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PREPARATION AND CHARACTERIZATION OF EDIBLE FILM FROM SORGHUM STARCH WITH GLYCEROL AND SORBITOL AS PLASTICIZERS

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Abstract

Preparation of edible film from sorghum starch as biodegradable food packaging has been studied in this research. Starch-based film commonly has drawbacks since it is brittle and absorbs moisture easily. Hence, an innovation was developed to produce sorghum starch edible film with low moisture absorption and solubility in water by the addition of various plasticizers (glycerol, sorbitol, and the mixture of glycerol and sorbitol). Formulations of sorghum edible films with these types of plasticizers have never been explored previously. In this work, plasticizer concentrations were varied (20%, 40%, and 60% w/w). The effects of types and concentrations of plasticizer on thickness, moisture absorption/water uptake, solubility in water, tensile strength, elongation, and young modulus were investigated. Among all types of plasticizer, the best treatment on synthesis of sorghum-based edible film was the addition of 20% glycerol, which provided the thickness of 0.1231 mm, tensile strength of 1.4793 MPa, and elasticity of3.6983 N/mm^2. Application of 60% glycerol demonstrated the best water uptake of 60% and elongation of 65%. An addition of 20% sorbitol has the lowest solubility in water (26%). This work contributes on the improvement of the sorghum starch-based edible film properties as alternative potential food packaging by the addition of plasticizers (glycerol, sorbitol, and the mixture of glycerol-sorbitol).

Keywords: Edible film, plasticizer, sorghum, starch, biodegradable

1. Introduction

Plastics are widely used as packaging materials since they are convenient, lightweight, transparent, unbreakable and cheap. However, because of their non-

Nomenclatures

A	Surface area, mm^2
F_{max}	Maximum force, N
W	Wet edible film weight, g
W_o	Dry edible film weight, g

Greek Symbols

τ	Tensile strength, MPa

Abbreviations

CA	Citric Acid
CMC	Carboxymethyl cellulose

biodegradability, plastics wastes cause environmental pollution [1]. Due to this concern, edible films have emerged as alternative to synthetic plastic for food packaging. The benefit of edible films over synthetics plastics is that they can contribute to the environmental pollution reduction due to their biodegradability character [2].

Edible films are defined as a thin layer of material which can be consumed and provided as barrier to moisture, oxygen and solute movement into the food [3]. Edible films act as carriers of antioxidants, antimicrobials, and have been particularly considered in food preservation due to their ability to extend food shelf life. Edible films can be prepared from low-cost natural biopolymers, such as starch, agar, pectin, alginate, gelatine, collagen, and fatty acids. Among those feed-stocks, sorghum starch can be chosen as raw material for edible film preparation since it is abundant, cheap, biodegradable, and easy to form matrix. It is also source of carbohydrate and protein. Sorghum contains 80.42% starch and the availability of sorghum's seeds is 26.63 million tons in Asia-Australia regions [4]. Based on the Indonesian Plan and Strategic 2015-2019, sorghum is among the strategic commodities as feed-stocks of bio-industry in Indonesia [5]. Sorghum starch can be converted to valuable bio-products such as sugar, ethanol, acetone, butanol, lactic acid, hydrogen, methane, and edible film [6]. Yet, it has not been optimally applied for bio-product synthesis. In this work, sorghum starch was developed as the feed-stock for edible film synthesis.

Preparation of edible plastics from sorghum needs a proper formulation to produce high quality material. Starch-based edible film usually has some weaknesses as food packaging since it tends to absorb moisture easily and it is brittle. In order to obtain good physical and mechanical properties of edible films for food packaging and preservation functions, addition of plasticizer at an appropriate concentration into base material is required. Plasticizer must be compatible with polymers of the feed-stocks of edible film [7]. Hydrophilic compounds, such as poly-ols (glycerol and sorbitol) are generally applied as plasticizer in starch-based films since it is able to overcome the brittleness of starch. Besides, glycerol and sorbitol have small hydrophilic content. Thus, it is easy to insert these plasticizers between polymer chains of edible film. To extend edible films shelf life, citric acid is required for preservation. Citric acid is among the common and safe preservatives for food application. Thickener agent is also essential in the film formulation. Carboxymethyl cellulose (CMC) can be selected as thickener since it is food grade, abundant, affordable, able to perform gelation by using heat, and yields excellent edible films [4].

