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Review Article

A Review on Treatment Efficiency of Pharmaceutical Effluents Using Natural Coagulants

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Abstract: Pharmaceutical effluents are wastes generated by pharmaceutical industries during the process of drug production. Pharmaceutical industry effluent is hazardous in nature due to its components such as toxic metals and active pharmaceutical ingredients and therefore should be treated before it is discharged into any water body or on to soil. But studies have shown that in most developing countries like Nigeria, industries dispose their effluents without treatment. The pharmaceutical effluents can be treated using inorganic or natural coagulants. This paper reviewed some studies on the treatment of pharmaceutical effluents using natural coagulants which are low-cost, under-utilized, economic friendly and readily available. The coagulants reviewed include *Moringa oleifera, Citrullus lanatus* (Seed of watermelon), *Treculia africana* (African bread fruit), Phoenix dactylifera (Date), *Zea mays* (Corn or maize), Banana peels, *Sesamum indicum* (Beniseed). The studies were critically reviewed with respect to the type of natural coagulant used, method of application to the effluents and result obtained. The results from the studies reviewed revealed that the treatment efficiency was maximally achieved in each case when the natural coagulants were used in combined form (either with another natural coagulant or inorganic coagulant) in appropriate ratios. Treatment efficieny of pharmaceutical effluents is obtained by using combined natural coagulant.

Keywords: Pharmaceutical Effluent, Natural Coagulant, *Zea mays*, *Sesamum indicum*

1. Introduction

Effluent is a liquid waste that is sent out from factories or places where sewage is dealt with usually flowing into rivers, lakes or the sea. The effluent is expected to be treated or disinfected in the plant prior to discharge using any type of process depending on the components of the effluent. It is a complex mixture containing several active ingredients from the entire production. Effluent can be generated by residential, institutional, commercial and industrial establishments. It includes household waste liquid from toilets, baths, showers, kitchens sink and so forth that is disposed of via sewers [1]. Pharmaceutical industry is considered as one of the highly polluting industry generating a huge amount of wastewater [2]. Pharmaceutical compounds are typically produced in batch processes leading to the presence of a wide variety of products in wastewaters which are generated in different operations. The presence of pharmaceutical compounds in drinking water comes from two different sources: production processes of the pharmaceutical industry and common use of pharmaceutical compounds resulting in their presence in urban and farm wastewaters. The wastewaters generated in different processes in the manufacture of pharmaceuticals and drugs contain a wide variety of compounds [3]. Pharmaceutical effluents are waste generated by pharmaceutical industries during the process of drugs manufacturing. The pharmaceutical industry manufactures biological products, medicinal chemicals, botanical products and the pharmaceutical products as well as other commodities. The industry is characterized by a diversity of products processes, plant sizes as well as waste water quantity and quality.

Generally, pharmaceutical industries do not generate uniform waste streams, due to the variety of medicines produced during any given processing period. In recent times, a wide variety of pharmaceuticals have been found in fresh and marine waters and some of these compounds are potentially capable of causing harm to both aquatic and terrestrial life. Pharmaceutical industry wastewaters may contain organic solvents, catalysts, additants, reactants, intermediates, raw materials and active pharmaceutical ingredients (APIs) which makes them difficult to treat [4]. The pharmaceutical effluent contain high amount of biological oxygen demand (BOD), chemical oxygen demand (COD) and nitrate which pose a great hazard to both human and aquatic environment when released untreated in to the environment [5]. The presence of toxic or recalcitrant substances in such wastewater results in lower chemical oxygen demand (COD) removal efficiencies [6]. It has been estimated that up to half of the pharmaceutical wastewater produced worldwide is released without any treatment [7].

Treatment of any wastewater is compulsory before it's discharge into the ecosystem. There are three general types of wastewater treatment, chemical method, biological method and physical method. Chemical method involve reactions to produce insoluble solids, reaction to produce insoluble gas and oxidation-reduction process and biological methods involve anaerobic and aerobic reactions while physical method includes gravity separation, reverse osmosis and ion exchange. The implementations of these methods depend on the type of wastewater treated and the efficiency of the treatments in economical way [8].

