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Review

The use of antioxidants in vegetable oils – A review

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The shelf life of vegetable oils in food uses and their applicability in industrial situations is greatly dependent on their oxidative stabilities. Methods of improving oxidative stability values currently available include genetic modifications, compositional changes via chemical means, as well as the inhibition of oxidation by means of substances known as antioxidants. This paper reviews the properties of vegetable oils which predispose them to auto oxidation reactions, explores the nature and mechanisms of the operation of antioxidants in brief, and finally summarizes current research efforts geared towards identifying and evaluating the effectiveness and commercial promise of less toxic new antioxidants derived as natural extracts from various plant species.

Key words: Vegetable oils, oxidative stability, natural extracts.

INTRODUCTION

By general definition, an antioxidant is a molecule capable of slowing or preventing the oxidation of other molecules (wikipedia.org). In general, oxidation reactions in organic systems produce free radicals which start off chain reactions. Antioxidants terminate these chain reactions by probably removing free radical intermediates, or by being oxidized themselves. Hence, most antioxidants are often reducing agents (wikipedia.org).

The role of free radicals in oxidation is universal. In living systems, free radicals, which oftentimes incorporate the highly reactive oxygen in their structure, are known to attack healthy cells of the body in oxidation reactions that result in their loss of structure and function. Antioxidants in living systems are capable of stabilizing or deactivating free radicals, thus controlling their formation (Percival, 1996). Auto oxidation occurring in food systems, and various substances employed in industrial and domestic applications is also recognized as occurring via a free radical chain reaction mechanism (Bennion, 1995). The most common molecules which are attacked by oxidation are unsaturated fats, which turn rancid as a result (Robards et al., 1988).

The chemical composition of fats and oils which confers on them specific properties has made them suitable for use as foods, fuels and lubricants. They are naturally occurring substances which consist predominantly of esters of fatty acids and glycerol. Their sources include substances of vegetable, animal and marine origin. Vegetable oils in particular are natural products of plant origin consisting of ester mixtures derived from glycerol with chains of fatty acid contain about 14 to 20 carbon atoms with different degrees of unsaturation (Ferrari et al., 2004). Knowledge of their specific characteristics provides insight into their being prone to auto oxidation reactions, thus necessitating the use of antioxidants as one major means of ensuring oxidative stability.

VEGETABLE OILS - GENERAL PROPERTIES

Vegetable oils are obtained from oil containing seeds, fruits, or nuts by different pressing methods, solvent extraction or a combination of these (Bennion, 1995). Crude oils obtained are subjected to a number of refining processes, both physical and chemical. These are detailed in various texts and articles (Bennion, 1995), (Fennema, 1985).

There are numerous vegetable oils derived from various sources. These include the popular vegetable oils: the foremost oilseed oils - soybean, cottonseed, peanuts and sunflower oils; and others such as palm oil, palm kernel oil, coconut oil, castor oil, rapeseed oil and

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others. They also include the less commonly known oils such as rice bran oil, tiger nut oil, patua oil, koème oil, niger seed oil, piririma oil and numerous others. Their yields, different compositions and by extension their physical and chemical properties determine their usefulness in various applications aside edible uses.

Cottonseed oil was developed over a century ago as a byproduct of the cotton industry (Bennion, 1995). Its processing includes the use of hydraulic pressing, screw pressing and solvent extraction (Wolf, 1978). It is classified as a polyunsaturated oil, with palmitic acid consisting 20 – 25%, stearic acid 2 – 7%, oleic acid 18 – 30%, and linoleic acid 40 – 55% (Fennema, 1985). Its primary uses are food related – as salad oil, for frying, for margarine manufacture, and for manufacturing shortenings used in cakes and biscuits.

Palm oil, olive oil, cottonseed oil, peanut oil, and sunflower oil amongst others are classed as Oleic – Linoleic acid oils seeing that they contain a relatively high proportion of unsaturated fatty acids, such as the monounsaturated oleic acid and the polyunsaturated linoleic acid (Dunn, 2005; Gertz et al., 2000). They are characterized by a high ratio of polyunsaturated fatty acids to saturated fatty acids. As a consequence of this, they have relatively low melting points and are liquid at room temperature. Iodine values, saponification values, specific compositions and melting points in addition to other physical properties have been determined and are widely available in the literature (Williams, 1966), (Oyedeji et al., 2006).