Based on the interesting findings on the mixing of glycerol and sorbitol in improving properties of edible film, the mixture could be a suitable plasticizer for sorghum starch. Hence, in this work, the effect of the application of glycerol, sorbitol and the mixture of sorbitol-glycerol as plasticizer on the film properties was studied. The best type and percentage of plasticizers in the manufacture of sorghum starch-based edible film were examined to perceive the best properties of edible film in term of thickness, solubility in water, water uptake, tensile strength, elongation, and elasticity. Based on the literature Formulations of sorghum edible films with glycerol, sorbitol, and the mixture of glycerol-sorbitol plasticizers have never been investigated previously. Hence, this work will contribute on the improvement of the sorghum starch-based edible film properties as alternative potential food packaging by the addition of those plasticizers.

2. Materials and Method

2.1. Materials

Sorghum starch was provided by Ganesha Farmhouse (East Java, Indonesia). Glycerol, Sorbitol and Carboxymethyl cellulose (CMC) (technical grade) were purchased from Merck (Germany). Citric acid (CA) (food grade) was purchased from chemical store in Semarang (Indonesia), and distilled water was obtained from the laboratory of Universitas Negeri Semarang.

2.2. Method

2.2.1. Preparation of films

Sorghum starch preparation: Sorghum flour as a feed-stock was sieved using sieve shaker to get a smooth texture with particle size of 120 mesh or finer. The smooth sorghum flour was then precipitated in distilled water for 4 hours to get the wet sorghum starch. Subsequently, the wet sorghum starch was dried under the sun for 2-3 days until the sorghum starch was dried. *Edible films preparation:*5 grams of dry starch was mixed with distilled water (100 mL), different plasticizer concentrations (glycerol, sorbitol, and mixed of glycerol and sorbitol at 20%, 40%, 60% v/w) and CA 10% w/w starch at room temperature (30℃) for 5 minutes. This suspension was then introduced to the oil-bath at 90℃and agitated using magnetic stirrer at 500 rpm for 30 minutes to yield starch suspension containing plasticizer and CA. This suspension then thickened using CMC. CMC (20%w/w starch) was solved in 75 mL of water. The suspended starch and CMC solution was then mixed and stirred at 75℃ in 10 minutes (pH=5.5) to obtain a homogeneous mixture. The mixture was then cooled at 40 and mixed for 20 min to release all air bubbles. Afterwards, it was discharged into an acrylic casting tray with the dimension of 21x29.7x1 cm and dried at 80 in the oven to obtain the films. The films were then characterized to determine the properties.

2.2.2. Film thickness measurement

Thickness indicates homogeneity of the film and it is related to the strength and barrier properties [8]. The films thickness was tested using a digital micrometre with an accuracy of ±1 µm (Krisbow KW06-85). To get the average thickness of edible films, seven tests were taken on each sample, and the mean value was used in the calculation.

2.2.3. Water uptake measurement

To determine water uptake, firstly, the initial weight (W_0) of the sample film was measured. Then, the film was soaked in a flask filled with distilled water for 10 seconds. The soaked film was then lifted from the flask and weighed to obtain wet weigh (W). The sample was soaked back in the flask, then it was lifted every 10 seconds and weighed again. This procedure was performed continuously until a constant final weight of the film was attained [4, 9]. The water absorbed by the film is measured with Eq. (1):