In most developing countries like Nigeria, most industries dispose their effluents without treatment. These industrial effluents have a hazardous effect on water quality, habitat quality and complex effects on flowing water [9]. In Nigeria, main contributors to the surface and ground water pollution are the by-products of various industries such as pharmaceutical, textile, metal, dying chemicals, fertilizers, pesticides, cement, petrochemical, energy and power, leather sugar processing, mining and others. The discharge of industrial effluents, municipal sewage, farm and urban wastes carried by drains and canals to rivers worsen and broadens water pollution [10]. Recently, scientists have raised concerns about pharmaceutical residues detected in rivers and coastal waters and their potential to cause adverse effects in humans and aquatic species [11]. The presence of pharmaceutical chemicals in the environment is a matter of concern today due to their lipophilic and non-biodegradability nature, as well as their biological activities.

In wastewater treatment operations, the processes of coagulation and flocculation are employed to separate suspended solids from water. Coagulation is the destabilization of colloids by neutralizing the forces that keep them apart. Cationic coagulants provide positive electric charges to reduce the negative charge (zeta potential) of the colloids. As a result, the particles collide to form larger particles (flocs). Rapid mixing is required to disperse the coagulant throughout the liquid. Coagulation process is necessary in wastewater treatment as it can help to sediment the flocs formed during chemical treatment of the wastewater.

However, there are different ways by which the effluents can be treated before discharge, these involve the use of inorganic/synthetic coagulants or natural coagulants. Natural coagulants are associated with biological processes. All biological-treatment processes take advantage of the ability of microorganisms to use diverse wastewater constituents to provide the energy for microbial metabolism and the building blocks for cell synthesis. This metabolic activity can remove contaminants that are as varied as raw materials and by-products. The use of biological materials, including living and non-living micro-organisms, to remove and recover toxic or precious metals from industrial waste waters has gained popularity over the years due to increased performance, availability and low cost of raw materials, [12-14] microorganisms including bacteria [15]. Inorganic/synthetic coagulant is associated with conventional processes. Conventional wastewater treatment processes are not specifically designed to degrade traces of dangerous organic contaminants, and these are consumed by aquatic organisms, which is a hazard to the whole food chain. Conventional techniques such as chemical precipitation, carbon adsorption, ion exchange, evaporations and membrane processes are found to be effective in treatment of waste and sewage water.

Natural coagulants (biopolymers) are natural low-cost products, characterized by their environmentally friendly behaviour, and presumed to be safe for human health [16]. They are also readily available especially those of the plant origin than animal-based coagulants, thus suggesting that they could be potential replacements to chemical coagulants and have since gained continuing impact over the years [17]. Natural coagulants include extracts of microorganisms, animal or plant origin, examples include Narmali seeds, *Moringa* seeds, Okra, Cassava, banana, *Phoenix dactyifera*, water melon seed, *Zea mays*, broad beans, flava beans, etc. Inorganic/synthetic coagulants, on the other hand, include aluminium sulphate (alum), aluminium chloride and sodium aluminate, ferric sulphate, ferrous sulphate, ferric chloride and ferric chloride sulphate. Others include hydrated lime and magnesium carbonate [18]. These chemical coagulants also have some drawbacks, which include pH alteration, large volume of sludge resulting in huge disposal cost, not being effective.

Some studies have revealed the efficacy of effluent treatment with natural coagulant over treatment with inorganic/synthetic coagulant. This study therefore reviews the efficiency of using natural coagulant in treating pharmaceutical effluent.

2. Treatment of Pharmaceutical Effluents Using Natural Coagulants

Biological treatment is mainstream for pharmaceutical wastewater at home and abroad, which is the most economical way to remove organic pollutants since organic matter are the main pollutants in pharmaceutical wastewater. This study reviewed seven natural materials that are locally sourced and constantly available to act as coagulants in pharmaceutical effluent treatment. Studies have been carried on the treatment of pharmaceutical effluents by natural coagulants. Some of the reviewed studies involving the use of natural coagulants are given thus:

2.1. Moringa oleifera

Moringa oleifera is the most widely cultivated species of a monogeneric family, the Moringaceae, native to the sub-Himalayan tracts of India, Pakistan, Bangladesh and Afghanistan [19]. The flocculant active cluster from *Moringa oleifera* extract consitute aliphatic primary amides and primary aliphatic alcohol. The amide cluster shows that polyelectrolyte flocculant is positively charged, while polyelectrolyte hydroxyl cluster is charged negatively [20]. The mechanism that occurs during the *Moringa oleifera* flocculant process is adsorption and voltage neutralization or adsorption and unstable particles bond. The seed has been found to have antimicrobial activity against bacteria, moulds and fungi. Many studies have proved *Moringa oleifera* to be very good natural coagulant both in combined and uncombined form. *Moringa oleifera* seeds are used as a primary coagulant in drinking water clarification and wastewater treatment due to the presence of a water-soluble cationic coagulant protein which is able to reduce turbidity of the treated water [21, 22]. The leaves are used for Cadmium (II) removal from waste water [23]. *Moringa* seeds are much more effective in water purification in terms of adsorption of metals. The percentage removal by *Moringa* seeds were 90% for copper, 80% for lead, 60% for cadmium and 50% for zinc and chromium [24].

Moringa oleifera contains coagulant proteins that play a major role in surface water treatment. On dosing extracts of *Moringa oleifera* that are soluble in water, turbidity was reduced to 5.9 NTU from 100 NTU in highly turbid water. Also 89-96% of coliform was reduced on using this natural coagulant. Laboratory investigations confirm the seed to be highly effective in the removal of suspended solids, Eri et al. (2018) ascertained that the percentage removal of total suspended solids herbal pharmaceutical effluent using moringa oleifera was 93.42-99.54% [20].

A research on moringa seed extract (*Moringa oleifera L.*) in coagulation activity proved that the use of moringa extract is more effective rather than alum [25]. The *Moringa* seed extract can reduce the amount of turbidity by 81%, while alum can only reduce 58% [26]. Studies revealed that among the large number of plant materials that have been used over the years, the seeds from *Moringa oleifera* have been proved to be one of the most effective primary coagulants for water treatment especially in rural communities [22, 27, 28]. The studies on *Moringa oleifera's* competence in water and wastewater treatment process had been undertaken by numerous scholars [29-33].

2.2. Citrullus lanatus (Seed of Water Melon)

Citrullus lanatus popularly known as watermelon is a popular seasonal plant found in sub-Sahara [34]. It is a member of the cucurbit family (Cucurbitaceae) and have four distinct parts which include the rind/peel, the seed, the fleshy white and the fleshy red/pink/yellow parts. The seeds can be brown, white green, or yellow and a few varieties are actually seedless [35]. The watermelon seed kernel composed of 35.7% crude protein, 50.1% crude oil, 4.83% crude fiber, 3.60% total ash, and 5.81% nitrogen [36]. The seeds of watermelon are considered as agrowaste and are spitted out in spite of its high nutritional value. Watermelon seeds are highly nutritional; they are rich sources of protein, vitamin B, minerals and fat among others as well as phytochemicals. Roasted watermelon seeds are served as a snack to garnish salads. Watermelon seed oil is extracted from these seeds which is beneficial for hair and skin. The seeds of watermelon are effective water purifiers because of their adsorbent properties like most of the indigenous seeds. [37, 38]. They treat water on two levels, acting as both a coagulant, an antimicrobial agent and can be used to enhance the filtration process during water treatment [39, 34, 40].

The Yeast (*Saccharomyces cerevisiae* and *Torulaspora delbrueckii*) extract from the spoilt water melon seed are capable of biological treatment of pharmaceutical effluent [5]. The yeasts showed considerable colour removal probably because of the adsorption of the anionic dyes in the effluent due to the nitrogen containing (or protein) group that comprises the yeast biomass and other cellular components as suggested by Kumar et al. (2007) [41]. Abioye et al. (2015) also confirmed significant reduction in BOD, COD, turbidity, nitrate and sulphate using the isolated yeast from spoilt water melon seed as a coagulant in treating pharmaceutical effluent thereby removing the offensive odour and the effluent clear [5]. The watermelon seeds when employed as a coagulant significantly decrease BOD, COD, TSS, TDS and turbidity of the synthetic wastewater and also the tannery effluent [42, 34].

Sathish et al. (2017) also reported that efficiency of seed of watermelon in turbidity removal was 98% from synthetic wastewater and 86.7% for the tannery effluent, BOD, COD and TSS of the wastewater were reduced to a greater extent with removal efficiency of 54.96%, 50.19% and 68.8% respectively for BOD, COD, TSS [42].