Other oils fall under various classes such as the erucic acid oils which are like the oleic linoleic acid oils except that their predominant unsaturated fatty acid is erucic acid (C_{22}). Rapeseed and mustard seed oil are important oils in this class. Canola oil is a type of rapeseed oil with reduced erucic acid content (Applewhite, 1978). It is a stable oil used in salad dressings, margarine and shortenings. Soybean oil is an important oil with numerous increasing applications in the modern day world. It is classed as a linolenic acid oil since it contains the more highly unsaturated linolenic acid. Other oils include castor oil (a hydroxy-acid oil) which contains glycerides of ricinoleic acid (Erhan et al., 2006). Also worthy of note is that coconut oil, which unlike most vegetable oils, is solid at room temperature due to its high proportion of saturated fatty acids (92%) particularly lauric acid. Due to its almost homogenous composition, coconut oil has a fairly sharp melting point (Bennion, 1995).

VEGETABLE OILS - LUBRICANTS, BIO-FUELS, AND TRANSFORMER COOLANTS

The application of vegetable oils and animal fats for industrial purposes, and specifically lubrication has been in practice for many years. Inherent disadvantages and the availability of inexpensive options have however brought about low utilization of vegetable oils for industrial lubrication (Honary, 2004). When applied in the science of tribology, vegetable oils fall under the class known as fixed oils (Gunther, 1971). They are so named because they do not volatilize without decomposing. Prior to recent developments, vegetable and animal oils in tribology have functioned mainly as additives to mineral lubricating oil formulations, although in some cases they are applied exclusively, or in blends. For instance, tallow (acidless) has been used as an emulsifying agent for steam cylinder oils, while castor, peanut and rapeseed oils have been used in blends with mineral oils to improve lubrication performance. Palm oil has been used in isolation as a fluxing dip in the tin plating of steel, while olive oil has applications as a yarn lubricant (Gunther, 1971).

Reasons for the use of vegetable oils in the science of lubrication abound. Their superior lubricity and emulsifying characteristics increase their desirability as additives to the cheaper but less effective mineral oil based lubricants. Their superior lubricity in industrial and machinery lubrication sometimes even necessitates the addition of friction materials in tractor transmissions in order to reduce clutch slippage (Honary, 2004).

Other advantages that encourage the use of vegetable oils include their relatively low viscosity-temperature variation; that is their high viscosity indices, which are about twice those of mineral oils (Honary, 2004). Additionally, they have low volatilities as manifested by their high flash points (Honary, 2004). Significantly, they are environmentally friendly: renewable, non toxic and biodegradable (Howell, 2007). In summary, engine lubricants formulated from vegetable oils have the following advantages deriving from their base stock chemistry:

- ✓ Higher Lubricity resulting in lower friction losses, and hence more power and better fuel economy.
- ✓ Lower volatility resulting in decreased exhaust emissions.
- ✓ Higher viscosity indices.
- ✓ Higher shear stability.
- ✓ Higher detergency eliminating the need for detergent additives.
- ✓ Higher dispersancy.
- ✓ Rapid biodegradation hence decreased environmental / toxicological hazards (Erhan et al., 2002).

In a comparison of palm oil and mineral based lubricants, palm oil based lubricants were found to be more effective in reducing the hydrocarbon and carbon monoxide emission levels, among other things (Masjuki et al., 1999).

Vegetable oils have also been identified as having a lot of potential as alternative diesel engine fuels (Kayisoglu et al., 2006). This is supported by an interest in a cleaner environment, as well as the increasing cost of mineral deposit based energy (Howell, 2007). Based on the potential availability to meet demand, soybean, peanut and sunflower oils have been identified as the most promising fuel sources (Kayisoglu et al., 2006). When used as a fuel, the term "biodiesel" is applicable.

Biodiesel is defined strictly as *"…the mono alkyl ester (usually methyl ester) of renewable fats and oils…"* (Howell, 2007). It consists primarily of long chain fatty acid esters, produced by the transesterification reaction of vegetable oils with short chain alcohols. Distinct advantages of biodiesel include a high flash point of over 100°C, excellent lubricity, a BTU content comparable to that of petrol diesel, and virtually no sulfur or aromatic content. Above all, biodiesel is non-toxic and biodegradable (Howell, 2007).

