Point-of-Use technology for clean water treatment in disaster areas: effectiveness of LDH plate in eliminating pathogenic bacteria

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Abstract. This study tackles the clean water crisis in West Sumatra, often exacerbated by natural disasters such as flash floods and landslides, by proposing an innovative point-of-use water treatment method. The method employs layered double hydroxide (LDH) synthesized on aluminum plates, created by soaking these plates in a solution of seawater and magnesium hydroxide. The X-ray diffraction analyses proved the formation of LDH. Experiments demonstrated that this method is suitable for water contaminated with E. coli. For the LDH plates, this removal rate of E. coli reached 100% with continuous stirring and 91% with occasional stirring within 24 hours. Stirring did enhance contact between LDH and water, thus improving the rate of removal of the bacterium. The process is low-cost and simple, with all materials easily available, so this offers a very reproducible solution to deliver clean water in disaster zones. Further studies shall be conducted for its performance against different bacterial strains and in more complex water matrices. This work substantially expands current knowledge of POU/HWT technologies and delivers a real-life solution for providing clean water in disaster situations.

1 Introduction

West Sumatra is one of the 38 provinces comprising Indonesia, located on the western part of Sumatra Island, and is predominantly recognized for its natural beauty. The geography of the region is varied, with a coastline along the Indian Ocean and the towering *Bukit Barisan* Mountains, and so the area is predisposed to a number of natural disasters. Flash floods, landslides, and earthquakes are major threats to the citizens of this region [1, 2]. Due to high tectonic activity, the *Bukit Barisan* Mountains commonly bring about earthquakes. Heavy rainfalls in many areas cause recurring flash floods and landslides [3, 4]. Such disasters not only bring physical damage and loss of life but even result in shortages of clean water supply, a basic human need.

Lack of water can lead to many serious health issues. As natural hazards, flash floods and landslides can damage the water supply systems, including clean water pipes and springs. In addition, these natural hazards can contaminate the water sources through mud, debris, and hazardous chemicals [5, 6]. For instance, if there is a flash flood, excess water from the river may overflow with a great number of materials originating from upstream, such as soil, debris, and even agricultural chemicals. This, in turn, contaminates the river water, which the community uses for clean water, and makes it unhygienic for consumption [6]. Landslides can also cause this. If a landslide happens, the material of the landslide may either cover the water sources or destroy clean water distribution infrastructures. Apart from that, landslide activities can introduce high loads of organic and inorganic materials, which, together with the pathogenic bacteria, require more intensive purification efforts to make the water sources safe for consumption.

In certain circumstances, various water treatment technologies and methods used at the point of use (POU) or in households (HWT) are the primary choices for providing clean water to the community. POU/HWT methods can serve as a temporary solution before permanent infrastructure becomes available [7, 8]. Currently available POU/HWT methods include boiling, chlorination, solar disinfection (SODIS), and bio-sand filters [9, 10]. These methods have proven effective in treating water, especially in eliminating pathogens. However, a challenge arises as chlorination and bio-sand filters require government assistance in providing the necessary equipment, while boiling and SODIS can be done without special equipment but have the potential for re-contamination [11]. Therefore, additional POU/HWT options are needed to address these challenges, particularly in West Sumatra.

Despite the various methods available for removing pathogenic bacteria from drinking water, adsorption is not popular. However, it is a well-known conventional method that eliminates almost all harmful

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microorganisms in drinking water [12]. Administer the powder after treating the water with a powdered adsorbent. One such adsorbent, layered double hydroxide (LDH), is gaining popularity in the scientific community due to its straightforward and inexpensive production and effectiveness in adsorbing pathogens. Research has also confirmed that LDH is safe for human use [13, 14]. However, when applied in drinking water treatment, the challenge lies in its nanomaterial form. To address this challenge, providing a structure to the adsorbent can make it easier to retrieve the adsorbent powder after the adsorption process.

The process of creating Mg-Al type LDH in situ was conducted by Fukugaichi et al. in 2022 [15]. In our study, we used an aluminum plate as a substrate to facilitate the movement of LDH when utilized as a water-purifying agent. We have redeveloped a single-step, in situ LDH manufacturing process. In this technique, LDH will be generated at the plate's surface by removing AlOH and then contacting aluminum with magnesium in one step or solution. We present a substitute way of POU/HWT that is low-cost, easy to apply, and suitable for disaster-prone areas, especially West Sumatra. We formed LDH on the surface of an aluminum plate by dipping it in seawater and $Mg(OH)_2$ fertilizer solution. This study is a feasible way to realize this method and to solve some problems caused during its realization in disaster areas.