The dosage, mixing speed and stirring time on coagulation have effect when using watermelon seed cake as coagulant [43, 36]. A research showed that at a dosage of 0.1g/L, stirring time of 10 minutes and mixing speed of 100rpm, the highest turbidity removal was observed [43]. The residual turbidity at optimum dosage of watermelon seed cake improved by 25% (14.4 NTU) [43]. While the research by Bello et al. (2019) showed highest turbidity reduction from 62.6NTU to 3.68 NTU at a dosage of 0.1g/L with stirring time of 8 minutes and mixing speed of 100rpm, which is below the World Health Organizations (WHO) recommended value of 5NTU [36].

Water melon seed cake in combination with chemical coagulant such as alum have been proved to give higher colour and turbidity removal, going as high as 100% clarification of colour. However the recommended ratio for the combined coagulant dose was 80% watermelon seed powder and 20% alum as best water treatment was obtained. This therefore establishes that watermelon seed powder as a natural coagulant can be more effective when used with 20% alum as a coagulant mixture. [40]

2.3. Treculia afaricana (African Bread fruit)

Treculia afaricana, (African bread fruit) belongs to Moraceae family. It is an evergreen forest fruit tree in tropical Africa countries like Ghana, Sierra Leone, Nigeria, etc. African breadfruit is an underutilized tree crop that produces large round fruits which are covered with rough pointed outgrowths. The seeds are embedded in the spongy pulp of the fruits. Three varieties of *Treculia* have been recognized based on the size of the seed: var. *africana* (large), var. inverse (medium) and var. mollis (small) [44]. African breadfruit is an important food item in various regions of tropical West Africa and is usually consumed as pottage or roasted snack sold with palm kernel or coconut. The breadfruit flour has a high potential usage for pastries production. The *africana* seeds are an important source of vitamins, minerals, proteins, carbohydrates and fats [45]. Ellis et al. (2007) reported that the crude protein content, fat, ash, crude fibre, carbohydrates, iron, calcium, potassium and phosphorus content of African breadfruit seed were of 13.35%, 10.12%, 1.96%, 2.83%, 62.01%, 8.70 mg·100 mg-1, 93.90 mg·100 mg-1, 464.60 mg·100 mg-1 and 1300.00 mg·100 mg-1, respectively [46]. The seed has been a natural resource, contributing significantly to the income and dietary intake of the people. The seeds are used for soup thickening and making bread fruit cakes, snacks and cookies [47, 48].

Treculia Africana seed coat (TA) which is one the agrowastes can serve as alternative coagulant for reduction of colour from Pharmaceutical effluent [49]. The study revealed that the optimum colour reduction efficiency was 64.36% at coagulant dosage of 252.32 mg/L, settling time of 25.31 minutes and pH of 2.89 [49].

2.4. Phoenix dactylifera (Date)

Phoenix dactylifera is called date or date palm in English. Date palm tree belongs to Arecaceae family (Angiosperms, monocotyledon) [50]. It is a one-seeded fruit, usually oblong. It has been the staple food and chief source of wealth in the irrigable desert from ancient times [51]. *Phoenix dactylifera* has been found to originate from North Africa and grows in country such as Arabia, Persian Gulf, Canary Islands, in the northern Mediterranean and in the south of the United States. It has palm with a very slender trunk, up to 30 m tall, conspicuously covered with the remains of sheaths from fallen leaves [52]. *Phoenix* seeds consist mainly of cellulose, hemicelluloses and lignin [53]. Both lignin and cellulose are organic polymers present in plant species, but they differ in chemical structure. Cellulose comprise of organic carbohydrate monomers, while lignin consists of organic non-carbohydrate monomers that are difficult to degrade. When date seeds are pyrolyzed, their cellulose and hemicellulose components decompose, giving rise to pores that directly increase the surface area, improving their application as adsorbents. Hence, they can be used in wastewater treatment, with surface modification under simple conditions [54]. Surface modification of the carbon materials was achieved with thermal energy on its own or in combination with chemical energy. Carbon adsorbent prepared from *Phoenix* seeds and their surface combined with potassium hydroxide can be used as an adsorbent for the removal of synthetic dye (methyl orange (MO)) from aqueous solutions [55].

