

CHARACTERIZATION AND PROPERTIES OF SEMI-BIODEGRADABLE LOW
DENSITY POLYETHYLENE/PALM PRESSED FIBRE BIOCOMPOSITE FILM

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*This thesis is
Specially dedicated to*

*My beloved Father and Mother
Aris Bin Deros and Siti Noorliah Binti Bakar*

*My lovely sibling and fiancé
Nurulakhwan Aris and Mohd Fariz Hasan*

Thanks for all the support, endurance, inspiring spirit, patience and prayers.

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ABSTRACT

Semi-biodegradable low density polyethylene/press palm fibre (LDPE/PPF) biocomposite film was successfully prepared. PPF was treated with sodium hydroxide (NaOH) at various concentrations and soaking durations. Oil content analysis showed that the removal of oil residue was largely influenced by soaking time rather than NaOH concentration. The result was supported by a Fourier transform infra red analysis, where the peak intensities of carbonyl groups (C=O) at 1630-1700 cm^{-1} and alkene (C=C) at 1596 cm^{-1} decreased with soaking durations. Longer soaking duration led to fully-swollen PPF cellulose chains, thus allowing more oil to be easily removed from the surface of the fibre. However, the removal rate was gradually decreased after 6 hours, probably due to the difficulty to remove the oil trapped among the entangled-swollen cellulose chains. The crystalline phase of PPF was changed into the amorphous rich phase, as shown by the shifted of bonded-OH stretching band at 3280 cm^{-1} to freed-OH at 3345 cm^{-1} . In addition, the decrease of area under the curve of melting temperature (T_m) and the heat of melting from the differential scanning calorimetry analysis supported the findings. Meanwhile, LDPE/PPF biocomposite was prepared by blending the LDPE resin with various percentages of PPF and glycerol. The mixtures were compounded using a single screw extruder prior to blown extrusion into films. Results from the melt flow index and rheological tests showed that the addition of PPF increased the shear stress and viscosity of samples, owing to the restriction of chain mobility of LDPE by PPF particles. Higher PPF loading had also decreased the tensile strength and elongation at break of the film due to agglomeration of PPF's particles that caused phase separation between PPF-LDPE matrix, thus weakening the strength of the samples. The formation of a discrete phase observed on the micro-images taken from the scanning electron microscopy supported the results. Meanwhile, increasing the amount of PPF from 3wt% to 7 wt% had increased at about 43% water absorption in 30 days due to the formation of hydrogen bonding between the OH groups of PPF, glycerol and water. This condition provided a suitable environment for the growth of microorganisms thus increasing the biodegradation rate of the samples at an average of 31% weight loss in 12 weeks. However, T_m and the degree of crystallinity were unaffected with increasing PPF content as these properties are strongly influenced by LDPE.