2 Methods

A commercial aluminum square plate (A1100, PT. Surya Logam Universal, Indonesia), 25 cm^2 in area and 0.3 mm thick, was exposed to seawater from Padang Beach, West Sumatra, at pH 9.9, which had been adjusted by adding magnesium hydroxide, $Mg(OH)_2$, in a volume ratio of 100 mL. The plate was immersed for 24 hours at room temperature and then rinsed with distilled water. We prepared two soaking solutions—one with 0.5 g and the other with 1 g of $Mg(OH)_2$—and labeled the ensuing samples as LDH plate 0.5 and LDH plate 1, respectively. X-ray diffraction (XRD) was used to investigate the characteristics of these samples.

Before conducting the experiments on disinfection, all glassware and solutions were autoclaved at 121°C for 20 minutes in order to render them sterile. A tryptic soy broth solution with an initial content of 3.0×10^7 CFU/mL E. coli was serially diluted with sterile distilled water in order to prepare a working concentration of around 1000 CFU/mL. Following this, 100 mL of the freshly diluted bacterial suspension was introduced into 250 mL sample bottles (polypropylene).

The ability of the LDH plate to remove bacteria in clean water treatment was then tested in triplicate using 9 sample bottles. Each bottle contained approximately 1000 CFU/mL E.coli in conditioned distilled water. Three bottles were blank; three contained LDH plate 0.5, and the remaining three contained LDH plate 1. The sample bottles were agitated using a reciprocal shaker for 24 hours at a speed of 72 rpm and a temperature of 25ºC. Samples of treated water were collected at the third and 24th hours and then analyzed using the plate count method to determine the viable count of E. coli. The culture medium used in this counting method is Lysogeny Broth agar.

3 Results and discussion

3.1 Synthesis and characterization of LDH plate

LDH plate formation occurs via a number of interesting chemical changes, starting with the preparation of a solution to soak. This soak solution is prepared by adding $Mg(OH)_2$ into seawater and results in quite a visibly distinct white, cloudy mixture. With vigorous agitation, the magnesium hydroxide does not fully dissolve, and some residual material can clearly be seen at the bottom of the container.

An aluminum plate is then promptly submerged into this freshly concocted solution. Within minutes, bubbles will begin to form on the aluminum surface, which should indicate an event of central significance: the break-down of the protective aluminum oxide layer native to the metal's surface, an event allowing for direct contact between metallic aluminum and magnesium—present in both the seawater (0.05 mol/L) and the magnesium hydroxide (0.01 g/L).

This dynamic interaction is a two-step process. First, the thin aluminum oxide layer reacts with the abundant hydroxide ions in the solution. After that, this oxide layer dissolves, and the metallic aluminum underneath appears. This newly exposed aluminum subsequently reacts with the solution, producing soluble compounds and liberating hydrogen gas. The net result of all this complicated chemistry is the formation of an Mg/Al-type LDH layer on the surface of the LDH plate.

Normally, after immersion, the surface of the LDH plate is adorned with a white powdery precipitate of magnesium hydroxide. After carefully rinsing and wiping this off, the brownish LDH plate is revealed. This whole process occurs in just about the same way with both LDH plate 0.5 and LDH plate 1 ((Fig. 1(b) and (c)).

During the process, seawater acts as an important medium with its rich composition of different salts. Above all, seawater acts as an electrolyte and helps in the flow of ions, which is necessary for these chemical reactions to take place. The magnesium ions in seawater play a part in the formation of the Mg/Al LDH layer.

Magnesium hydroxide provides a supplementary source of magnesium ions and also helps raise the solution's alkalinity. Such an alkaline medium is essential for the solubility of the aluminum oxide layer.

Aluminum oxide is very thin but forms a strong, adherent protective film on the aluminum surface. The hydroxide ions in the solution react with the oxide layer and dissolve it into mobile aluminate ions.