Results from investigating performance of vegetable oils in blends with diesel indicate that blending up to 25 percent biodiesel (sunflower) with mineral diesel has no adverse effect on performance. (Kayisoglu et al., 2006)

Vegetable oils have also been applied as transformer coolant oils and have been found to conform to all industry standards with performances and cost profiles comparable to the conventional mineral oils applied in transformer cooling (ABB Inc., 2002). Transformer oil products have been produced from soybean oils as well as castor oils (Honary, 2004).

Whether applied for lubrication purposes or as biodiesel or as transformer cooling fluid, one of the major challenges in the utilization of the more environmentally friendly vegetable oils is their poor oxidative stability (Honary, 2004), (Howell, 2007).

Combating the issue of oxidative instability in vegetable oils for industrial use is a continuing research area. In the United States, for instance, three avenues are being pursued. These are:

- ✓ Genetic modification of oils to give higher mono unsaturated compounds
- ✓ Chemical modification
- ✓ The use of various additives and property enhancers (Howell, 2007).

Genetic modification has been made possible by recent advances in biotechnology. DuPont Technology has developed a soybean seed that presents 83% oleic acid as against having the more unsaturated linolenic acid as the major constituent. This new seed provides oils that show about 30 times the oxidative stability and viscosity stability of the conventional oil. High oleic varieties of rapeseed, canola and sunflower seed oils are increasingly being used as base stocks for lubricant formations (Honary, 2004).

Chemical modifications involve the partial hydrogenation of the vegetable oil and a shifting of its fatty acids (Honary, 2004). In one study, epoxidized soybean oil was chemically modified with various alcohols in the presence of sulfuric acid as a catalyst. Better performance was recorded (Hwang et al., 2001).

The use of additives known as antioxidants to control the development of oxidative rancidity has been applied in the US since 1947 (Bennion, 1995). They still remain one of the most efficient and cost effective ways to improve the oxidative stability of oils in both domestic and industrial conditions.

AUTO OXIDATION AND OXIDATIVE STABILITY IN VEGETABLE OILS

As just stated, the major drawback in the application of vegetable oils for industrial uses is the fact that in their natural forms, they lack sufficient oxidative stability. By definition, the oxidative stability of oil is a measure of the length of time taken for oxidative deterioration to commence. On a general level, "the rates of reactions in auto-oxidation schemes are dependent on the hydrocarbon structure, heteroatom concentration, heteroatom speciation, oxygen concentration, and temperature (Ferrari et al., 2004).

If untreated, oils from vegetable origin oxidize during use and polymerize to a plastic like consistency (Honary, 2004). Even when they are not subjected to the intense conditions of industrial applications, fats and oils are liable to rancidity (Eastman Chemical Company, 2001; Morteza- Semnani et al., 2006). This happens more so in fats that contain unsaturated fatty acid radicals (Charley, 1970). Indeed the oxidisability of a vegetable oil is dependent on the level of unsaturation of their olefinic compounds.

In general terms, oxidative rancidity in oils occurs when heat, metals or other catalysts cause unsaturated oil molecules to convert to free radicals. These free radicals are easily oxidized to yield hydroperoxides and organic compounds, such as aldehydes, ketones, or acids which give rise to the undesirable odors and flavors characteristic of rancid fats (Eastman Chemical Company, 2001). The role of peroxides is exploited in monitoring oxidative deterioration by measuring peroxide values (POV) (Mochida et al., 2006).

Lipid oxidation occurs via auto oxidation or lipoxygenase catalysis. Auto oxidation refers to a complex set of reactions which result in the incorporation of oxygen in lipid structures. Auto oxidation reactions are seen to progress more rapidly in oils that contain predominantly unsaturated fat molecules; other relevant factors include the presence of light, transition metal ions, oxygen pressure, the presence or absence of antioxidants and pro oxidants, temperature and moisture content. Auto oxidation reactions occur at an increasing rate after the initial induction period. This behavior can be explained by assuming that oxidation proceeds by a sequential free radical chain reaction mechanism.