ABSTRAK

Filem biokomposit polietilena berketumpatan rendah/serat sabut buah kelapa sawit (LDPE/PPF) separa urai telah berjaya dihasilkan. PPF telah dirawat dengan sodium hidroksida (NaOH) pada pelbagai kepekatan dan tempoh rendaman. Analisis kandungan minyak menunjukkan penyingkiran sisa minyak lebih dipengaruhi oleh tempoh rendaman berbanding kepekatan NaOH. Keputusan ini disokong oleh analisa inframerah transformasi Fourier di mana keamatan puncak kumpulan karbonil (C=O) pada $1630-1700\text{ cm}^{-1}$ dan alkena (C=C) pada 1596 cm^{-1} berkurang dengan tempoh rendaman. Lebih lama tempoh rendaman membawa kepada pembengkakan penuh rantaian selulosa PPF, sekaligus membenarkan lebih banyak minyak disingkirkan dengan mudah dari permukaan serat. Walau bagaimanapun, kadar penyingkiran menurun secara perlahan selepas 6 jam, berkemungkinan disebabkan oleh kesukaran untuk menyingkirkan minyak yang terperangkap di celah rantaian selulosa yang bengkak dan berselirat. Fasa penghabluran PPF telah bertukar kepada fasa yang kaya dengan amorfus, seperti yang ditunjukkan oleh peralihan puncak rengangan OH-terikat pada 3280 cm^{-1} kepada OH-bebas pada 3345 cm^{-1} . Tambahan lagi, pengurangan luas bawah lengkung suhu lebur (T_m) dan haba lebur dari analisis kalorimetri pengimbas pembezaan menyokong penemuan ini. Sementara itu, biokomposit LDPE/PPF telah disediakan melalui pencampuran resin LDPE dengan PPF dan gliserol pada pelbagai peratus. Campuran tersebut telah diadun menggunakan penyemperit satu skru sebelum ditiup dengan penyemperitan tiupan untuk menghasilkan filem. Keputusan dari indeks aliran lebur dan ujian reologi menunjukkan bahawa penambahan PPF meningkatkan tegasan ricihan dan kelikatan sampel, disebabkan oleh sekatan pergerakan rantaian LDPE oleh zarah-zarah PPF. Kandungan PPF yang tinggi juga mengurangkan kekuatan regangan dan pemanjangan pada takat putus filem kerana penggumpalan zarah-zarah PPF menyebabkan pemisahan fasa antara PPF-matrik LDPE, lantas melemahkan kekuatan sampel tersebut. Pembentukan fasa terputus-putus yang dilihat pada imej mikro dari mikroskopi pengimbas elektron menyokong keputusan tersebut. Sementara itu, penambahan jumlah PPF dari 3wt% kepada 7wt% telah meningkatkan sebanyak 43% jumlah penyerapan air untuk tempoh 30 hari yang disebabkan oleh pembentukan ikatan hidrogen di antara kumpulan OH PPF, gliserol dan air. Keadaan ini menyediakan persekitaran yang sesuai untuk pembesaran mikroorganisma, sekaligus meningkatkan kadar biopenguraian sampel pada purata 31% kehilangan berat dalam masa 12 minggu. Walau bagaimanapun, T_m dan darjah penghabluran tidak terjejas dengan penambahan kandungan PPF kerana kedua-dua sifat ini lebih dipengaruhi oleh LDPE.

TABLE OF CONTENTS

CHAPTER	TITLE	PAGE
	DECLARATION	ii
	DEDICATION	iii
	ACKNOWLEDGEMENTS	iv
	ABSTRACT	v
	ABSTRAK	vi
	TABLE OF CONTENTS	vii
	LIST OF TABLES	x
	LIST OF FIGURES	xii
	LIST OF ABBEREVIATIONS	xiv
	LIST OF SYMBOLS	xvi
1	INTRODUCTION	1
	1.1 Background of Study	1
	1.2 Problem Statement	3
	1.3 Objective	5
	1.4 Scope of Study	5
2	LITERATURE REVIEW	7
	2.1 Plastics and Environment	7
	2.2 Biodegradable Plastic	8
	2.3 Biodegradable Process	12
	2.4 Low Density Polyethylene (LDPE)	15
	2.5 Natural Fibre	19
	2.6 Palm Pressed Fibre	24
	2.7 Chemical Modification or Mercerization	27

2.8	Chemical Coupling	31
2.9	Plasticizer-Glycerol	32
2.10	Glycerol on LDPE	34
2.11	Natural Fibre Blending with Polymer Matrix	34
2.12	Blow Film Extrusion Process	36
3	METHODOLOGY	38
3.1	Materials	38
3.1.1	LDPE	38
3.1.2	Palm Pressed Fibre (PPF)	39
3.1.3	Sodium Hydroxide (NaOH)	39
3.1.4	Silane	40
3.1.5	Glycerol	40
3.2	Preparation of Composite Film	40
3.2.1	Chemical Treatment of PPF	40
3.2.2	Compounding Formulation	41
3.2.3	Mixing	42
3.2.4	Compounding with Single Screw Extruder	42
3.2.5	Blown Film Extrusion	43
3.3	Sample Characterization	44
3.3.1	Fibre Analysis	44
3.3.1.1	Oil Content Analysis	44
3.3.1.2	Fourier Transform Infrared (FTIR)	44
3.3.1.3	Differential Scanning Calorimetry	44
3.3.2	Biocomposite Film Analysis	45
3.3.2.1	Melt Flow Index	45
3.3.2.2	Rheological Properties	45
3.3.2.3	Fourier Transform Infrared (FTIR)	45
3.3.2.4	Tensile Test	46
3.3.2.5	Differential Scanning Calorimetry	46
3.3.2.6	Scanning Electron Microscopy (SEM)	47
3.3.2.7	Water Absorption Test	47
3.3.2.8	Biodegradation Test (Soil Burial)	48