When the barrier of aluminum oxide is broken, the metallic aluminum reacts with water in the presence of hydroxide ions to form the respective aluminate ions and hydrogen gas. These aluminate ions then react with magnesium ions, which are present both in seawater and in magnesium hydroxide, to form a precipitate of Mg/Al LDH. The constitution of this LDH may differ in different reaction conditions; however, it normally includes positively charged layers of magnesium and aluminum hydroxide intercalated with negatively charged anions and water molecules.

The excess magnesium hydroxide in the white precipitate on the LDH plate after immersion can be easily rinsed off to leave a brownish underlying LDH layer. The brownish color of the LDH plate suggests that some iron impurities might have been introduced into the LDH structure from aluminum or seawater. Iron tends to give its compounds a brownish hue.

There are complex chemical reactions in the plate formation process of an LDH. On the other hand, deep understanding may be important in optimizing the synthesis and properties of LDH materials, which have tremendous potential for a wide range of applications, from catalysis and environmental remediation to energy storage.

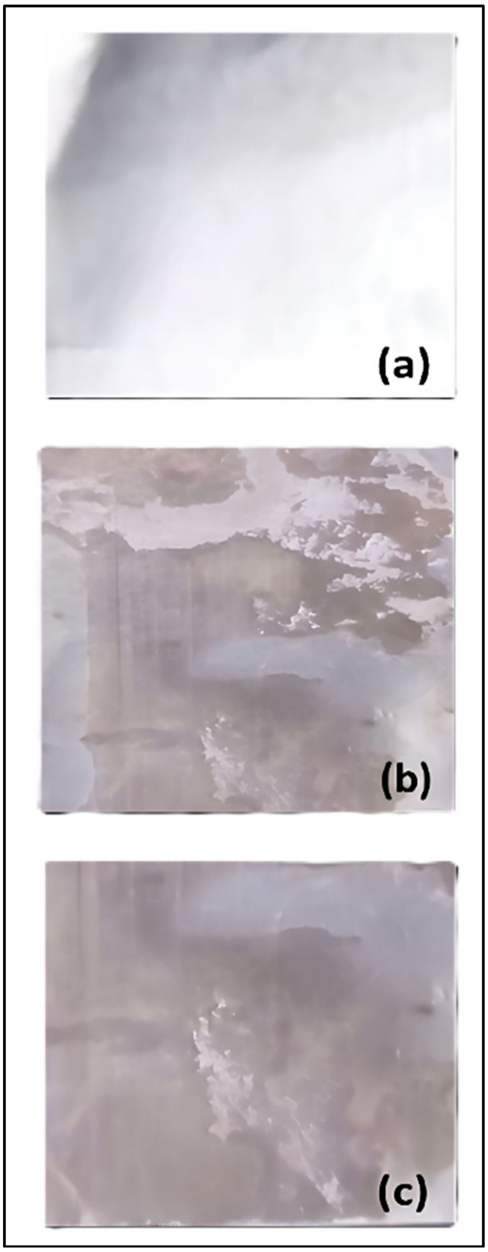

Fig. 1. (a) aluminum plate; (b) LDH plate 0.5; (c) LDH plate 1

Figure 1 shows the XRD pattern, which can be taken as conclusive evidence of the formation of an Mg-Al layered double hydroxide on the LDH plate surface. Characteristic peaks of Mg-Al LDH can be observed clearly enough, though broad in nature, which indicates a relatively low degree of crystallinity. This was not totally unexpected, considering the process of LDH formation onto this aluminum substrate is rapid and heterogeneous.

Notably, peaks for aluminum oxide are not found in the XRD pattern. This clearly implies that the native aluminum oxide layer on the aluminum plate has been completely dissolved by the soaking process. The dissolution of aluminum oxide already natively present from the aluminum plate is an important prerequisite to form the layer of Mg-Al LDH, exposing the metal aluminum to direct contact with reactive species in the soaking solution.

A comparison between the obtained XRD patterns of LDH plate 0.5, shown in Fig. 2(a), and LDH plate 1, shown in Fig. 2(b), gives a very fine but important difference. Obviously, peaks for LDH plate 1 have much stronger intensity than LDH peaks for LDH plate 0.5. This difference most likely implies that on LDH plate 1, more LDH has formed than on LDH plate 0.5, probably due to soaking conditions or due to features of the aluminum substrate itself.

