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Evaluating the effectiveness of eggshells to remove heavy metals from wastewater

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ABSTRACT

Bio sorbent such as banana peel, pumpkin, papaya seed, crab shell, and eggshell uses effectively to remove contamination from the wastewater. This experiment is designed to evaluate the factors that influence the adsorption efficiency of eggshells to remove heavy metals particularly, Pb(II), Cu(II), and Zinc(II) from wastewater. Various methods were applied to determine the impact of temperature, size, and quantity of eggshell on the efficiency to remove heavy metal contamination from the wastewater. Results revealed that the efficiency of a sorbent is inversely related to the size of the sorbent. Moreover, higher efficiency was obtained at higher concentrations. The critical concentration lies between 20 and 25 (mg/L) where efficiency was at maximum level. Furthermore, adsorption efficiency was directly related to temperature after 35°C. Before 35°C, the variation in temperature did not show any impact on the efficiency. An optimal reaction temperature was determined at 65°C when the efficiency of adsorbent was at the maximum level. The study results provided the potential to get economic and environmental benefits by removing contamination from wastewater using eggshells.

Keywords: Adsorption; Sewage water; Eggshell powder; Lead; Copper; Zinc

1. Introduction

Water contamination is the main problem in resource-poor and industrial countries [1]. From 2.5% of freshwater, only 0.5% of water is available for humans to use [2]. Water contamination is an emerging problem caused by domestic, commercial, industrial, and agricultural waste [3]. Particularly, battery plants, pharmaceuticals, metal processing, hospitals, agrochemicals, and pesticides are releasing various types of heavy metals (zinc, copper, lead, mercury, and cadmium) that directly or indirectly discharge into surface water [4–8]. These toxics impact negatively on the ecosystem [9]. Heavy materials are non-biodegradable and adversely affect human health if consumed [10]. Various studies of heavy metals cause kidney, liver, memory problems, infertility, hepatotoxic, hemolysis, and cramps [11]. In the developing countries, 70%–80% of illnesses in

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the women and children were found due to the use of contaminated water [12]. In the reported literature, dealing with the control and the management of the water pollution as well as its related sources and effects, Owa [13] reported that 14,000 deaths per day in the world is mainly caused by water pollution. Hence, it is imperative to solve this situation by elucidating new methods susceptible to eliminate water contamination.

Portable water filters have been invented; however, these are expensive to buy especially for poor people. Adsorption, chemical precipitation, ion exchange, electrodialysis, membrane filtration, reverse osmosis, and flotation are few methods to remove heavy metals from aqueous solutions [11,14–17]. Biosorption is one of the methods that has been recognized as a reliable and economical method to remove heavy metal ions from aqueous solutions [18]. The adsorption method is easy handling and cost-effective [9,19,20]. The effectiveness of adsorption is more than 80% and it depends on the different factors such as structure, specific surface area of the adsorbent, and chemical nature of pollutants [21]. Various natural bio sorbents are available such as banana peel, pumpkin, papaya seed, crab shell, and eggshell which can be used effectively to remove contamination from the wastewater [6,22]. Removal of soluble microbial products and heavy metals from contaminated water using eggshell wastes is a cost-effective and efficient method [23]. The eggshell waste is available in abundance, global eggshell wastes generation is around 5.92 million per year and it may cause serious environmental consequences if it is not to be dumped into landfills.

Many studies have used eggshells to remove heavy metals from wastewater [17,24–26]. The effectiveness of eggshells to remove heavy metals from aqueous solution depends on various factors such as concentration, sorbent particle size, and temperature [27]. Removing contamination using eggshells is a cost-effective and efficient method because of its adsorbent ability [27,28]. The reason behind the effectiveness of eggshells may be two layers which make many circular pores [29,30]. Previous literature has focused on a particular size of eggshells; however, to the best of our knowledge, limited studies have focused on critical concentration and optimal temperature of eggshells during the treatment of wastewater [31–33]. Therefore, the current study examines the adsorption capability of eggshell as an effective adsorbent to remove contaminations (lead, copper, and zinc) from solutions. Batch adsorption experiments were carried out to evaluate the effects of process parameters such as (adsorbent size, initial metal concentration, and temperature) on adsorption efficiency and find out the critical concentration and optimal temperature.