$$Water\ (\%) = \frac{W - W_0}{W_0} \qquad (1)$$

2.2.4. Solubility in water measurement

Solubility in water was defined as the percentage of the dry substance of film which is solubilized after 24 h immersion in water. The water solubility test was conducted following Ghanbarzadeh's procedure. The film was stored in a desiccator containing potassium silica gel until it reached a constant weight (initial dry weight). Thereafter, about 500 mg of film was immersed in a glass containing 50 ml of distilled water at 23 for 24 hours by mild agitation. The film was then taken from the water and placed in the desiccator until it attained a constant weight (final dry weight) [10]. The percentage of total soluble substances (water solubility) of the film was determined using Eq. (2):

$$Water\ Solubility = \frac{Initial\ dry\ weight - final\ dry\ weight}{Initial\ dry\ weight}\ x\ 100\% \qquad (2)$$

2.2.5. Tensile strength measurement

Measurement of tensile strength was conducted by using Brookfield Texture Profile Analyser. The value of tensile strength was determined by dividing the maximum force (F) with the surface area (A) of edible films as shown in Eq. (3):

$$\tau = \frac{F_{max}}{A} \qquad (3)$$

2.2.6. Elongation at break measurement

The measurement of elongations is carried out using the similar method as tensile strength test. The value was expressed as a percentage and calculated by Eq. (4):

$$Elongation\ (\%) = \frac{strain\ when\ broken\ (mm)}{initial\ length} x100\% \quad (4)$$

2.2.7. Young modulus (elasticity) measurement

The young modulus, which could be related with elasticity, is equal to the tensile strength divided by the elongation of the edible film [11].

3. Results and Discussion

Edible films from sorghum starch using various types and concentration of plasticizer has been synthesized. Properties each edible film were tested in terms of film thickness, moisture absorption (water uptake), solubility in water, tensile strength, elongation at break, and young modulus as shown in Table 1. The influences of the types and concentration of plasticizer are also discussed.

Table 1. Properties of edible film formulated with different types and concentration of plasticizer.

Properties	Plasticizer								
	Glycerol			Sorbitol			Glycerol + Sorbitol		
	20%	40%	60%	20%	40%	60%	20%	40%	60%
Thickness, mm	0.123	0.13	0.149	0.15	0.154	0.156	0.148	0.164	0.17
Water Uptake, %	110	104	60	110	64	64	108	90	72
Solubility in Water, %	26	44	44	30	38	40	28	40	40
Tensile Strength, MPa	1.478	1.175	1.042	1.008	1.009	0.984	0.998	0.978	0.895
Elongation at Break, %	40	50	65	30	35	45	30	30	45
Young Modulus, N/mm^2	3.696	2.35	1.603	3.359	2.884	2.186	3.326	3.26	1.99

3.1. Effects of the types and concentration of plasticizers on film thickness

Figure 1 shows that the increasing concentration of plasticizer led to the higher edible film thickness. The increasing thickness of the film occurs because the plasticizers will inhabit the voids the film network and interact with edible film to form polymer. This condition increases the distance between polymers, hence enhancing the film thickness [12]. It was also reported that the plasticizer added to edible film can bind with starch, creating starch-plasticizer polymer. Thus, the starch-starch bond is substituted by the starch-glycerol-starch bond, leading to the increasing of the film thickness [3]. It has also been reported that the film contains higher concentrations of glycerol has a higher dry substance content, which yielded a thicker film. The thicker film means that it is more rigid to be formed, but it provides better mechanical protection to the packaged material.

The thickness of sorghum starch-based edible film is more or less comparable to that of the hanjeli starch-based edible film. Edible film made of hanjeli starch provided film thickness ranging from 0.15 to 1.16 mm [13]. The difference in edible film thickness can be caused by several factors, such as the difference between the edible film being poured into the mold and the drying temperature. In fact, edible films can be adjusted in thickness by varying the amount of solution poured to the mold and the area of mold used. The more volume of edible film solution is poured, the thicker edible film will be obtained. It is because the total solid in the edible film solution gets greater [3].

