA and creating new denture base materials. Fiber-reinforced resin seemed to be the finest option[2,3]. Reinforcing fibers were presented to the denture base materials family. Plant fibers are one of the favorable biomaterials among natural fibers because of their biocompatibility[4]. The natural fibers have numerous advantages over artificial fibers includes renewable resource, less damage to treating equipment, low weight, inexpensive, good relative mechanical properties and environmentally friendly[5,6].

Ihsan and et.al, studied the incorporation of different filler such as (Ag,TiO₂,ZrO₂,Al₂O₃,SiC,SiC-nano,Si₃N₄ and HA-nano) in ratio of 10% wt on thermal conductivity and flexural strength of PMMA. The thermal conductivity

of PMMA improved with the inclusion of filler materials, according to the results of the experiments. The flexural strength values did not change appreciably [7].

Jawad and et.al, studied the thermal conductivity and water absorption of self-cured acrylic resin reinforced with natural fibers (siwak and bamboo) in different concentration (3,6 and 9%) wt and different length (2,4 and 6mm). The findings showed that as the fiber concentration and fiber length of both types of fibers increased, the values of thermal conductivity and water absorption increased [8].

Asopa and et.al, evaluated the flexural strength, impact strength surface hardness and water sorption of high impact acrylic resin reinforced with 10% and 20% ZrO₂. The reinforcement of acrylic resin with ZrO₂ has an effect on its physical and mechanical properties, according to the experimental results[9].

Fadhil and et al, prepared composite specimens consist of PMMA reinforced with two kinds of natural fibers (Bamboo and Siwak) in three concentrations (3,6&9%)wt and different lengths (2,6&12)mm. The results indicate that both the concentration of fibers and fiber length affect the impact strength and compression strength of PMMA [10].

Hamad investigated the effect of introducing two types of reinforcing particles, nano-alumina (Nano-Al $_2O_3$) and nano-silica (Nano-SiO $_2$), at varied volume fractions (1, 2, and 3)% wt, on the mechanical properties of composite prosthetic complete denture base materials. The results revealed that increasing the volume percentage of both (nano-Al $_2O_3$ and nano SiO $_2$) particles in PMMA increased the values of (compression strength and hardness) characteristics. While the impact strength values declined[11].

Sharhan and et al, studied the influence of adding synthetic fibers (polypropylene and polyacrylonitrile) on physical properties (water absorption, thermal conductivity and density) of PMMA. The results indicate that the values of water absorption increased as the reinforcement ratio increased, while the thermal conductivity and density properties decreased [12].

Hameed and Ali, utilized natural fibers (wheat and barley legs) as a reinforcing materials within polyester resin as a matrix materials to make composite materials. The findings indicate that the chemical treatment of these fibers with ethanol provides optimistic results and increases the adhesion of the interface between the fibers and matrix of the composite which lead to an increase in the strength of composite [13].

The aim of this research is to investigate the influence of sisal fibers on some properties; thermal conductivity, water sorption and solubility of PMMA composite.

2. Experimental Procedure

2.1 Materials and Methods

2.1.1 Materials

Heat cured denture base acrylic resin (Meliodent, Kulzer, Germany) was used as a matrix materials and natural sisal fibers as a reinforcing materials. The concentrations and the amounts of polymer and monomer where shown in table 1.

Table 1. Concentration and amounts of Polymer, Monomer and Sisal fibers used in this study.

Concentrations	Amount of reinforcement	Amount of PMMA (g)	Amount of monomer (ml)
(%)	materials (g)		
0%	0	35	14
5%	1.75	33.25	14
10%	3.5	31.5	14

2.1.2 Salinization of Sisal Fibers

10 grams of fibers were salinized with 3- Aminopropyltriethoxysilane by impregnated into the solution, which contained 25 ml of saline in 70 ml of absolute ethanol and 30ml of distilled water at PH 4.5-5.5 for one hour. Then the powders were dried at room temperature for 2 weeks.