4	RESULT AND DISCUSSION	49
4.1	Fibre Analysis	49
4.1.1	Oil Content Analysis	49
4.1.2	Fourier Transform Infrared (FTIR)	52
4.1.3	Differential Scanning Calorimetry (DSC)	55
4.2	Biocomposite Film Analysis	56
4.2.1	Melt Flow Index	56
4.2.2	Rheological Properties	58
4.2.3	Fourier Transform Infrared (FTIR)	60
4.2.4	Tensile Test	63
4.2.5	Difference Scanning Calorimetry (DSC)	67
4.2.6	Scanning Electron Microscopy (SEM)	69
4.2.7	Water Absorption Test	70
4.2.8	Biodegradable Test	72
5	CONCLUSION AND RECOMMENDATION	75
5.1	Conclusion	75
5.1.1	Fibre Analysis	75
5.1.2	Biocomposite Film Analysis	76
5.2	Recommendation	77
	REFERENCES	78
	Appendices I-III	85

LIST OF TABLES

TABLE NO.	TITLE	PAGE
2.1	Differences of type of natural biodegradable	10
2.2	T _m and chemical structure of polymer	12
2.3	Definition of biodegradation	13
2.4	Physical properties of various type of PE	16
2.5	Application of PE	17
2.6	Properties of natural fibre and glass fibre-E	21
2.7	Chemical composition of common natural fibre	22
2.8	Mechanical properties of natural fibre	23
2.9	Chemical composition of OPEFB and PPF	25
2.10	Solubility of OPEFB and PPF in different solvent	27
2.11	Comparison on chemical composition of raw jute and treated jute	28
2.12	Effects of mercerization treatment on properties of biocomposite	30
3.1	Product of data sheet of titanlene LDF260GG	39
3.2	Parameter of alkali treatment on PPF	41
3.3	Sample formulation	42
3.4	Processing parameter for compounding process	43
3.5	Processing parameter for blown film extrusion	43
4.1	Percentages of oil residue in PPF	51
4.2	Differential functional group between untreated and treated PPF	53
4.3	Effect of PPF treatment on MFI value without glycerol	57
4.4	Effect of PPF treatment on MFI value with glycerol	57

4.5	Summary of major wavelength detected in all sample	61
4.6	Comparison of mechanical properties of LDPE/PPF biocomposite film	65
4.7	T_m and degree of crystallinity of LDPE/PPF biocomposite film	68
4.8	Percentages of water absorption of LDPE/PPF biocomposite film	72
4.9	Percentages of weight loss for LDPE/PPF biocomposite film	74

LIST OF FIGURES

FIGURE NO.	TITLE	PAGE
2.1	Mechanism reaction between hydrogen and ester bond	10
2.2	Comparison between degradation and biodegradation	12
2.3	Path reaction between of aerobic and anaerobic process	14
2.4	Structure of Low Density Polyethylene (LDPE)	18
2.5	Main application of LDPE in industries	18
2.6	Classification of natural fibre	20
2.7	Picture of a) EFB b) PPF	25
2.8	Fruitlets of oil palm bunch	26
2.9	Typical structure a) untreated fibre b) treated fibre	28
2.10	Schematic diagram of swelling process in cellulose	29
2.11	Formation of alkali cellulose and cellulose-II	29
2.12	Schematic reaction of silane (3-APM) and fibre	32
2.13	Diagram of blow film extrusion	37
3.1	Specimen dimension for tensile test	46
4.1	Percentages of oil residue in PPF after 1 hour at various NaOH concentration	51
4.2	Percentages of oil residue on PPF at 1% of NaOH concentration at various soaking duration	52
4.3	FTIR spectra of PPF with various soaking time at 1% NaOH concentration	54
4.4	FTIR spectra of PPF with various NaOH concentration at 1 hour soaking duration	54
4.5	Tm of untreated and treated PPF with various concentrations of NaOH at 1 hour soaking duration	55