Iloamaeke et al. (2019) disclosed that the surface of this coagulant look sticky and rough with tiny pores in it [56]. The rough sticky surface and pores are the point of attachment of the colloidal particles of the pharmaceutical effluent. This property makes *Phoenix dactylifera* to be qualified as suitable coagulant. At optimum condition, *Phoenix dactylifera* is found to be an effective natural coagulant in the treatment of pharmaceutical effluent. Coagulant dosage of 100 mg/L, pH 2 and time of 50 mims were found to give the highest colour removal efficiency of 99.86% [56].

Phoenix dactylifera is efficient coagulant both in combined and uncombined form. The combination of *Mangifera indica* (mango) and *Phoenix dactylifera* (dates) seeds powder as coagulants in wastewater treatment removed 96% turbidity, 96% BOD, 87% COD and 98% total coliform of the wastewater [57]. The research showed the efficacy of combining *Mangifera indica* and *Phoenix dactylifera* seeds powders as coagulants in wastewater treatment at the effective combined ratio (of 20mg/L of *Mangifera indica* with 5mg/L *Phoenix dactylifera*) for turbidity and total coliforms removal. The statistical analyses showed that over 85% of the turbidity and total coliforms removed can be attributed to the change in dosage administered to the wastewater. Thus, the use of *Mangifera indica* and *Phoenix dactylifera* seeds powders have great potentials as coagulants and to some extent in wastewater treatment [57].

2.5. Zea mays (Maize or Corn)

The use of corn as a food ingredient also increase corn production, the wastes resulting from the production of corn such as the leave, stem, tassel, cob and the husk have not been optimally utilized. Therefore to increase the economic value and utilization of this plant, these corn waste products can be utilized as a basic material of activated carbon as bio adsorbent through activation process.

Investigations have confirmed the seeds of *Zea mays* (corn) to be highly effective in the removal of suspended solids from water containing medium to high initial turbidities [58]. The turbidity reduction (81.0%) was achieved with 1g of *Zea mays* for one hour treatment [58]. Treatment with *Zea mays* showed the significant decreased up to 92% in the reduction of microbial counts [59].

Zea mays tassel which is a zero-value agricultural waste was used to produce a low-cost activated carbon using phosphoric acid as the activating agent, the prepared *Zea mays* tassel activated carbon (an adsorbent) was applied for adsorption of an emerging contaminant, metformin hydrochloride (MH) from pharmaceutical effluent [60]. The pH, contact time, adsorbent dosage, and initial MH concentration upon which the adsorption depend were investigated using a response surface methodology and the following were obtained: pH 9.5, contact time 67.50 min, dosage 0.5750 g, and MH concentration 152.50 mg/L. Consequently, the study demonstrated that *Zea mays* tassel is a potential precursor for preparation of adsorbents for the removal of the MH from pharmaceutical effluent [60].

Maize husk, an agricultural waste has potential of iron removal by adsorption from groundwater or other polluted waters [61]. The authors indicated that the adsorption was dependent on PH, adsorbent dose, initial iron concentration and contact time. The activated carbon from corn cobs can reduce the heavy metal content of Fe, Cu and Pb which are heavy metals that are widely distributed in industrial waste [62]. The use of activated carbon in the process of industrial waste purification could be a good option because of its effect in reducing heavy metal content. *Zea mays* seed also have ability to remove heavy metals from polluted water.

2.6. Banana Peels

Mainly banana peels contain nitrogen, carboxylic acid, carbohydrate, fiber, potassium, manganese, iron, sodium which attracts toxic metals present in the water. Metals found in impure water are lead, copper, mercury, and iron, fruit peels can be used to remove these metals. Banana peel contains lipids (1.7%), proteins (0.9%), crude fiber (31%) and carbohydrates (59%). The various minerals present are potassium (78.10 mg/g), manganese (76.20 mg/g), sodium (24.30 mg/g), calcium (19.20 mg/g) and iron (0.61 mg/g) [63]. Banana peels and Fish Scales, which are discarded waste materials and are in abundance in the local market, can be used for the removal of heavy metal from pharmaceutical waste water. Efficiency of removal of heavy metal concentration is more with banana peel and then with fish scale. Mixture of both the adsorbents gives more efficiency [63].