2.2 Specimen preparation

The conventional processing technique for complete dentures was followed was used. The control specimens were prepared by mixing PMMA powder with MMA liquid at ratio (2.5:1) by wt. Sisal fibers were mixed in different percentages (5 and 10) %wt with monomer at room temperature before adding powder. Curing was carried out by placing the clamped flask in boiling water and switch the heat source off and leave for 15 min, then boil for 20 min. After curing has been concluded, the metal flask is allowed to cool down slowly in the water bath. Then the acrylic specimens were removed carefully from the mold.

2.3 Structural Test

2.3.1 Fourier Transformed Infrared Spectroscopy (FTIR)

The (FTIR) test was carried out to reinforcement material (sisal fibers) before and after salinization to determine whether or not there is chemical bond between reinforcements materials and saline coupling agent. A little amount of materials are mixed with KBR and placed inside the device. Infrared spectrums were collected in absorption and set to work in the (400–4000 cm⁻¹) range.

2.4Thermal Test

2.4.1 Thermal Conductivity Test

Lee's Disc method was used for calculating the thermal conductivity of materials using the instrument which is manufactured by (Griffin& George/ England). It is made up of three copper discs (A,B,C). The sample (S) is inserted among discs A and B, while the heater is inserted between discs B and C. The heater is powered by a voltage of 6 volts, and the current flowing complete the unit was roughly 1 amp (0.25A). The heat is transferred across the sample from the heater to the first two discs, formerly to the third disc. Figure 1 shows the Lee's disk scheme. The temperature of the three discs $(T_A, T_B,$ and $T_C)$ is determined by thermometers located within them. The temperatures are recorded once they have reached thermal equilibrium.

$$k\left[\frac{T_B - T_A}{ds}\right] = e\left[T_A + \frac{2}{r}\left(d_A + \frac{1}{4ds}\right)T_A + \frac{1}{2r}dsT_B\right] \tag{1}$$

T(A, B, C) are the temperatures of A, B, and C brass discs, respectively, and d(A, B, C) are the disc thicknesses (mm).

r: is the radius (mm).

e: the amount of heat flowing through the cross sectional area of the specimen per unit time $(W/m^2.^{\circ}C)$ calculated from the following equation:

$$V = \pi r^2 e(T_A + T_B) + 2\pi r e[d_A T_A + d_S \frac{1}{2}(T_A + T_B) + d_B T_B + d_c T_C$$
 (2)

where:

I: current through the heater (Ampere)

V: applied voltage (Volt)

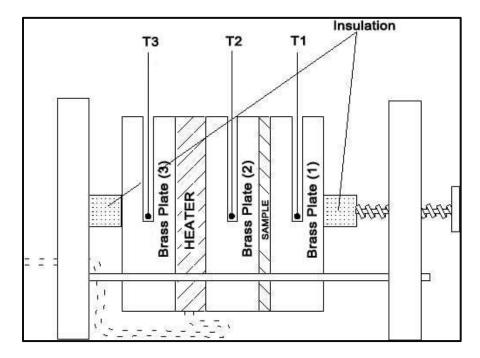


Figure 1: Scheme represents a lee's disk method for measuring thermal conductivity

2.5 Physical Tests

2.5.1 Water Sorption and Solubility Test

Acrylic disc specimens were made with a plastic model measuring 50 mm x 1 mm in diameter and 0.5 mm x 0.1 mm in thickness.)(ANSI/ADA specification No.12, 1999). Specimens were dehydrated in a dissector comprising newly dried silica gel, figure 2 . The discs were kept in an incubator at 37 $^{\circ}$ C ± 2 $^{\circ}$ C for 24 hours. The specimens were then allowed to cool for one hour before being evaluated using a numerical balance with a precision of (0.0001g). This cycle was repeated every day at the same time until, after 5 days, a constant mass called "conditioned mass" (M1) was attained.

The specimens were then immersed in purified water for 7 days at 37 °C ± 2 °C, following which each disc was withdrawn from the water using tweezers and wiped for 30 seconds with a clean dry hand tewel, then left in air for 15 seconds before being weighted (M2).