4.6	Area under the curve of treated and untreated PPF with various concentration of NaOH at 1 hour soaking duration	56
4.7	Viscosity versus shear rate of LDPE/PPF biocomposite film	59
4.8	Shear stress versus shear rate of LDPE/PPF biocomposite film	59
4.9	FTIR spectrum of pure LDPE (reference)	61
4.10	FTIR spectrum of a) LDPE/3wt%PPF/silane/1wt%glycerol b) LDPE/3wt%PPF/silane/2wt%glycerol	62
4.11	FTIR spectrum of a) LDPE/7wt%PPF/silane/1wt%glycero b) LDPE/7wt%PPF/silane/2wt%glycerol	63
4.12	Young's modulus of LDPE/PPF biocomposite film	65
4.13	Tensile strength of LDPE/PPF biocomposite film	66
4.14	Elongation at break of LDPE/PPF biocomposite film	66
4.15	DSC curve of LDPE/PPF biocomposite film	68
4.16	Surface morphology of pure LDPE	69
4.17	Surface morphology of LDPE/PPF biocomposite film at 500X magnification	70
4.18	Water absorption behavior for LDPE/PPF biocomposite film	71
4.18	Percentages of weight loss of LDPE/PPF biocomposite film	74

LIST OF ABBREVIATION

3-APE	-	3-aminopropyltriethoxysilane
3-APM	-	3-aminopropyltrimethoxysilane
ASTM	-	American Society for Testing and Material
DOP	-	Diocetyl Phthalate
DSC	-	Differential Scanning Calorimetry
FFB	-	Fresh Fruit Bunch
FTIR	-	Fourier Transform Infrared
HDPE	-	High Density Polyethylene
LDPE	-	Low Density Polyethylene
LLDPE	-	Linear Low Density Polyethylene
ICI	-	Imperial Chemistry Industries
MFI	-	Melt Flow Index
OPEFB	-	Oil Palm Empty Fruit Bunch
OPF	-	Oil Palm Frond
OPT	-	Oil Palm Trunk
PE	-	Polyethylene
PET	-	Poly (ethylene terephthalate)
PHA	-	Polyhydroxyalkanoates
PLA	-	Poly (Lactic Acid)
PKC	-	Palm Kernel Cake
PKS	-	Palm Kernel Shell
POME	-	Palm Oil Mill Effluent
PP	-	Polypropylene
PPF	-	Palm Pressed Fibre/Mesocarp Fibre
PS	-	Polystyrene
PVC	-	Poly (vinyl chloride)

SEM	-	Scanning Electron Microscopy
UV	-	Ultraviolet
VLDPE	-	Very Low Density Polyethylene

LIST OF SYMBOLS

ΔH	-	Change of enthalpy
ΔS	-	Change of entropy
ΔH_m	-	Heat of melting
ΔH_f	-	Heat of formation
%	-	Percent
$^{\circ}\text{C}$	-	Degree Celsius
wt%	-	Weight percentage
T_m	-	Melting temperature
hr	-	Hour
MPa	-	Mega Pascal
Gpa	-	Giga Pascal
g/cm^3	-	Gram per centimeter cubes
kg/cm^2	-	Kilogram per centimeter square
$\text{g}/10\text{min}$	-	Gram per 10 minutes
$\text{g}/\mu\text{m}$	-	Gram per micrometer
kg	-	Kilogram
g	-	Gram
rpm	-	Revolution per minute
kN	-	Kilo Newton
μm	-	Micro meter
X_c	-	Degree of crystallinity
Mt (%)	-	Percentages of weight gain
CO_2	-	Carbon Dioxide
CH_4	-	Methane
H_2O	-	Water
H_2S	-	Hydrogen Sulphide

NaOH	-	Sodium Hydroxide
NH ₃	-	Ammonia
NO ₂	-	Nitrogen Oxide

CHAPTER 1

INTRODUCTION

Introduction

This chapter consists of the overview of the project including project background, problem statement, objective and scope of study.

1.1 Background of Study

Plastics are typical organic polymers that mostly derived from petrochemical resources. They are found in many applications such as transportations, packaging, agricultures, electrical appliances, textiles and many more. Plastics are replacing metals, woods, glasses and ceramics due their lightweight, processability and good mechanical, thermal and barrier properties (Siracusa *et al.*, 2008; Tharanathan, 2003). The main drawback is their non-environmental friendly natures which take from hundreds to million years to degrade under environment condition thus, contributing huge piles of plastic wastes in the landfills (Sepulveda *et al.*, 1999).