The adhesion of the Mg-Al LDH layer to the underlying aluminum substrate seems to be pretty good since no LDH delamination or peeling is observed during sample preparation or analysis. This is very important for long-term stability and functionality with respect to the LDH plate, mainly under conditions that mean exposure to aqueous environments.

While Mg-Al LDH has been reported as safe and biocompatible [13, 14], its long-term safety is of essential interest, especially considering the long-time immersion in water. One can presume that the surface LDH layer would gradually dissolve over a very long period and release magnesium and aluminum ions from the surrounding area into the environment. A knowledge gap exists regarding the biological implications in this respect.

Further studies should be directed to the systematic assessment of the dissolution kinetics of Mg-Al LDH as a function of variable conditions, such as different pH, temperature, and ionic strength. These studies will help gain relevant insights into the long-term stability of the LDH plates and inform about safe and effective uses.

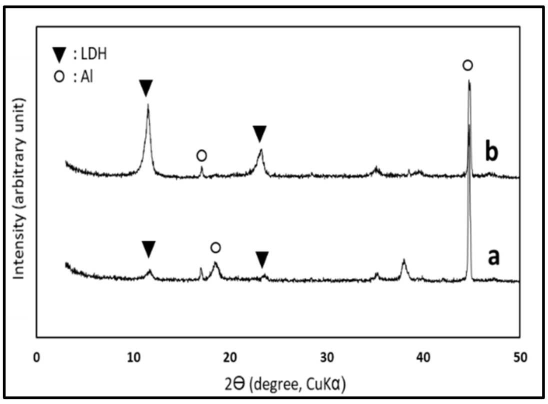

Fig. 2. XRD patterns of (a) LDH plate 0.5; (b) LDH plate 1

3.2 E. Coli removal test in water

The experimental results, exemplified by Fig. 3, show a good demonstration of the antibacterial properties of the LDH plates, particularly their potential for efficiently removing E. coli from an aqueous environment.

This control experiment, conducted in the absence of LDH plates, was important in creating a baseline for comparison, as there was no significant reduction in the viable count of E. coli bacteria even after an extended 24-hour incubation time. Therefore, this observation identified the resilience of E. coli and underlined that an active intervention needs to be implemented in order to eradicate it.

The introduction of LDH plate 0.5 into the E. coli suspension initiated a drastic drop in bacterial viability. Within 3 hours, the drop had reached as high as 87% in E. coli count. This antibacterial effect continued to increase over time until the complete eradication of E. coli after 24 hours.

LDH plate 1 demonstrated even more remarkable antibacterial power by reducing E. coli to nearly complete elimination, reducing it by 99% within the first 3 hours. After 24 hours, the count of E. coli was undetectable, which means the suspension was totally sterilized.

These results suggest that the superior performance of LDH plate 1 compared to LDH plate 0.5 is due to its higher LDH content, which was supported by the XRD measurements. This kind of observation could indicate a direct relationship between the amount of LDH present on the plate and its antibacterial efficacy. Increased LDH content on LDH plate 1 should have resulted from a higher $Mg(OH)_2$ dosage in its preparation.

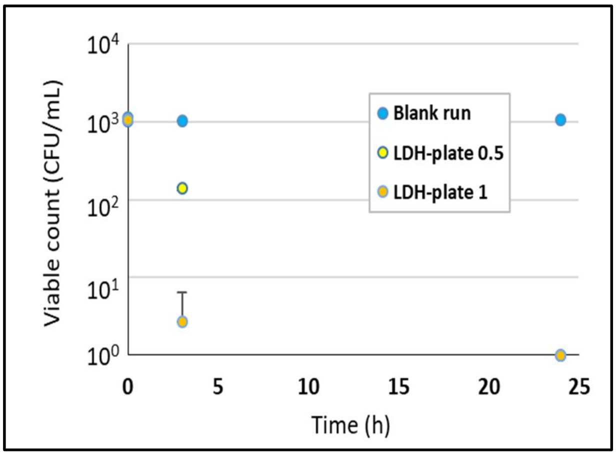

Fig. 3. The viable count of E. Coli was measured at different shaking times after adding LDH plates, starting with an initial viable count of approximately 10^3 CFU/mL. The viable count of LDH plates at 24 hours was determined to be 0 CFU/mL. error bars in the data represent the standard deviation.