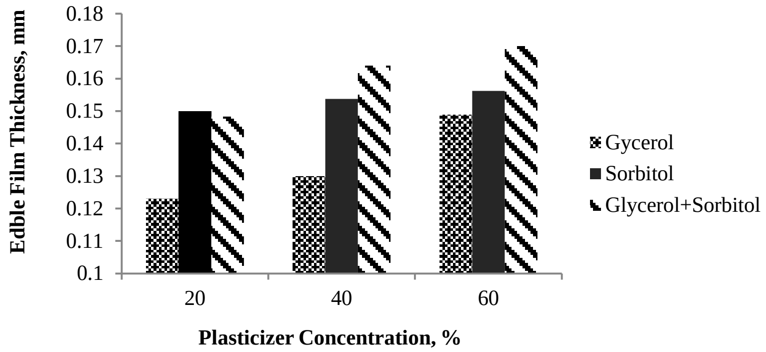

Fig. 1. Effect of type and concentration of plasticizer on film thickness.

3.2. Effects of the types and concentration of plasticizers on water uptake

The disadvantage of polysaccharide films is the weak water vapour barrier and the tendency to absorb moisture from the surroundings due its hydrophilicity [14]. The properties of plastic film resistance to water are commonly determined by swelling test (water uptake), which indicates the percentage of film inflation by water. Hence, the best water uptake characteristic means that edible film absorbs the smallest amount of water or moisture. In this work, as depicted in Fig. 2, the higher concentration of plasticizer leads to lower water uptake. This phenomenon was revealed for all types of plasticizers. The best water uptake was 60%, provided by edible film with addition of 60% glycerol. Meanwhile, the water uptake of control was 146%.

This research showed that the water uptake value of edible film was smaller when the higher concentration of plasticizer was employed. It is not surprising since the film with a lower plasticizer concentration has more empty space than the film added with a higher concentration of plasticizer. The higher volume of empty room will enable water to easily enter the empty space, leading to the greater water uptake. Figure 2 also reveals that the addition of 60% glycerol plasticizer resulted in the best water uptake compared to the addition of other plasticizers. Glycerol has a smaller molecular size than sorbitol. Therefore, it easily and efficiently enters into the film network and fills the space. This condition will decrease the opportunity of water to be adsorbed and reduce the transfer rate of water in the film. Thus, glycerol plasticizer resists the rate of water and moisture uptake more efficiently [15].

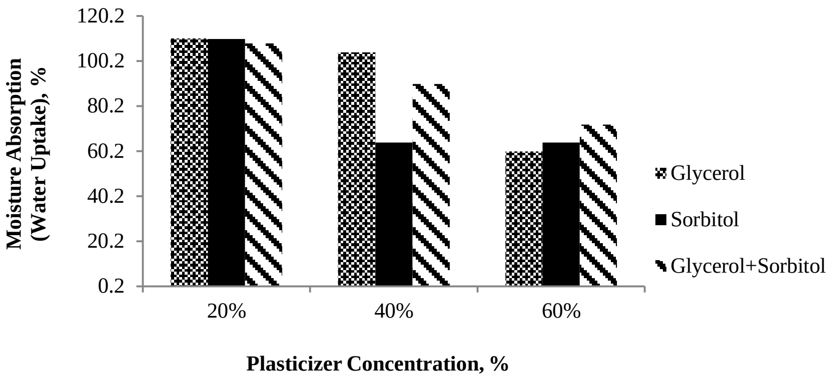

Fig. 2. The effect of type and concentration plasticizer on water uptake.

3.3. Solubility in water

Solubility of edible films in water constitutes a dry weight percentage of edible films that has been dissolved in water for 24 hours. Edible film with the lowest solubility in water indicates the best quality of edible film. Edible film with 20% concentration of sorbitol as a plasticizer has the lowest solubility value (26%). Solubility of edible film in water is influenced by composition of the materials that are used. The increased concentration of plasticizer added will increase solubility of edible film in water. It is due to the fact that the addition of the higher amount of plasticizer which has hydrophilic character will increase the solubility of edible film in water [3].