The assessment of water sorption calculated from the following equations:

$$ws = M2 - M1/S \tag{3}$$

WS: Water sorption (mg/cm²)

M1: The trained mass (mg)

M2: Mass of specimens after 7 days immersing in water (mg)

S: The surface area of the disc (cm²)

To determine the assessment of solubility, the discs were renovated to a constant mass in desiccators at 37°C±2°C, as was done formerly for the sorption test, and the renovated mass was detailed as (M3). Within 5 days, the entire troop had arrived at M3.

The assessment of water solubility, designed for each disc as of equivalence:

$$WSL = M1 - M3/S \tag{4}$$

WSL: Water solubility (mg/cm²)

M3: The reconditioned mass (mg)

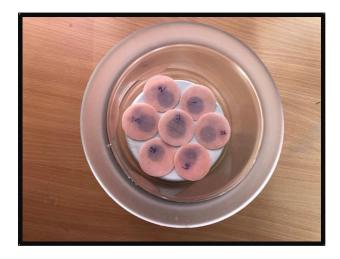


Figure 2: Specimens of water sorption and solubility were dehydrated in a dissector comprising newly dried silica gel.

2.6 Statistics Analysis

Data analysis was performed using (ANOVA) test. If p- value less than 0.05 was regarded as significant [14].

3. Results and Discussion

3.1 FTIR Results of Sisal Fibers

Figures 3 and 4 show the FTIR spectra of untreated and treated sisal fibers, respectively. Table 2 shows the characteristics of treated and untreated sisal fibers [15,16].

Table 2: Characterization spectral analysis of sisal fibers [15,16]

Functional group	Untreated sisal fibers	Treated sisal fibers(wavenumber, cm ⁻¹)
	(wavenumber, cm ⁻¹)	
O-H stretching, H-	3435.25-3406.40	3342.75
bonded		
C-H stretching	2912.61	2931.90
C=C stretching	1631.83	1639.55
C=C stretching	1508.38	1575.89
C-H bending	1427.37, 1371.43	1471.74
C-O stretching	1247.99	1120.60

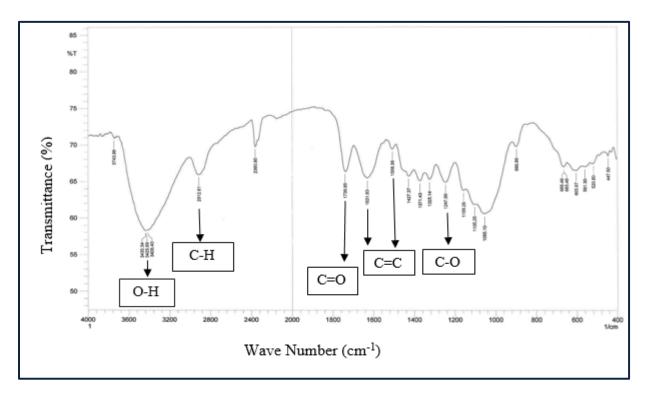


Figure 3: FTIR spectra of sisal fibers

The FTIR spectra of sisal fibers following salinization are shown in Figure 4. New absorption bands developed in the region after the silane treatment, ranging from 600 cm-1 to 1200 cm-1, and are distinctive to silane coupling agents. The stretching vibration of the Si-O-Si bonds produced from the condensation of the silane agents corresponds to the bands at 690.54 cm⁻¹ [17].

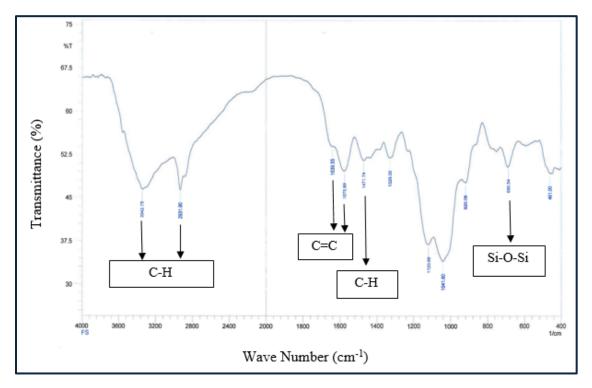


Figure 4:FTIR spectra of sisal fibers after salinization

3.2 Thermal Conductivity Test

The mean values, standard deviations, and standard error of thermal conductivity are shown in Table3. One way ANOVA suggested a highly significant differences among the group of thermal conductivity. Table 4 shows one way ANOVA results.