2. Materials and methods

Chicken eggshells were washed five times with deionized water. The eggshells were then air-dried for 120 min and incubated in a hot air oven at 50°C for 35 min. This has been done to denature the organic proteins in the eggshells. The eggshells are divided into three batches. The first batch of eggshells was grinded into powder by a motor and further screened by no. 150 mesh. The second batch of eggshells was crushed by hand. While the third batch of eggshells was not crushed. Determination of metal ions using spectrophotometry is very interesting. The quantitative determination of metal ions is often carried out by spectrophotometry or extractive spectrophotometry, though other techniques, such as gravimetry, fluorimetry, and titrimetry are also employed. Among these methods, spectrophotometry is preferred because it is economical and easier to handle with comparable sensitivity, selectivity, and accuracy. Metals in aqueous solutions of eggshell were determined by flame atomic adsorption spectrophotometer (FAAS) based on size of eggshell, concentration, and temperature. It should be emphasized that similar methods to the one presented herein have been proposed by many researchers [1,4,12,34,35].

2.1. Characterization

2.1.1. Fourier transform infrared (FT-IR) spectrometry

The functionality of eggshells was investigated by Avatar 330 FT-IR Thermo Nicholet, (Thermo Electron Corporation, USA) with attenuated total reflectance (ATR) accessory (smart partner) within the range of 4,000–600 cm^{-1}.

2.1.2. pH meter

The pH of solutions was taken before and after adsorption by Orion 5-Star pH meter (Orion Private Ltd., Boston, MA, USA) connected with combined glass electrode and internal reference electrode was used to measure the pH of solutions. The quality of solution and performance of eggshell for removing heavy metals from solution was carried out using the (FAAS) and UV-visible spectroscopy.

2.1.3. Flame atomic adsorption spectrometry (FAAS)

Atomic adsorption studies were carried out on Elmer Perkin AA-800 (Elmer Perkin, Singapore) atomic adsorption spectrometer coupled with air-acetylene flame with standard burner head to compute the results and was operated at the conditions as prescribed by the manufacturing agency. The instrument was controlled by the computer with WinLab32 software. The analysis was performed at least in triplicate ($n = 3$) with integration time 3S and delay time 3S. FAAS is a very common technique for detecting metals present in samples. Metal ions in a solution are converted to atomic state by means of a flame. It is a very accurate quantitative and qualitative technique.

2.1.4. UV/visible spectroscopic studies

Over the last several years, an experimental and a theoretical picture of the usage of UV/visible spectroscopy in the water pollution field, particularly those dealing with hydrogen bonding, hydration, and adsorption of water and aqueous solutions has emerged which has proven to be useful for interpreting the main qualitative features of water pollution [36]. The UV-visible spectroscopy is a very powerful tool for studying the structural configuration, nature of bonding, and transitions in compounds. It is important to note that the adsorption of visible and ultraviolet radiation is associated with the excitation of electrons which allows to determine the directly and/or inversely proportion to the percent transmittance and/or absorbance as well as the precision and accuracy of the different measurements. Moreover, we must underline that the reproducibility of the results remains a challenging problem in this field [37].

Over the years, there has been much interest in the biological treatability of chemical and petrochemical wastewaters. Indeed, many methods have been developed and discussed the role played by organic and inorganic compound mixtures encountered in chemical and petrochemical wastewaters [38]. Within the context of these methods, it was shown that UV analysis is easy, economic, less time consuming, and better performed for understating and/or underlying molecular interactions within wastewaters in contrast with other procedures which has been demonstrated to be not suitable for complex mixtures such as wastewaters. Spectrophotometric studies were performed on double beam Perkin Elmer 35 spectrophotometer (Perkin Elmer, Singapore) with dual 1 cm quartz cuvettes. The computer-controlled UV-vis instrument with Win lab Revision-6.0.0.0718 software.