However, this issue can be minimized or eliminated by using semi-biodegradable or totally biodegradable plastics that produced from natural, synthetic or combination of both polymers that decompose either through aerobic or anaerobic processes (Tharanathan, 2003). The common practice to impart degradability to

polymers is by blending the polymers with natural fibres such as jute, kapok, hemp, sisal and oil palm biomass (Mwaikambo *et al.*, 2001). Natural fibres offer many advantages such as good stiffness, specific strength and gravity, non toxic, cheap and readily available (Swamy *et al.*, 2004). Many plastic resins such as polyethylene (PE) and polypropylene (PP) are used as matrices for biocomposites. Among the PE families, low density polyethylene (LDPE) is commonly used in packaging film sectors due to longer branches molecule chains which impart good melt strength during blown film extrusion. The branches also contribute to highly amorphous regions (about 50%) that offer good mechanical and optical properties. Ismail *et al.* (2010) had prepared LDPE/kenaf fibre biocomposite where the kenaf loading was limited to 20wt% to achieve optimum tensile strength. Higher kenaf loading gave poor interfacial adhesion between LDPE's matrix and fibre due to agglomeration of the fibre. The same observation was reported by Ku *et al.* (2011) which had blended LDPE with 3 different fibers; rice hull, flax and hardwood. Each fibre had different optimum loading (3% and 1% volume fractions) to achieve higher mechanical properties.

Meanwhile, as the second largest palm oil producer in the world with the oil palm plantation at about 5.1 million hectares, Malaysia is facing tonnage of biomass wastes from oil palm industries (SimeDarby, 2013). Empty fruit bunch (EFB), palm pressed fibre (PPF), oil palm frond (OPF) are among the biomasses that contribute to environmental pollutions. Among these biomasses, PPF that obtained after oil extraction process of palm fruit offers an extra benefit for the plastic's film manufacturers. Previous study reported that PPF has 5-6% oil residue remains in the fibre (Neoh *et al.*, 2011). This oil residue is thought could act like lubricant to assist uniform fibre dispersion across the LDPE matrix during the film blown process. However, the biggest problem when dealing with natural fibre is its hydrophilic nature. This character leads to poor interfacial adhesion between the fibre and the hydrophobic polymer matrix. This incompatibility causes the fibre to aggregate and induces stress concentration area thus, giving poor mechanical properties and dimensional stability of composite materials (John *et al.*, 2008; Hashim *et al.*, 2012).

Mercerization or chemical modification is one alternative solution to improve the compatibility between polymer and natural fibre. This treatment has changed the chemical constituent behavior of the fibre during swelling process due to relaxation of crystalline structure of the fibre. The swelling reaction leads new formulation of lattice, where the distance of molecule cellulose increases and filled with water molecule, thus converting the OH-groups to O-Na groups (Hashim *et al.*, 2012). The treatment increases the wettability, surface roughness and reduces moisture absorption in natural fibre. These enhance the strength and toughness of system (John *et al.*, 2008; Hashim *et al.*, 2012).

Apart from mercerization, coupling agent silane is also utilized in many biocomposite systems to improve the interfacial adhesions by forming crosslink networks between the fibres and the polymer matrices. Abdelmouleh *et al.* (2007) reported that silanol reacts with hydroxyl group of cellulose, reduces the hydrophobicity and increases the compatibility between fibre and polymer matrix. In this study, 3-aminopropylmethoxysilane (APM) was used as coupling agent with fixed amount at 5wt%. Another important component in plastic film manufacturing is plasticizer. Plasticizer such as glycerol is used to lower the glass transition temperature (T_g) of the system and enhances flexibility, which is important during the film blowing process (Mutuana *et al.*, 2007). In this project, the amounts of glycerol were varied at 1wt% and 2 wt%. Effects of glycerol against the amount of PPF were investigated in terms of flexibility, degree of crystallinity and morphology.