The surface activity of banana peels is mainly due to the presence of carboxyl, hydroxyl and amide groups at its surface due to which it is able to chelate with various metals and help in their removal. High surface area of banana peels adds to the property and makes it an excellent and economic adsorbent than fish scale, for water purification process [63]. Banana peel seems to be good adsorbent and can be used as valuable material for cleaning of water. Bioadsorbent prepared from banana peels has been reported for the removal of chromium, cadmium and copper ions from aqueous solution [53]. Also the fish scales are used as the bioadsorbent for removal of copper, chromium, zinc and iron from waste water [64].

Aftab et al. (2005) treated diary wastewater using low cost adsorbents, the peels of orange and banana by carbonization and dehydration (biosorption) methods [65]. The removal of organic substance from dairy waste water using de-hydration and carbonization methods for orange and banana peels was studied by investigating the effect of pH, time, adsorbent dosage and particle size. The carbonization method was found to be more efficient than the de-hydration method for both orange and banana peels with highest percentage removal of 50.1% and 44.1% respectively. The carbonization method is considered to be better since carbon is a strong oxidant and has a unique pores structure which adsorbs the organic substances to its surface easily. Some studies have revealed the removal of heavy metals from industrial effluents using banana and orange peels [65, 66]. The authors found out that the maximum efficiency of removal of heavy metal using banana and orange peels is 60% and 70% respectively [65, 66].

Studies show that the removal of COD, SS and turbidity by banana stem juice was efficient at a pH 7, where the percentages were at 83.9, 84.1, and 90.9%, respectively. Turbidity removal percentages showed marginal difference in which more than 98% removal was achieved. At a dosage of 90mL, the highest COD removal percentage was 80.1% and the suspended solid removal percentage was 88.6%. This shows that banana stem has the potency to act as a natural coagulant and it can be used in the pretreatment stage as a fine powdered coagulant. Adsorption of heavy metals from water using banana and orange peels was studied [67]. The effect of pH was experimented for both orange and banana peels on different metals. It was concluded that the maximum adsorption occurs at pH 6.8 for orange and banana peels.

2.7. Sesamum indicum, (Beniseed) Extract

Sesamum indicum is a flowering plant of *Sesamum genus*, also called beniseed belongs to the family Pedaliacea [68]. The tiny spherical seeds are edible, having milky flowering flavour. It is an important oil seed believed to have originated from tropical Africa with the greatest diversity [69]. Beniseed is a staple food among many ethnic groups in Nigeria and it is cultivated in most areas of the middle belt and some northern states of Nigeria [70]. The seed are consumed fresh, dried, fried or when blended with sugar. It is also used as a paste in some local soups. Sesame seeds have also been found to possess numerous medicinal properties. *Sesamum indicum* seeds contain relatively high protein content of 30.6% and this is an indication of the good performance of the seeds as a precursor for the treatment of effluents [17, 71-73]. This is essential for neutralization (the adsorption of an oppositely charged coagulant on the colloidal surface) of the colloidal particles; these, in turn, induces the process of coagulation via bridging, which stimulates the flocs development [17]. Beniseed was used as a coagulant for the removal of turbidity from surface water [74]. They obtained maximum turbidity removal of 97.6% at the optimum coagulant dose of 12 g/L at the pH of 6.8 and coagulation time of 10 min. Another study showed that total dissolved solids (TDS) can be removed effectively from aquaculture wastewater with the use of *Sesamum indicum* seeds extract as coagulants [75]. The process was found to be dependent on the dosage of coagulant, settling time, temperature and pH of the effluent. Optimum TDS removal of 82% was obtained at pH of 2, dosage of 0.4

g/L, solution temperature of 303 K and settling time of 60min [75].

3. Conclusion

Natural coagulants are natural low-cost products, characterized by their environmentally friendly behaviour, presumed to be safe for human health and readily available. The review of many studies on the treatment of wastewater precisely pharmaceutical wastewater has shown the efficiency of using natural coagulants both in combined and uncombined form. It can concluded from the results of these reviewed studies that the use of the natural coagulants from plants and animal origins are more effective in treating effluents when combined with either another natural coagulant or inorganic coagulant in appropriate ratios. Pharmaceutical effluents can therefore be treated efficiently with natural coagulants combined in correct proportions.

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