Table 3: Descriptive statistic of thermal conductivity test (W/m.°C)

Studied	N	Mean	Std. Deviation	Std. Error	Minimum	Maximum
group						
Control	10	0.3300	0.06470	0.02046	0.22	0.46
5%	10	0.3031	0.03028	0.00958	0.26	0.35
10%	10	0.2377	0.02325	0.00735	0.20	0.28

Table 4: One way ANOVA test of thermal conductivity test (W/m.°C)

	Sum of	Df	Mean	F	Sig
	squares		square		
Between groups	0.045	2	0.023	11.996	0.000
Within groups	0.051	**	0.002		
Total	0.096	79			

The post HOC test among groups of thermal conductivity revealed a highly significant difference between the control group and group of (5%) wt and high significant differences between group of (5%) wt and (10%) wt, but non-significant difference between control group and group of (10%). Table 5 shows the multiple comparisons of thermal conductivity among the groups.

Table 5: Post HOC test between groups of thermal conductivity test(W/m.°C)

(I) values	(J) values	Mean Difference (I-J)	Std. Error	Sig.
Control	5 %	0.09237	0.01940	0.000
	10 %	0.02695	0.01940	0.528
5%	10 %	-0.06543	0.01940	0.007
	Control	-0.09237	0.01940	0.000
10%	5 %	0.06543	0.01940	0.007
	Control	-0.02695	0.01940	0.528

Thermal conductivity is a material attribute that is determined by porosity (volume of air content). The thermal conductivity of the composite-fibers reinforcement decreases as the content of the fiber ratio increases. This behaviour could be due to the fact that every fibre has a large amount of air caught within its structure [18]. Natural fibers feature a distinctive microstructure termed lumen, which is well known. The natural fibres have a tube form because this lumen is normally filled with air. This interior microstructure is the source of natural fiber composites' unique functional capabilities [19]. With increased fiber content, the volume percent of air increases. When a result, as the fiber content increases, thermal conductivity decreases.

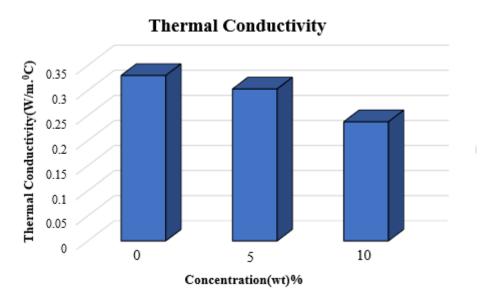


Figure 5: Mean values of thermal conductivity as a function of fiber concentration

3.3 Water Sorption and Solubility Test

Means, standard deviations, standard error and ANOVA test showed a significantly significant difference between the groups in the Water Sorption test as presented in table 6 and table 7.

Studied group	N	Mean	Std. Deviation	Std. Error	Minimum	Maximum
Control	10	0.4577	0.08242	0.02606	0.34	0.61
5 %	10	0.4433	0.07844	0.02480	0.30	0.59
10 %	10	0.7639	0.09955	0.03148	0.64	0.92

Table 6: Descriptive statistic of water sorption test (mg/cm²)

Table 7:One way ANOVA test of water sorption test(mg/cm²)

	Sum of squares	df	Mean square	F	Sig
Between groups	0.656	2	0.328	43.041	0.000
Within groups	0.206	77	0.008		
Total	0.862	۲ 9			

The post HOC test among groups of water sorption showed a highly significant difference between the control group and group of (10%) wt and between group of (5%) wt and (10%) wt, but non-significant difference between control group and group of (5%) wt of natural sisal fibers. Table 8 shows the multiple comparisons of water sorption among the groups.

