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PRODUCING GEL WITH VARIOUS INGREDIENTS: A REVIEW

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1. INTRODUCTION

Health problems are one of the most vulnerable things that occur in the world. The reason is, most people don't choose the right food in their daily lives. Inappropriate food choices can later cause one of the diseases, namely obesity. Obesity has become a public health problem that is becoming increasingly prominent in various countries, causing several food industries to be involved in developing new products with characteristic tastes and textures that are much desired by the public regardless of the high amounts of fat, sugar and salt content [1].

The association between consumption of hydrogenated unsaturated fats and solid fats is not only associated with obesity, but is also associated with increased cholesterol levels, risk of malignant tumors, and heart disease [2]. As a result, the demand for foods packed with nutrients containing health-beneficial compounds that can prevent the onset of these diseases is increasing [3]. The World Health Organization (WHO) identifies obesity as a major health concern worldwide, based on their data. A worldwide study in 2020 indicated a significant public health challenge: over 650 million adults (aged 18+) were classified as obese. Additionally, nearly 39 million children under 5 years old were found to be overweight or obese [4]. These figures highlight the importance of public health awareness and the importance of efforts to prevent and treat obesity in various age groups.

Certain goods, such as ice cream, yogurt, cheese, and processed meats, contain significant levels of saturated fat. However, efforts to minimize the total quantity of fat in a diet may cause modify in the sensory, structural, and rheological properties of the meal. Furthermore, lipids play a significant role as transporters of vitamins and critical fatty acids [5]. Some ingredients can be used as fat replacement gels in a variety of applications. For example, Carrageenan is used as a fat substitute in frozen yogurt, producing a thicker and softer texture with a fresher taste compared to frozen yogurt which contains high fat [6]. Vegetable proteins such as soybeans and peas are used as fat substitutes in burgers, resulting in a tender texture and savory taste with a lower saturated fat content compared to beef burgers [7].

Gel is a semi-solid system formed from small inorganic particles or large organic molecules that is capable of permeating liquids [8]. The gel is a semi-solid preparation dispersion of small and large molecules in aqueous liquid carriers.. Gel has several distinctive properties, namely; Gels resist flow and maintain their shape [9], The texture of a gel depends on the type of gel-forming material and its concentration. They can be smooth, soft, or brittle, gels can aid in stabilizing mixtures of oil and water [10], and natural gels are often free of cholesterol and trans fats, rich in fiber and protein, and have a low glycemic index [11] [10]. This review aims to investigate the use of healthy natural ingredients as fat substitutes in gel preparation and to examine the effects of such substitutions.

2. DEFINITION OF GEL

Gels are a type of material made from long, chain-like molecules (polymers) that can trap large amounts of liquids (hydrogels), gases (aerogels), or even oils (oleogels) within their three-dimensional structures. These unique properties make gels interesting to scientists working in many different fields [12] [13]. Since the early $20th$ century, gels have been utilized as biomaterials for injectable soft-tissue fillers derived from hyaluronic acid or for wound healing in the case of gels with an alginate foundation [14].

Gels, also known as solidified liquids, are a type of soft, solid, semi-solid material that holds a lot of water in its structure. Gels can be divided into two primary categories based on the sort of bond they have: physical and chemical gels. Physical gels are linked non-covalently, while chemical gels are joined covalently. In the food industry, gels have become more significant since they provide consumers convenience in the form of desserts, jam, jelly, and quick-setting products, among other products. Gels are enjoyed because of their flavor, low-calorie content, nutritional value, and satisfaction [15].

Gel formation involves the interaction of various forces and requires certain triggers, namely temperature, pressure, or solvent quality. Covalent interactions, such as disulfide bonds [16], and non-covalent interactions, such as hydrophobic interactions, hydrogen bonds, and electrostatic interactions, play an important role in the process of protein gel formation and stabilization [17]. There are various methods used to characterize gels. One of these is the rheological characterization of the gel. Rheology, a study that investigates how materials respond to applied forces, can be done utilizing minor distortions and several types of rheometers. Alternatively, examine the gel's mechanical qualities (large distortion). When tiny distortions are used, popular metrics examined include storage modulus (G'), which indicates the gel's solid qualities, and loss modulus (G''), which reflects the gel's liquid-like characteristics. In tests with large deformations, numerous metrics are acquired, including fracture stress, which reflects the strength of the gel; fracture strain, which reveals the level of gel brittleness; and Young's modulus, which depicts the stiffness of the gel [18].