3.4. Tensile strength

Figure 3 demonstrates that for all types of plasticizers, the value of tensile strength decreased with the increase of plasticizer concentrations. This finding is in accordance with the result of Arkam’s work which applied glycerol plasticizer on agar-based edible film [12]. The highest tensile strength was 1.4793 MPa, obtained by the addition of 20% glycerol plasticizer. Glycerol exhibited a better result than the other types of plasticizers examined in this work. Glycerol can reduce the intermolecular force on the polymer chain, thus it enhances the edible film flexibility and extends the distance between molecules. It additionally will decrease the hydrogen bond on the polymer.

3.5. Elongation

The elongation value resulted by the addition of various types of plasticizer at different concentration is demonstrated in Table 1. It was revealed that the employment of 60% glycerol provided the highest elongation value (65%).

Furthermore, the increased on the plasticizers concentration results in the increased elongation value of edible films. The plasticizer plays important role to decrease the inter-molecular force. Thus, it will increase the mobility between polymers, and consequently edible film becomes more elastic and flexible [3].

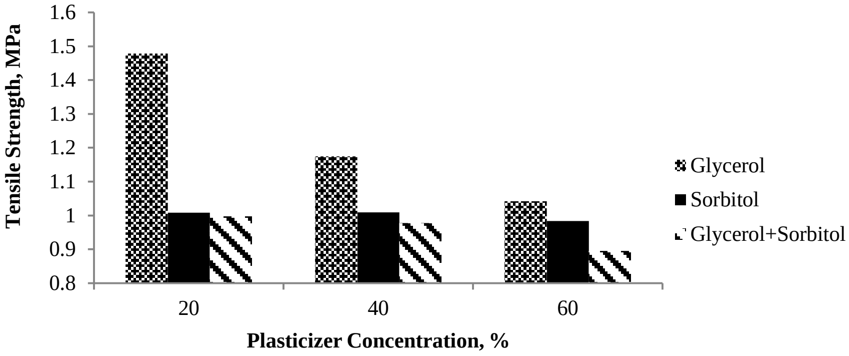

Fig. 3. The effect of type and concentration plasticizer on tensile strength.

1.1. Young Modulus (Elasticity)

Young modulus (elasticity) is the opposite of percentage of elongation in which it decreases as the increases amount of plasticizer applied in edible film. Based on the data presented in Table 1, it is shown that the adding of 20% glycerol results in the highest value of elasticity (3.6983 N/mm^2).

2. Conclusion

For all types of the variation of plasticizer, addition of 20% glycerol demonstrated the best value of thickness (0,1231 mm), tensile strength (1,4793 MPa), and elasticity (3,6983 N/mm2). An addition of 60% glycerol has the best water uptake (60% water content) and elongation (65%). In the other hand, an addition of 20% sorbitol has the lowest solubility in water (26%).