Relatively stable radicals that can abstract hydrogen atoms from the allylic methylene groups in olefinic compounds are formed. Hence auto oxidation is a radical induced chain reaction which proceeds through the traditional stages of initiation, propagation and termination. Detailed proposed mechanisms for these free radical chain reactions are available in literature (Fennema, 1985).

Lipoxygenases are metal proteins with an iron atom as the active center. They catalyze the oxidation of unsaturated fatty acids to hydroperoxides as with auto oxidation. Enzyme activation usually occurs in the presence of hydroperoxides, even though enzyme catalyzed oxidation can occur even in the absence of hydroperoxides (Fennema, 1985). As earlier stated, the more unsaturated the fatty acid involved is, the greater its susceptibility to oxidative rancidity. For instance, the linolenic acid esters present in soybean oil (with twice the unsaturation as monounsaturated esters) is particularly sensitive to even oxidation of the slightest kind, commonly referred to as flavor reversion, resulting in beany, grassy or painty flavors (Wolf, 1978). A highly saturated fatty acid level is confirmed to be of benefit in terms of storage ability when compared to more unsaturated vegetable oils (Ferrari et al., 2004). Indeed, the tendency of an oil to combine with oxygen of the air and become gummy (known as drying) is measured with the iodine number, which in fact is merely a measure of the level of unsaturation of the oil in question (a higher iodine number will indicate higher unsaturation seeing that iodine is absorbed primarily by the mechanism of addition to the double bonds characteristic of unsaturation) (Gunther, 1971).

Based on studies by Toshiyuki. (1999), the oxidative stability of refined vegetable oils is found to be determined considerably by the fatty acid composition, the tocopherols content and the carbonyl value (Toshiyuki, 1999). When observed at frying temperatures, it is seen that in general, non-refined oils prove to have a better stability than refined oils (Gertz et al., 2000). This could be attributed to the fact that refining steps, in particular deodorization, remove a percentage of the tocopherols, which act as natural anti-oxidants in vegetable oils (Applewhite, 1978). Corn oil has a better stability than soybean oil, while rapeseed oil is seen to give a better performance than olive oil. This can be explained in terms of their compositions (Isbell et al., 1999).

When investigated at a temperature of 110°C, vegetable oils still show the trend of increased stability in the unrefined state than when refined. Meadow foam oil is reported as the most stable oil in the study conducted by Isbell et al. (1999). High oleic sunflower oil and crude jojoba oil also had good values of oxidative stability (Isbell et al., 1999).

Other studies indicate that the presence of free fatty acids has a pro-oxidant effect on vegetable oils (Frega et al., 1999). Hence refining practices are important, seeing that improper handling and raw material abuse can result in the stimulation of enzymatic activity which could produce free fatty acids (Applewhite, 1978).

Further investigations on manufacturing practices also reveal research which indicates the importance of the solvent used in the extraction of vegetable oils. Traditional solvents utilized such as hexane or petroleum ether have the characteristic of extracting only non-polar species. Isopropanol however, as documented by Oyedeji et al. (2006) would extract some polar and high molecular weight compounds. Among these compounds are the natural antioxidants and pigments in oilseeds which presence lead to extended shelf life and hence better oxidative stability (Oyedeji et al., 2006).

ANTIOXIDANT OPERATION AND MECHANISMS

The word anti-oxidant is used in a general sense to refer to any type of chemical agent which inhibits attack by oxygen or ozone (Scott, 1965). As applied to vegetable oils, anti-oxidants are compounds which interrupt the oxidation process by preferentially reacting with the fat radical to form a stable radical which does not quickly react with oxygen (Eastman Chemical Company, 2007). When the reference is to food uses, they are grouped as a food additive which has the effect of increasing the shelf life of foods by protecting them against deterioration caused by oxidation which leads to rancidity and color changes (FSANZ, 2006).

Antioxidants function either by inhibiting the formation of free alkyl radicals in the initiation step or by interrupting the propagation of the free radical chain (Ohio State University, 2008). In truncating the propagation step, the antioxidants function as hydrogen donors.