The physical attributes of gels, such as Young's modulus indicating material stiffness, recoverable energy and storing energy elastically in the structure, critical stress intensity factor, and serum release, are all of great importance in the context of food and model systems. All these factors can impact sensory perceptions related to food and food products [19]. When a food gel experiences deformation, the noncovalent bonds in the gel will lose energy due to bending and breaking. In addition, these bonds will reform with low energy. Certain food additives, known as gelling agents, are incorporated into food products to modify their texture. Common examples of gelling agents include pectin, starch, natural gums, and agar-agar. These additives function by increasing the viscosity (thickness) and promoting the formation of a stable gel network within the food, ultimately influencing its overall texture [15].

Figure 1. A visual representation outlining the transition from dispersal of proteins within a solution to the formation of aggregates, which subsequently amalgamate to create a gel network [18].

For our purposes here, a gel is a substance where microscopic particles clump together, creating a 3D web that fills the entire container. Heat-induced (thermal) gels emerge when native proteins are partially unfolded, exposing nonpolar residues and causing aggregates to cluster and form a spatial gel network. When we make protein gels, using more protein makes the gel stronger. This is because the protein molecules stick together better, forming a more stable network. As a result, the gel can hold its own shape without any help [18]. Think about building a sandcastle. You want the sand to stick together well enough to hold its shape (like a self-supporting gel), otherwise your creation crumbles easily. In food, this is like how well a jello holds together. The better the

"stickiness" (fracture properties), the easier it is to cut and handle the food without it falling apart [18]. Making a gel is like making jelly. Different ingredients need to come together in a certain way, and then you need a trigger, like adding cold water, to make everything set and form a wobbly network. Strong bonds (like tiny handcuffs) [16] and weaker attractions (like magnets and sticky patches) between protein molecules play a key role in making and keeping protein gels stable. Figure 1 shows a picture of this whole process.

Forming Materials

2.1. Polysaccharides

Certain sugars with long chains (polysaccharides) can only form gels (thicken and trap water) when used at a specific amount (critical concentration). The leftover polysaccharides can still be used in food to thicken and stabilize things, even if they don't form gels. Compared to proteins, these critical concentrations for polysaccharides are usually lower (you don't need as much). After the polysaccharides soak up water (hydration process), the chains start to connect with each other (crosslinking) at specific points, forming a strong network. When there's enough polysaccharide and enough crosslinking, the liquid mixture (dispersion) turns into a gel with a solid structure. These connection points (junction zones) can be different depending on the type of polysaccharide and how the gel is formed. Some polysaccharides need heating and then cooling to form gels (heat-set gels), while others can form gels at room temperature (cold-set gels) using specific minerals (cations), changing the acidity (pH), or adding other ingredients (cosolutes). There's even a special polysaccharide called curdlan that can form both heat-set and cold-set gels, we'll talk about some of the most common polysaccharide gels in the next sections $[10]$.

There are several ways to achieve polysaccharide gelation, however it usually depends on the polymer structure, or the source of the polysaccharide. Common gelation techniques include freezing and thawing cycles, pH, temperature, and salt. Different forms of temperature-induced gelation exist [14]: Gelation occurs in four different ways: (1) Thermosensitive Gelling: Gelling agents like agarose, carrageenan, and gellan form gels upon cooling from a hot solution. As the temperature decreases, the molecules interact and form a three-dimensional network, trapping water and creating a gel structure. (2) Thermosetting Gelling: In contrast, gelling agents like methylcellulose and hydroxypropylmethylcellulose require heating a solution to induce gelation. Temperature increases promote interactions between the molecules, leading to network formation and gelation. (3) Temperature-Sensitive Sol-Gel Transitions: Some gelling agents, like partially galactose-removed xyloglucan, exhibit sol-gel transitions only within a specific temperature range. Outside this range, the molecules exist in a liquid state (sol). (4) Mixed-Mode Gelation: Combining gelling agents with different temperature responses (e.g., mixing types (1) and (2)) can create gels with more nuanced temperature sensitivities. This allows for tailoring gel properties based on specific needs . Although thermal hysteresis in these gels can be controlled by ionic strength, molecular weight, or side residue profusion, they are frequently thermoreversible [20]. For instance, increasing the ionic concentration of κ-carrageenan solution broadens the thermal hysteresis by strengthening the gel and rising the melting temperature [20].