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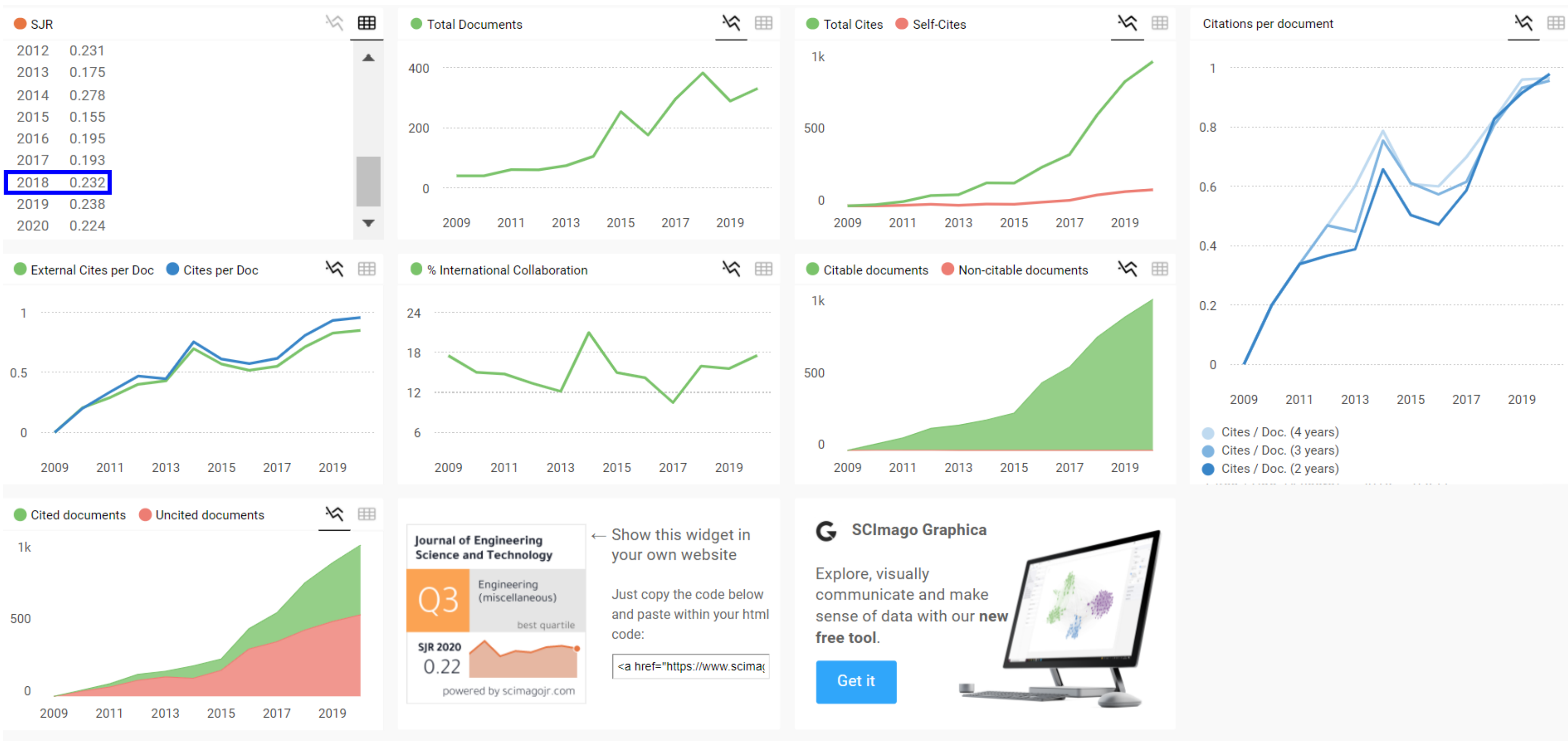