(I) values	(J) values	Mean Difference (I-J)	Std. Error	Sig.
Control	5 %	0.01438	0.03904	1.000
	10 %	-0.30621	0.03904	0.000
5%	10 %	-0.32059	0.03904	0.000
	Control	-0.01438	0.03904	1.000
10%	5 %	0.32059	0.03904	0.000

Table 8: Post HOC test between groups of water sorption test (mg/cm²)

Water sorption of fiber-reinforced composites has a major role in the stability of fiber-reinforced dental appliances in watery environments like the oral cavity [20]. The mean values of water sorption increase with an increase in the weight fractions of sisal fibers, as shown in tables 6. This was because fibers' affinity for moisture, and it could also be related to the natural of the fibers' as a high moisture absorption levels in polymer matrix, which are caused by polar hydroxide groups in the fibers. Natural fibers become more hydrophilic when their hydroxyl groups are polarized. There are a lot of hydrogen bonds in the fiber cell wall. When water comes into contact with the fiber, old hydrogen bonds dissolve and new hydrogen bonds form between hydroxyl groups and water molecules, allowing the fiber to absorb more water[21].

0.03904

0.000

0.30621

Control

0.8 0.7 0.6 0.5 0.4 0.2 0.1 0 0 Concentration(wt)%

Figure 6: Mean values of water sorption as a function of fiber concentration

Descriptive statistic of water solubility are presented in table 9.

Table 9: Descriptive statistic of water solubility test(mg/cm²)

Studied	N	Mean	Std. Deviation	Std. Error	Minimum	Maximum
group						
Control	10	0.03358	0.011251	0.003558	0.016	0.049
5 %	10	0. 02730	0.013267	0.02480	0.30	0.59
10 %	10	0.7639	0.09955	0.03148	0.64	0.92

One way ANOVA suggested a highly significant differences among the group of water solubility. Table 10 shows one way ANOVA result

Table 10:One way ANOVA test of water solubility test (mg/cm²)

	Sum of squares	df	Mean square	F	Sig
Between groups	0.008	2	0.004	42.807	0.000
Within groups	0.004	77	0.000		
Total	0.013	79			

The post HOC test among groups of water solubility showed a highly significant difference between the control group and group of (10%) wt and between group of (5%) wt and (10%) wt, but non-significant difference between control group and group of (5%) wt of natural sisal fibers. Table 11 shows the multiple comparisons of water solubility among the groups.

Table 11: Post HOC test between groups of water solubility test (mg/cm²)

(I) values	(J) values	Mean Difference	Std. Error	Sig.
		(I-J)		
Control	5 %	0.06277	0.005752	0.854
	10 %	-0.31524	0.005752	0.000
5%	10 %	-0.37801	0.005752	0.000
	Control	-0.006277	0.005752	0.854
10%	5 %	0.37801	0.005752	0.000
	Control	0.31534	0.005752	0.000

Solubility

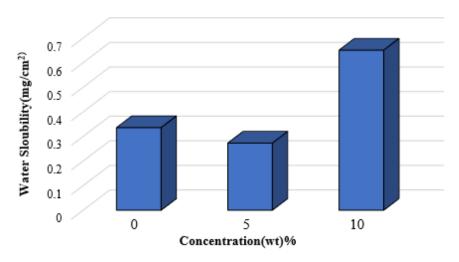


Figure 7: Mean values of water solubility as a function of fiber concentration

3.4 Morphology test

3.4.1 Filed emission scanning electron microscope (FE-SEM)

For the pure PMMA resin and PMMA composite specimens, field emission scanning electron microscopy (FE-SEM) micrographs of the fracture surface of flexural test specimens were taken. As shown in the figure 8. FE-SEM images of fracture surface morphology of pure PMMA at different magnifications [22]. It looked to be a homogenous morphology in the majority of cases. The ductile fracture of pure PMMA was confirmed by FE-SEM analysis.