Another significant element that can cause polysaccharides to gel is salt, since many polysaccharides have polyelectrolytic properties. The most basic way that gelation can happen is by the screening of electrostatic contacts, which favors hydrophobic or hydrogen bonding connections and causes gelation, as shown in chitin fibrils and filamentous nanocellulose [21]. In this regard, pH accommodate a comparable mechanism, modifying the distribution of charged groups along the length of the filaments [22].

In Zhang's (2023) research, κ-carrageenan was used with the inclusion of rennet in the composite gel. It was found that with increasing κ-carrageenan concentration, the gel strength decreased due to reduced bonds between proteins in the gel structure. There was a slight increase in the hardness of the composite gel at a temperature of 25℃ for 12 hours when compared with the same temperature condition for 2 hours, indicating that extending the reaction time can increase cross-linking between proteins. In addition, at a temperature of 25℃, no microscopic phase separation occurred in the composite gel. The network structure remains dense with small pores when the concentration of κ-carrageenan is increased [23].

In research conducted by [24] it was stated that extraction using acid produces low methoxyl pectin which has the potential to be acted as a thickening ingredient for sugar-reduced and calorie-controlled foods. When neutral pH is applied to the Subcritical Water Extraction (SWE) method with a high extraction temperature, it produces high levels of methoxyl pectin. In this condition, pectin gel formation occurs when sugar is added at a low pH. In addition, in this research, scientists also investigated how well leftover pectin (a substance in apples) could hold together oil and water (emulsifying) after processing apples with varying levels of a molecule called methoxyl. They found that pectin with more methoxyl worked better at forming emulsions than pectin with less methoxyl. This led to emulsions with oil and water droplets that were all similar sizes (uniform particle size distribution).

In our prior research [25], About 100 times as much G′ was present in the G70 (high-amylose starch with 68% amylose content) gel as in the NCS (Normal Corn Starch) gel. In this investigation, however, the addition of G70 to the NCS gel decreased G′ while increasing the gel's flowability. The existence of PG (Pregelatinized)-HAS (High-Amylose Starch) and its rehydration procedure are responsible for this disparity. In our previous research [26], upon rehydration in 90℃ water, PG-G70 demonstrated a 7% WSI (Water-Soluble Index) and a 12 g/g WAI (Water-Absorption Index). This suggests that while most of the starch chains in PG-HAS absorb water and form tangled and aggregated micro-domains, only a small portion of the chains can redissolve in water. As a result, the added PG-G70 is not able to mix and combine with NCS starch chains in an effective manner, which hinders the creation of a uniform network. Rather, their chains group together to form micro-domains that break the original NCS network's continuity and lessen the interactions between the starch chains, which makes the starch gel more flowable. Within the NCS gel, there are two main parts: the NCS phase and the PG-G70 domains. The NCS phase itself is not very strong. However, the PG-G70 domains, formed by tangled amylose chains, are much stronger. These stronger domains help the NCS gel form better (improves formability) and become more self-supporting (higher τy). But be careful! If you add too much PG-G70, the gel becomes too runny (excessive flowability) and loses its ability to form properly. Figure 2 illustrates the structure of this NCS/PG-G70 gel network in more detail.

Figure 2. Network structure schematic design for NCS/PG-G70 gel [27].

2.2 Proteins

In the gelation process caused by enzymes, the formation of covalent protein cross-links occurs through the action of the transglutaminase enzyme. This enzyme facilitates the acyl transfer reaction between lysine residues and glutaminyl residues [28] [29] [30]. These cross-linking characteristics result in the formation of gels with different mechanical responses compared to gels formed via heat setting or acid setting. Gels produced from enzymatic processes tend to have more elastic rheological characteristics [31]. "Cold-set" gels are created by adding ions. This gel is created by preheating the dispersion, resulting in protein denaturation. In the early stages of denaturation, aggregation occurs without gelation development. These aggregates are ultimately capable for the creation of the gel network.