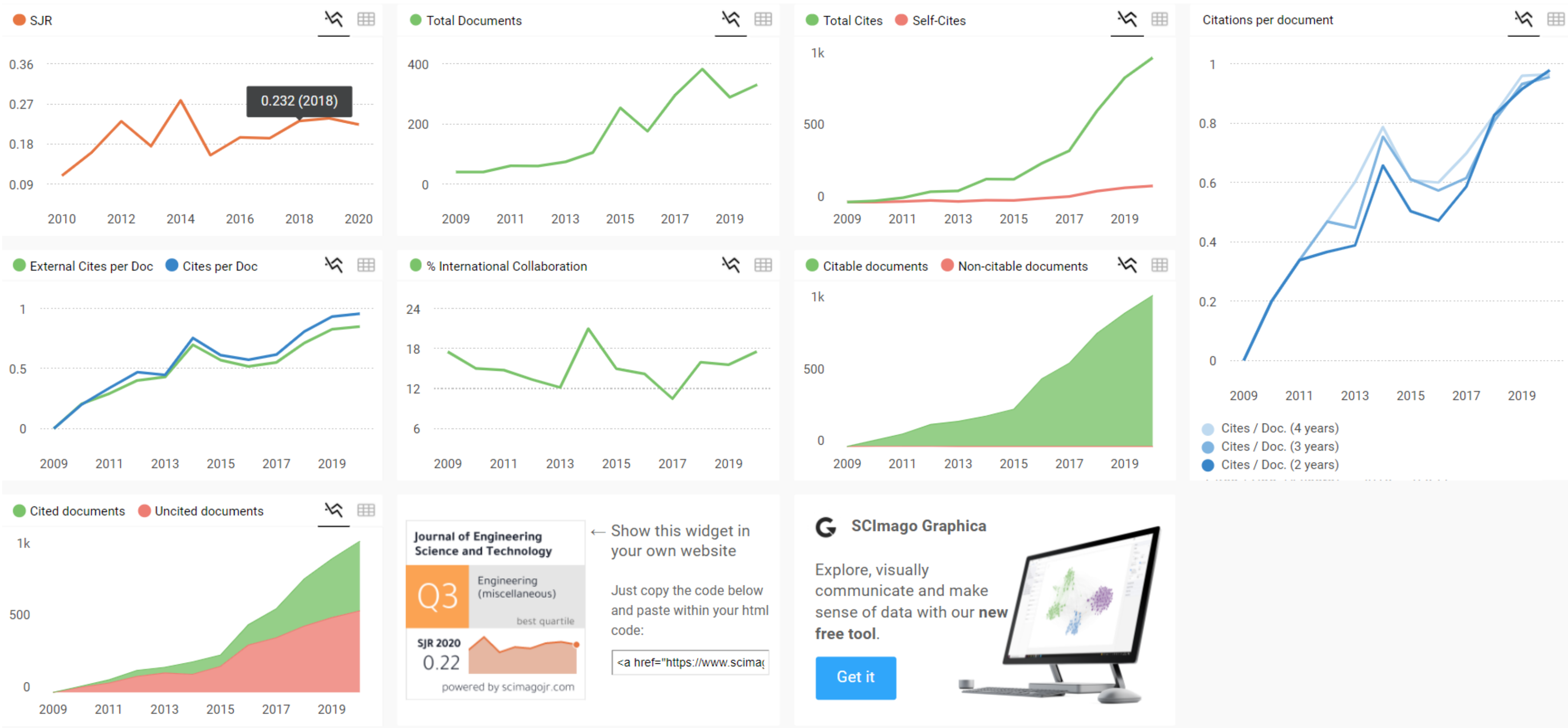
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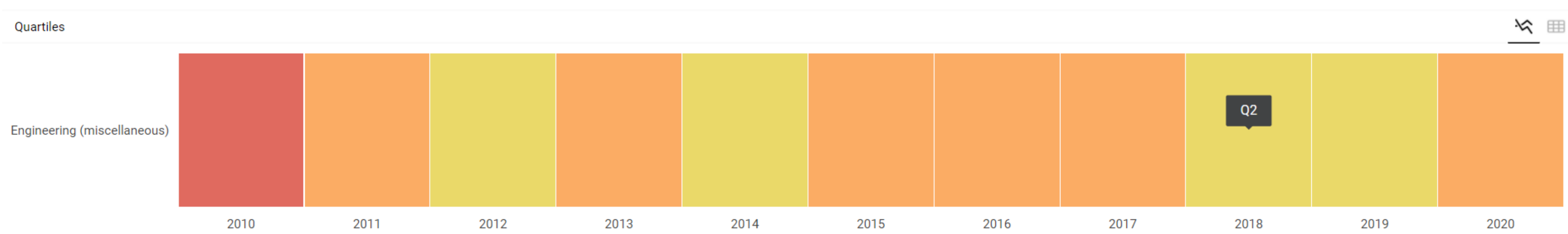

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