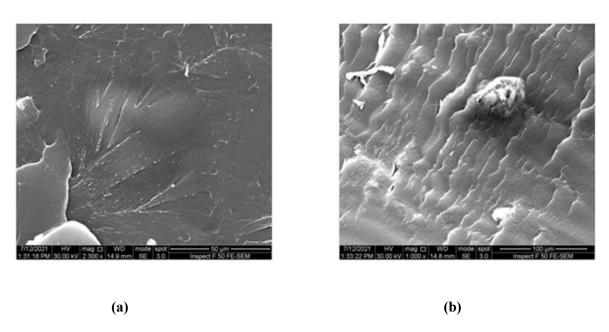


Figure 8: FE-SEM image of fractured surface morphology of pure PMMA with different magnification,(a)FE-SEM magnification 2000x, 50μm, (b) FE-SEM magnification 1000x, 100μm.

FE- SEM images of the fracture surface morphology of PMMA reinforced with 5% wt and 10% wt of sisal fiber, were shown in figure 9 and figure 10 at different magnifications respectively. It is noticed through FE-SEM image that the majority of the sisal fiber converted portion of the matrix material, incorporated in PMMA matrix material. The fibers were submerged within the matrix material, so the figures demonstrate a strong association

between the sisal fiber and the PMMA material, indicating stronger interfacial adhesion between fibers and composite materials constituents. The brittle fracture of PMMA reinforced with sisal fibers was confirmed by FE-SEM analysis.

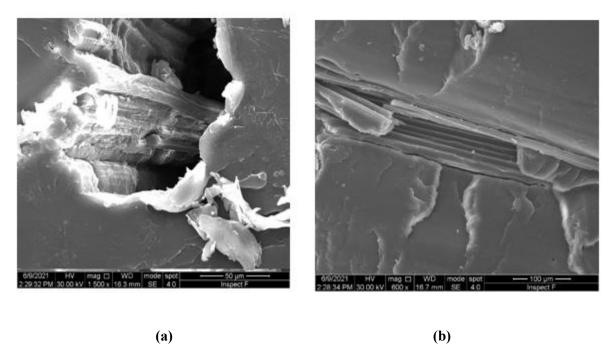


Figure 9: FE-SEM image of fractured surface morphology of PMMA reinforced with 5% wt of sisal fibers with different magnification,(a)FE-SEM magnification 1500x, 50μm, (b) FE-SEM magnification 600x, 100μm.

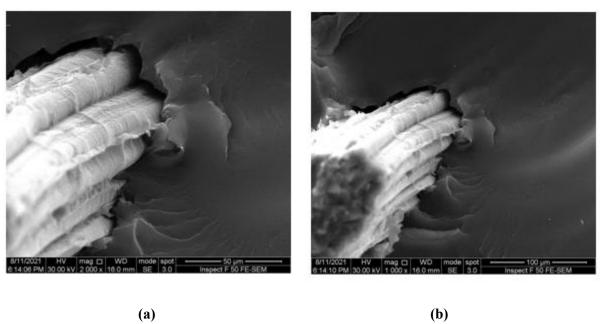


Figure 10: FE-SEM image of fractured surface morphology of PMMA reinforced with 10% wt of sisal fibers with different magnification,(a)FE-SEM magnification 2000x, 50μm, (b) FE-SEM magnification 1000x, 100μm.

4. Conclusions

Within the constrains of this investigation, it can concluded that the PMMA reinforced with natural sisal fibers showed significant changed in the physical properties. Thermal conductivity of PMMA decreased as the

concentration of sisal fibers increased,the values of thermal conductiovity of 5% wt and 10% wt sisal fibers was (0.3031W/m.°C) and (0.2377 W/m.°C) respectively, while of pure PMMA was (0.3300 W/m.°C). Also, it obsetved that a significantly increase the water sorption and solubility of acrylic resin. FTIR results showed a new absorption band was developed after sialne treatment.

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Conflict of Interest

There are no conflicts of interest regarding the publication of this manuscript.

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