The high capacity for E. coli removal that the LDH plates, particularly LDH plate 1, exhibited presents a strong potential for technologies in water purification and disinfection. Their efficiency in removing a common but highly pathogenic bacterium such as E. coli presupposes their wider application against other classes of microorganisms.

Future experiments are needed to evaluate the antibacterial spectrum of LDH plates against a number of bacterial and viral cultures. Investigations into long-term stability and reusability are also essential for the practical application of such plates. The optimization of LDH composition and load on the plates for the maximum expression of their antibacterial activity with the least possible adverse effects is another important direction for further investigation.

Given that LDH has the ability to attract bacteria due to its positive charge (since most bacteria and viruses carry a negative charge), this discovery confirms that bacterial adsorption occurs through an electrostatic mechanism [16, 17]. This adsorption was evidenced by a disinfection experiment in which LDH plates were immersed into agar-containing plates. After 24 hours, signs of E. coli colonies were observed on the plate surface (data not shown). This indicates that the LDH plate is adsorbing bacteria rather than killing them. Further research is necessary to understand better the process of capturing E. coli and to determine how long the bacteria can survive on the surface of the LDH plate.

Using LDH plate 1, we tested the potential for bacterial removal without stirring (Fig. 4). In samples without agitation, the number of live E. coli decreased by 41% after 24 hours. In samples with occasional stirring at hours 1, 3, and 5, the decrease in E. coli was 55% after 3 hours and 92% after 24 hours. This indicates the importance of stirring to allow the LDH plate to come into contact with water and adsorb E. coli.

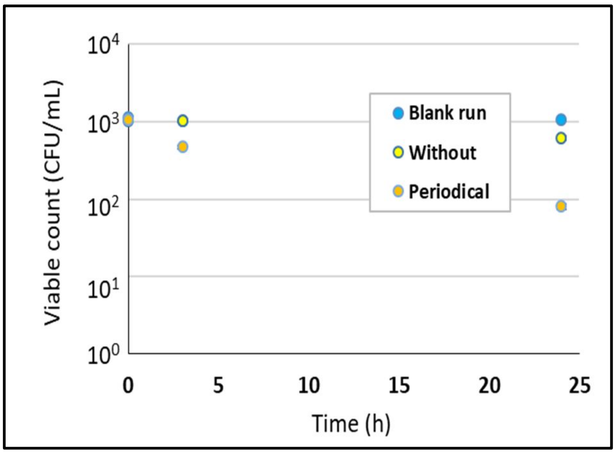

Fig. 4. The effect of mixing frequency without stirring and periodically stirring. error bars in the data represent the standard deviation.

This is a very complex phenomenon, given the many confluences of factors involved. The understanding of these factors is, therefore, highly important for the optimization of the design and application of LDH plates in any antibacterial setting.

The addition of $Mg(OH)_2$ used as a dosage in the preparation during the soaking solution is a very important process in the fabrication of LDH plates and has a very large degree of effect on the adsorption of the bacteria. While differences in $Mg(OH)_2$ dosage could indeed make a clear difference in E. coli removal at shorter time periods, for example, 3 hours, these differences seem to fade out over a long time period, for instance, 24 hours. This would imply that while the dosage of $Mg(OH)_2$ could affect the initial rate of bacterial adsorption, the ultimate adsorption capacity of the LDH plate is not very much influenced.

Stirring has a significant impact on bacterial adsorption to LDH plates. By offering continuous mixing and agitation, the act brings LDH plate surfaces into better contact with the bacterial suspension; therefore, successful adsorption is more likely to take place. Very interesting data shows that E. coli adsorption using stirring increases significantly.

Other factors that would probably exert a lot of influence on the adsorption of bacteria onto LDH plates, besides the amount of dosed $Mg(OH)_2$ and stirring, are:

1. The influence of the surrounding pH can change the surface charges of both the LDH plate and the bacterial cell, which, in turn, can lead to changes in potential electrostatic interactions between the two, hence affecting adsorption.
2. Temperature is an important factor that will influence adsorption kinetics. Temperature has a direct influence on metabolic activity and the viability of bacteria. The latter are factors that can regulate their adsorption behavior.
3. Finally, initial bacteria suspension concentration can have an effect on the quality behavior of the saturation kinetics of adsorption. Increased initial concentrations could lead to the earlier saturation of the LDH plate surface.