Generally, the most popular antioxidants are hydroxylphenol compounds with various ring substitutions. They are characterized by possessing low activation energies for the hydrogen donation process. The antioxidant radical which results is stabilized with its local electrons delocalized; hence antioxidant free radicals do not readily initiate other free radicals. They rather even react with lipid free radicals to form stable and complex compounds (Ohio State University, 2008). In investigating phenolic antioxidants, it is found that their antioxidative capabilities bear a relationship to the number of phenol groups occupying 1,2 or 1,4 positions in an aromatic ring as well as to the volume and electronic characteristics of the ring substituents present (Fennema, 1985). In elucidating the mechanism of oxidative inhibition, it is generally established that anti-oxidants function as oxygen interceptors in the oxidative process thereby breaking the chain reaction that perpetuates the process (Bennion, 1995).

Common phenolic antioxidants include Butylated Hydroxyanisole (BHA), Butylated Hydroxytoluene (BHT), Propyl Gallate (PG) and Tertiary Butyl Hydroquinone (TBHQ). Simply put, conventional antioxidant activity involves the donation of hydrogen to free radicals followed by the formation of a complex between a lipid radical and the antioxidant radical formed as a result of the hydrogen loss. Here the antioxidant radical functions as a free radical acceptor. The general scheme is presented below:

$$R\bullet + AH \longrightarrow RH + A\bullet$$

$$RO\bullet + AH \longrightarrow ROH + A\bullet$$

$$ROO\bullet + AH \longrightarrow ROOH + A\bullet$$

$$R\bullet + A\bullet \longrightarrow RA$$

$$RO\bullet + A\bullet \longrightarrow ROA$$

$$ROO\bullet + A\bullet \longrightarrow ROOA$$

$$\text{Antioxidant} + O_2 \longrightarrow \text{Oxidized Antioxidant}$$

(Ohio State University, 2008).

Mention must be made of synergists – substances which increase the effectiveness of a primary antioxidant. Certain metallic ions such as copper and iron act as pro oxidants, catalyzing the oxidation process. Such metal ions can be sequestered or chelated by certain organic acids. They effectively contribute to lower transition metal activity. Synergists are not as effective when used alone; rather, they work best when combined with an antioxidant. Examples of such compounds are citric acid, phosphoric acid and some of their derivatives (Fennema, 1985). Synergism has been studied, not just in relation to antioxidants alone, but in relation to combinations of antioxidants, anti wear and other additives (Sharma et al., 2007). An example of a synergist used in conjunction with phenolic antioxidants is citric acid (Bennion, 1995).

ANTIOXIDANTS AND STABILITY OF VEGETABLE OILS

Numerous experimental works have established the positive effect of anti-oxidants on the oxidative stability of vegetable oils for both edible uses and industrial uses. An important class of anti-oxidants consists of the phenolic compounds Butylhydroxyanisole (BHA), Butylhydroxytoluene (BHT), Propyl Gallate, and Tert-butyl Hydroquinone (TBHQ). Their use in vegetable oils meant for domestic and industrial processes is widespread.

Vegetable oils in their natural form possess constituents that function as natural antioxidants. Amongst them are ascorbic acids, α-tocopherole, β-carotene, chlorogenic acids and flavanols (Ullah et al., 2003). Tests conducted to investigate the effectiveness of natural anti-oxidants contained in red pepper oil added to soybean and sunflower oils indicate that they provide variable protection against light induced auto-oxidation. Measuring fatty acid formation and the measurement of peroxide values as a means of monitoring oxidation, results indicate an inhibitive effect on oxidation (Ullah et al., 2003). In another study which monitored the inhibitive action of tocopherols on rapeseed and palm kernel oils by measuring the presence of the oxidation product, monoaldehyde, indicate some measure of protection provided by these natural anti-oxidants (Emanuel et al., 1967).

In the above mentioned study on the inhibitive effect of natural antioxidants contained in red pepper oil, it was additionally observed that the phenolic anti-oxidant Butylated Hydroxytoluene (BHT) shows more effectiveness generally than natural anti-oxidants (Ullah et al., 2003).