A study by Guo (2023) investigated how adding sodium (Na⁺) and calcium (Ca₂⁺) ions affects how mung bean protein and wheat gluten clump together (gelation) in a mixed protein gel. When these proteins come together, they usually repel each other. But the researchers found that interactions between the proteins become stronger than this repulsion as the amount of sodium and calcium ions increases, leading to a better gel. Calcium ions, in particular, seem to be better at protecting the negative charges on the protein surfaces, which also helps the proteins clump together more effectively. The study also showed that adding more sodium chloride (NaCl) and calcium chloride (CaCl₂), from 0 to 0.001 and 0.002 grams per milliliter (g/mL), respectively, made the proteins in the gel less soluble (dissolvable). The protein solubility went down from 35.11% to 30% and 30.11%, respectively [32].

When there's not much salt in a mixture of proteins forming a gel, the attraction between the protein molecules themselves gets weaker [33]. The study found that adding NaCl and CaCl₂ significantly increased the hardness, elasticity, and homogeneity of protein gels formed from mung bean protein and wheat gluten $(P < 0.05)$ [32]. Adding small amounts of sodium chloride (NaCl) and calcium chloride (CaCl₂), $(0.001$ and 0.002 grams per milliliter [g/mL] respectively) caused the biggest improvement in the gel's structure. This means the NaCl and CaCl2 helped strengthen the network of molecules within the gel. The strength of this gel (measured by something called storage modulus, G') is also affected by how often a force is applied (frequency-dependent).

In research conducted by Zheng (2023), Scanning Electron Microscopy (SEM) analysis demonstrates that the composite gel comprised of modified soybean protein and sunfish protein exhibits a more uniform morphology with a smoother surface, reduced pore size, and a denser network compared to the unmodified control [34].

In research [35] investigated Pea Protein Isolate (PPI) and Egg White Protein (EWP) gels. They found that as the amount of EWP increased in the mixture, the surface became less water-repelling (hydrophobic). This is likely because EWP itself has a low tendency to repel water. The research also showed that adding EWP increased the gel's elasticity (G'' value), reaching a peak at a ratio of PPI to EWP of 1:5. At lower EWP concentrations, the EWP distributes throughout the PPI gel, making it harder. However, when EWP is more abundant than PPI, the PPI gets dispersed within the EWP gel, also increasing hardness. Interestingly, a 1:1 ratio of PPI to EWP did not show an increase in hardness compared to other ratios This may be caused by the EWP concentration being higher than its lowest gelation concentration at this ratio, whereas the PPI concentration is not low. This increases the association between the two proteins and disrupts the formation of the protein network, thereby reducing the gel hardness [36].

2.3 Mixture of Proteins with Polysaccharides

Mixing two sugars with long chains (polysaccharides) that don't usually form gels on their own (or only weak gels due to tangled chains) can sometimes create strong gels. This is often seen when mixing xanthan with glucomannan or galactomannan, or when mixing alginate with pectin without calcium [20]. In the case of food polymers that have opposite charges, such as protein–polysaccharide, polysaccharide–polysaccharide, and protein–protein polymers, the formation of mixed gels can be attributed to a basic electrostatic complexation mechanism that can be adjusted based on many factors such as ionic strength, pH, and mixing ratio. We refer to this specific type of gels as coacervates. According to a recent assessment, protein–polysaccharide combination gelation offers several benefits over single gels, including exceptional water-holding capabilities [37]. Another benefit of mixing these gelling agents is that they can create gels with many different tiny structures (microstructures) inside. By changing these structures, scientists can design gels that feel a certain way in your mouth (texture) since how food feels when you eat it is strongly affected by its microstructure.

In research [38] found that gels formed in composite systems had a better structure when the acidity (pH) was moderate. This was determined by analyzing the gel's texture and how it flowed (rheology). Specifically, when the WPI (Whey Protein Isolate)-KGM (Konjac Glucomannan) composite system is heated to a pH of 8, it yields a gel with a generally smooth surface and a compact texture. On the other hand, after heating the WPI-KGM composite system at pH 4, a thick but slightly rough gel structure is produced. Konjac glucomannan is regarded as a nutritional component that is low in calories. Its indigestible dietary fiber, which is known to help lower cholesterol levels, alter gut microbial metabolism, and reduce body weight, is one of its key advantages [39]. Apart from its health benefits, konjac glucomannan also has various applications in the food industry as an additive. This is because of its properties as a thickener, providing texture, forming a gel, acting as an emulsifier, stabilizer, and water binder in foods such as soup, gravy, mayonnaise, and jam [40].