That is why these factors have to be examined in detail with regard to both their individual and synergetic effects on bacterial adsorption.

The detailed mechanisms of bacterial adsorption onto LDH plates are still under study, but some plausible models have been proposed, as shown in Fig. 5. One model based on the electrostatic attraction suggests that the positively charged surface of LDH plates could attract the negatively charged phosphate groups (PO_4^-) present in E. Coli. This could improve the electrostatic attraction between the coli cell surface and enable preliminary attachment of the bacteria to the LDH plate. One evident means is that phosphate groups on the outer surface of the bacteria exchange with the hydroxyl OH^- groups on the LDH plate. This form of ligand exchange would lead to a stronger and more time-proof binding of the bacteria on the LDH plate [18].

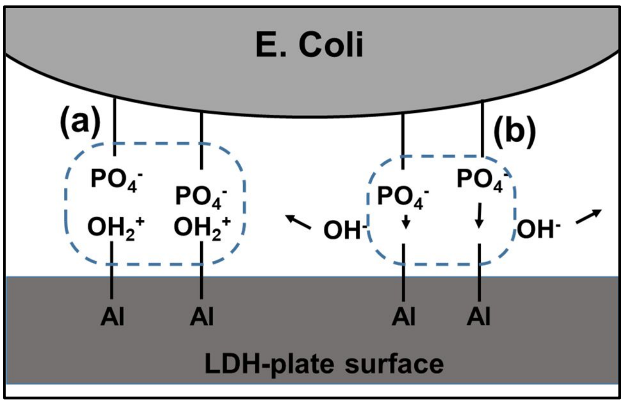

Fig. 5. Possible modes of E. coli binding to the LDH plate

Indeed, many studies have employed an ability to indicate aluminum minerals in soil, with a high number of attached microorganisms near them, and attached to $Al(OH)_3$ surfaces support a concept of electrostatic attraction as a major one to drive bacterial adhesion.

3.3 Future challenges

Our research on bacterial removal showcases the potential of using LDH plates as a practical and easy-to-produce disinfection tool. In West Sumatra, seawater can serve as a source of magnesium, and magnesium hydroxide ($Mg(OH)_2$), a commonly available fertilizer, enables the creation of highly feasible LDH plates.

Compared to most other POU/HWT methods used in disaster-stricken areas, the LDH plate method has a number of advantages. First, while chlorination and bio-sand filters require the assistance of the government for equipment, this method uses readily available materials at low cost, such as seawater and $Mg(OH)_2$ fertilizer. Second, while the boiling and SODIS methods involve risk due to re-contamination, the bacteria are adsorbed on the surface of the LDH plate. Such simplicity and low cost of the LDH plate method create opportunities for providing safe drinking water in emergency situations and resource-limited settings.

Although LDH plates represent an inexpensive and straightforward method for the purification of water, especially in emergency cases, further testing of these plates on other bacterial models and also on several natural sources of water is required. Also, as natural water can have a vast content of different organic compounds that may influence the extent to which microorganisms are present, it will be obligatory to perform tests on natural water samples, although it is put forward that, in general, low counts of E. coli bacteria are present in natural water [19].

4 Conclusion

In this study, a facile and effective method was developed for the removal of pathogenic bacteria from water using layered double hydroxides grown on an Al plate. An aluminum plate had to be immersed in a mixture of seawater and $Mg(OH)_2$ to form LDH, with a relatively high efficiency of removal for E. coli bacteria in water.

Beyond the efficacy of the method, it presents the cost-effective and facile execution of the technique, in particular in disaster-stricken areas like Sumatra Barat. Materials available on hand, or easily procured, are seawater and $Mg(OH)_2$, which are coupled with quite basic equipment: aluminum plates.

The research, however, also raised a number of further challenges that need to be addressed in future studies by testing on other bacterial models and natural water sources varying in different organic contents. More studies will hence be required to establish the efficacy and safety of LDH plates under more diversified and complicated conditions.

This research, therefore, contributes to the further development of point-of-use/household water treatment technologies by providing a practical, efficient means to produce clean drinking water in a disaster area.

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