1.2 Problem Statement

The common technique to produce semi-biodegradable plastic is by blending the resin with natural fibre such as oil palm biomass. In this research, palm pressed fibre (PPF) from oil palm biomass was chosen to be incorporated in LDPE to form biocomposite film. However, like other natural fibres, PPF tends to agglomerate in the LDPE's matrix due to its hydrophilic nature, thus giving poor fibre distribution across the matrix. Mercerization process with alkali such as sodium hydroxide

(NaOH) has proven to improve this drawback. The process is strongly influenced by three parameters, namely concentration of NaOH, soaking duration and soaking temperature (Hashim *et al.*, 2012). Interestingly, PPF contains 5-6% oil residue in the fibre. The oil is thought could assist the dispersion of fibre in the matrix by acting like lubricant. Therefore, the effect of mercerization on the oil content is crucial in this project. Two mercerization parameter were considered; NaOH concentration and soaking duration. The percentage of remaining oil residue in the fibre before and after the treatment was calculated.

Apart from good effort to produce semi-biodegradable plastic film for environmental sustainability, the incorporation of natural fibre into plastic can affect certain properties of the system such as flowability, tensile strength, degree of crystallinity, and morphology (Pasquini *et al.*, 2007; Garg and Jana 2007; Chandra *et al.*, 1998). Amount of fibre loading and plasticizer play important role especially in plastic film production in terms of processability. The addition of natural fibre increased the mechanical properties of biocomposite but must be limited at certain optimum value to avoid phase separation between the polymer matrix and the fibre (Pasquini *et al.*, 2007). Meanwhile plasticizer such as glycerol helps in reducing the viscosity and increase flexibility, thus making the molten plastic easy to be blown into thin sheet. On the other hand, higher amount of glycerol i.e. 15-20 wt% caused drastic reduction in tensile and modulus strength (Garg and Jana 2007). Therefore, it is important to investigate the flowability, tensile strength and morphology of the biocomposite against the amount of PPF and glycerol.

Because this project is aimed to produce semi-biodegradable plastic film, the ability of the product to degrade must be measured. Although PPF had undergone mercerization process, study showed that the fibre still contained some hydroxyl (OH) group (Hashim *et al.*, 2012). This group is responsible to form hydrogen bonding with water moisture from environment. The absorbed water helps to increase the rate of biodegradation as the wet and moist conditions are suitable for the growth of soil microorganisms. In this study, the absorption rate of water and biodegradability of biocomposite were measured and the values were compared with unfilled LDPE film.

1.3 Objective

Few objectives need to be achieved as the followings;

1. To investigate the effects two treatment parameters i.e. NaOH concentration and soaking duration towards the content of oil residue of PPF and thermal property before and after the treatment.
2. To select the best percentages of PPF and glycerol loadings by investigating the flowability of LDPE/PPF biocomposite film.
3. To analyze the effects of PPF and glycerol loadings on the properties of LDPE/PPF biocomposite film in terms of tensile strength, thermal property, surface morphology, water absorption and biodegradability.

1.4 Scope of Study

Following are scopes of study in order to achieve the objectives.

1. Alkali treatment of PPF by using sodium hydroxide (NaOH) at various concentrations (1%, 3%, 5%, 8% and 10%) and soaking durations (1 hr, 4 hrs, 8 hrs, 24 hrs and 48 hrs). Size of fibre was set at 63 μm .
2. Biocomposite film was prepared by premixing LDPE, PPF, silane and glycerol by using high speed mixer. Later, the premixture was compounded with a single screw extruder followed with blown film extrusion process to produce thin film.
3. Testing and characterization.

The treated PPF underwent several characterizations including:

- i. Oil content analysis using soxhlet extractor and rotary evaporator.
- ii. Fourier Transform Infra Red (FTIR) analysis.
- iii. Differential Scanning Calorimetry (DSC) analysis.

The LDPE/PPF biocomposite films that containing various amounts of PPF and glycerol underwent several test and characterization including:

- i. Rheology analysis by using capillary rheometer.

- ii. Flow behavior by Melt Flow Index (MFI) analysis.
- iii. Fourier Transform Infra Red (FTIR) analysis
- iv. Tensile Test according to ASTM D882 Standard Test Method for Tensile Properties of Thin Plastics.
- v. Water Absorption test according to ASTM D570
- vi. Biodegradability test by soil burial test.
- vii. Differential Scanning Calorimetry (DSC) test to determine melting temperature (T_m) and degree of crystallinity.
- viii. Scanning Electron Microscopy (SEM) with 500X magnification to analyze the PPF dispersion within LDPE matrix.

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