Gluten, the protein that gives dough its elasticity, can be classified into two categories: glutenin (30-40%) and gliadin (40-50%), based on their solubility in alcohol and water. A special technique called Fourier Transform Infrared (FTIR) spectroscopy was employed to investigate how heat alters the internal organization, particularly the secondary structure, of gluten molecules. Temperature affects how many bends and flat shapes (β-turns and βsheets) are present in the protein by breaking hydrogen bonds at high temperatures. Additionally, these high temperatures weaken the bonds between gluten and water molecules (KGM). The strength and elasticity of the dough can be measured by a property called dynamic storage modulus (G'). On the other hand, the loss modulus (G'') tells us how much energy is lost as heat when the dough is deformed, indicating how "sticky" the dough is. As the temperature increases, the slope of all dough types goes down, suggesting that the gluten molecules become more tightly linked together. At moderate temperature heating (55°C), KGM appears to strengthen the thermal denaturing effect on gluten. However, at higher heating temperatures (above 75°C), KGM actually weakens the aggregation caused by heat [41].

In research conducted by [42] showed that sugars with long chains (polysaccharides) can affect how different molecules (intermolecular) and parts of the same molecule (intramolecular) interact in complex systems. This interaction may also change how proteins behave. Increasing the concentration of KGM in SPI (Soy Protein Isolate)-KGM results in an increase in molecular strength, which supports the structural strength and stability of the gel network. Research by [43] consistent with this, showing that the presence of 5% KGM can increase the disulfide bonds in the KGM-gluten composite gel by 2.57-fold, which contributes to the stability of the gel network. [44] also stated that a number of denatured proteins are needed in the gel formation process. This is because the unfolded and denatured structure of the protein reveals hydrophobic and sulfhydryl groups, which then interact with each other, forming irreversible protein aggregations and strengthening the gel network.

Without KGM, the gluten surface has a lot of irregular holes of different sizes. But as KGM was added, the sample's overall structure became denser and more solid, and the number of holes started to decline. Nearly no holes were apparent on the sample surface when 5% KGM was present, however there was a significant quantity of chipped structure. It is possible that the 5% KGM will absorb the gluten and cause an increase in water volume, allowing the sample to become integrated into a single piece. KGM might also help gluten hold more water, which can be a problem during freeze-drying. When you freeze-dry something, the water inside turns to ice. But with KGM, the water in the less organized parts of the gluten can clump together and form large ice crystals. These big ice crystals can damage the gluten structure, causing the sample to break apart when it dries [45]. Adding KGM made gluten more elastic and cohesive, especially when up to 4% KGM was added. This might be because the KGM filled in the tiny holes (pores) within the gluten network, creating a stronger connection between gluten molecules. Interestingly, KGM also affected the chemical bonds in gluten. It reduced the number of loose "sulfhydryl groups" and increased the number of stronger "disulfide bonds." In fact, adding 5% KGM increased the amount of disulfide bonds by over two and a half times! Because KGM helps prevent these stronger bonds from breaking down, it improves the overall function and stability of the gluten structure [43].

Figure 3. This work investigates the correlations between power-law strength exponent (n), fractal dimension (df), and gel strength (Sg) as they depend on the concentration of konjac glucomannan within SPI-KGM complex gels [42].

3. AFFECTING FACTORS

Several variables that influence the rheological properties of gels include: (i) Amount of gelling agent [46] [47] (ii) Material used [10] (iii) Lipid state (liquid or solid) [48] (iv) Morphology of the gel network [49] (v) Gel production process (heat setting, cold setting, souring, and catalyzed by enzymes), pH, and strength ionic [50] [51] [52] (vi) Fats, salt, and sugar's influence on gel formation [53] fats can hinder gel formation by coating proteins and polysaccharides, preventing them from interacting. Salt can enhance gel strength by promoting electrostatic interactions between polysaccharides and proteins. Sugar can disrupt gel formation by drawing water away from proteins and polysaccharides.