In the work done by Robert (2005), the common phenolic anti-oxidants were tested for their effectiveness in improving the oxidative stability of biodiesel obtained from soybean oil. Dunn monitored the oxidative stability by means of pressurized differential scanning calorimetry (P-DSC). For both static and dynamic conditions, improvements in oxidative stability are observed with the application of anti-oxidants, which included BHA, BHT, TBHQ, propyl gallate (PrG) and α-tocopherol. The work of Dunn further showed that the relative effectiveness of the different anti-oxidants differed for static and dynamic conditions, although all showed superior performance when compared with α-tocopherol (Dunn, 2005). In addition to these observations, the relationship between anti-oxidant loading and oxidative stability was established. Increased loading led to increased anti-oxidant activity, with Propyl Gallate showing a slight advantage at lower loadings, and BHA showing the greatest effectiveness at amounts of about 5000 ppm. Little improvements in effectiveness were recorded for increased loading with α-tocopherol. Interestingly, in another study of the effects of tocopherols on the oxidative stability of soybean oil by Jung et al. (1990) concentrations of the tocopherols that were higher than what was determined as optimum resulted in pro-oxidant effects (Jung et al., 1990). The general conclusion is that up to a point, increased loading of antioxidants is beneficial in terms of improving oxidative stability. What remains to be answered is how the environmental benefits of using vegetable oils industrially are affected by the presence of these anti-oxidants.

Ruger et al. (2002) investigated the ability of various anti-oxidants and chelators (including TBHQ, BHT, hydroquinone and ascorbyl palmitate) to delay viscosity increase in soybean oil brought about by auto-oxidation.

Concentrations of 0.01 and 1.28% were used, and the results again confirmed the effectiveness of the given anti-oxidants, with TBHQ being the most effective at the two temperatures (Ruger et al., 2002). Other studies have investigated synergism amongst anti-oxidants (Sharma et al., 2004).

A recent area of interest in antioxidant research is concerned with finding effective replacements for the conventional synthetic antioxidants from among various natural extracts from plant species which are seen to possess antioxidant properties. Such research is in the main prompted by the reported possibility of synthetic antioxidants having adverse health effects on humans exposed to them. Specifically, they are known to contribute to liver enlargement and an increase in microsomal activity (Khanahmadi et al., 2006; Morteza- Semnani et al., 2006).

Maduka et al. (2003) investigated the effectiveness of a Nigerian alcoholic beverage additive, *Sacoglottis gabonensis* stem bark extract as an antioxidant for common stored vegetable oils. Inhibition of lipid peroxi-dation was found to be comparable to inhibitions obtained with treatment with vitamins C and E (Maduka et al., 2003).

The *Ferulago angulata* plant indigenous to the west of Iran also has proven antioxidant properties. Experimental studies documented indicate that this plants essential oil and extract begins to show preservative properties on vegetable oils at a minimum concentration of 0.02%. In fact, it even shows more effectiveness that TBHQ at concentrations of 0.5% (Khanahmadi et al., 2006).

When evaluated by measuring reducing power, ability to inhibit linoleic acid peroxidation, and 2,2-diphenyl-β-picrylhydrazyl radical scavenging activities, the alkaloid extracts of *Fumaria capreolata* and *Fumaria bastardii* demonstrated strong total antioxidant activity, with effectiveness marginally less than that of the common synthetic antioxidant butylated hydroxyanisole, and better than quercetine and caffeine. These species have wide distribution in the Mediterranean region and have a reputation for effectiveness in treating hepatobiliary disfunction and gastrointestinal disorders via local therapies (Maiza et al., 2007).

Methanolic extracts of *Phlomis bruguieri, P. herba-venti, P. olivieri, Stachys byzantine, S. inflata, S. lavandulifolia* and *S. laxa* were tested in sunflower oil stored at 70°C for antioxidant effectiveness, using peroxide values as a measure. Comparisons included samples containing BHA. Highest effectiveness in stabilizing sunflower was obtained from methanolic extracts of *P. bruguieri,* and *S. laxa.* These tests and their findings suggest strongly the possibility of having in these plants a viable source of natural antioxidants of high performance (Morteza- Semnani et al., 2006).

CONCLUSION

The applicability of vegetable oils as foods and industrial fluids is diminished by their relatively low oxidative stability. Genetic and chemical modifications to improve this property are still in the developmental stage, while the use of antioxidants is well established as a relatively cheap and effective method of obtaining oxidative stability improvements. The ideal antioxidant would combine effectiveness in low concentrations with minimal toxicity. Research is still ongoing to investigate the applicability of natural extracts found to possess antioxidant activity, and is yielding encouraging results. The next generation of safer, environmentally friendly, effective antioxidants is undoubtedly soon to come.

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