Gel strength is also significantly influenced by water concentration. Because thick gel domains are present in natural tissues like connective tissue, a lower water content (about 60%–80%) in comparison to synthetic polymer hydrogels (around 90%) can result in better gel strength [54]. But regular hydrogels tend to be weak because they're mostly water (about 90%) held together by a web of long molecules [55].

Hydrogels can be made stronger by reducing their water content in two broad ways. Making hydrogels at high concentrations and adding more solution is one method. However, the first strategy can be influenced by viscosity. Generally, hydrogel concentrations in the range of 2-7% will reach a sticky state that does not allow further increase in solids content to increase gel strength.

In terms of the textural properties of composite gels, it has been proven that freezing treatment and humidity control can effectively regulate the textural characteristics of composite gels. Freezing strengthens the composite gel by increasing its elasticity, tensile strength, toughness, and resistance to shear forces and indentation. This improvement can be enhanced by properly managing humidity. Based on the mechanical tests carried out, it is recommended that freezing of the gel be carried out for 46 hours at -20°C [56].

4. RHEOLOGICAL TEST

Imagine a gel like a complex recipe. Studying how it flows (rheology) is like figuring out how the ingredients and cooking methods affect the final texture. We can even design gels with specific feels (sensorial attributes) by tweaking the tiny building blocks (polymer microstructure) and the environment. There are new machines (rheometers) that can measure exactly how these gels respond to pressure or stretching, but we won't delve into those here. The key takeaway is understanding how gels flow, because this knowledge is crucial for controlling how they form and ultimately how they feel. Gels are stretchy solids because their building blocks (polymers) are connected in a special way. The strength of these connections (crosslinking) determines how firm the gel is, which scientists can measure with a special value called the equilibrium modulus. As these connections become stronger, the building blocks themselves also get bigger, and this process is what causes the liquid state (sol) to transition into a solid gel structure. The gel point is like the tipping point for a gel. It's the moment when the liquid mixture turns into a gel with a web-like structure throughout. At this point, the gel can't flow anymore (infinite viscosity) but it's also not quite solid yet (zero equilibrium modulus) [10]. Flory's theory and percolation theory suggest that for a gel to form properly, the connecting pieces (crosslinkers) should have at least three attachment points. This ensures that the gel structure is like a web, with branches connecting throughout the material. [10].

Figure 4. Illustrates the measurement of zero shear viscosity [59].

Dynamic oscillatory shear testing is a commonly used method in rheological research to evaluate the characteristics of complex fluids. There's a special test called SAOS (Small Amplitude Oscillatory Shear) that scientists use to understand how soft materials like gels behave when they're gently squeezed and stretched. This test is based on well-established scientific principles [57]. An investigation into the linear viscoelastic response of gel networks, the parameters usually studied include elasticity (G'), stickiness (G"), and wiggle factor (δ).

The G' value illustrates the elastic response of the material being tested and reflects its capacity to store energy, while G" indicates the viscoelastic properties of the material being tested and shows how much energy is dissipated [58]. The wiggle factor, which is the ratio between G"/G0 and represents the curvature of the tangent curve, describes the delay in response to stress applied to the material. This reflects the significant proportion of elastic and viscous elements in the structure of the gel.

Figure 5. Demonstrates the introduction of the material exhibits yield stress behavior [59].

5. CONCLUSIONS AND RECOMMENDATIONS

A common classification for foods like yogurt, ice cream, cheese, and processed meats is fat-containing gels. An important challenge in the food industry is to replace fat without compromising product quality. With their well-balanced functionality, biopolymers such as proteins and polysaccharides could replace fat in various food applications, including their ability to form gels that can improve structural quality.

Rheological research related to replacing ingredients in foods that have a soft-solid texture is very important to ensure optimal product quality and processing. In this review, we discuss recent research on gels with various fat substituted materials, namely from proteins, polysaccharides, and their mixtures, as well as the contribution of rheological studies in understanding the characteristics of these materials. In this study, we studied how rheological parameters influence gel attributes and how manipulation of these parameters can influence the structure and properties of food products.

By combining food science, digestion research, and non-invasive measuring tools, scientists can develop food gels with applications beyond just food. These gels could even impact fields like medical nutrition and tissue engineering. This review aims to spark creative ideas for a deeper understanding of food gels and their potential in new and exciting areas.

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