2.2. Preparation of standard solutions

Stock solution (1,000 mg/L) of Pb(II) was prepared by dissolving the appropriate weight of pure salt $Pb(NO_3)_2$ in deionized water. As an ionic substance, the dissolution of lead(II) nitrate involves dissociation into its constituent ions.

$$[Pb(NO_3)_2(s) \rightleftharpoons Pb^{2+}(aq) + 2NO_3^-(aq)] \quad (1)$$

Stock solution (1,000 mg/L) of Cu(II) was prepared by dissolving the appropriate weight of copper sulfate ($CuSO_4$) in distilled water.

$$[CuSO_4 \rightleftharpoons Cu^{2+}(aq) + SO_4^{2-}(aq)] \quad (2)$$

Stock solution (1,000 mg/L) of $ZnCl_2$ was prepared by dissolving the appropriate weight of zinc chloride ($ZnCl_2$) in distilled water.

$$[ZnCl_2 \rightleftharpoons Zn^{2+}(aq) + 2Cl^-(aq)] \quad (3)$$

The adsorption efficiency of Pb(II), Cu(II), and Zn(II) was measured and compared using FAAS.

3. Results and discussions

In order to study the binding mode of the various groups present in the egg shell, the IR spectra of the egg shell were recorded to identify the coordination of these groups in the solid-state. The IR spectral data also supported the structure with peaks appeared at cm^{-1} 3,247 assigned for OH, sharp peak at cm^{-1} 1,393 ascribed for –C–O or CH_3 bending, 1,084 for C–O, 712 for –CH group, (Fig. 1). Ali et al. [34] reported in the study on the adsorption of Cu(II) and Cd(II) ions onto eggshell and observed OH group was present in the eggshell powder.

To study the effect of pH on adsorption process, the influence of pH on the adsorption of lead(II), copper(II), and zinc(II) using eggshells was carried out in the range 3–10 at different pH. A series of buffer solutions each differing by 0.5 pH unit intervals was prepared and in the presence of these buffers, the percentage removal of metals lead, copper, and zinc were checked as per the procedure. The percentage removal efficiency values of each of the solutions were measured. From Fig. 2, it is observed that the percentage removal of all three metals (i.e., lead, copper, and zinc) was increased with an increase in pH value from 3.5 to 6.5. While further increase in pH value from 6.5 to 10, it started decreasing. It may be revealed due to active adsorption sites remain protonated by hydrogen ions at lower pH, and become less available for metal adsorption. As pH increases, H^+ ion concentration decreased, hence they did not compete with metal ions on the adsorption sites. The results are in consistency with existing literature, that optimum pH value was 6.0 to remove lead and copper using chitosan composites and garden grass, respectively [39–41].

We have taken 20 g of eggshells of different sizes which were added in 20 mL of 100 mg/L Pb(II), Cu(II), and Zn(II) aqueous solution. The mixture was shaken for 1 h

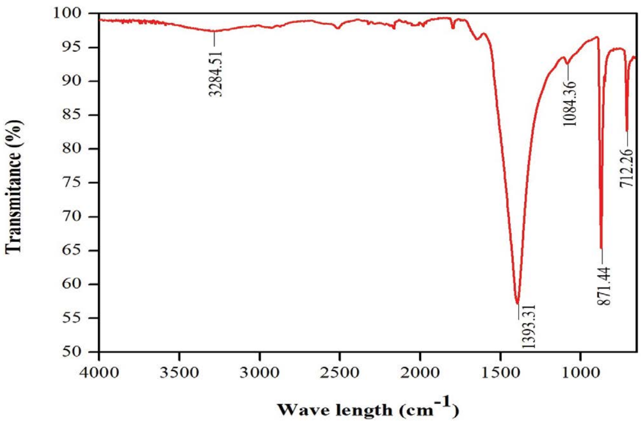

Fig. 1. FTIR spectra of eggshell powder.

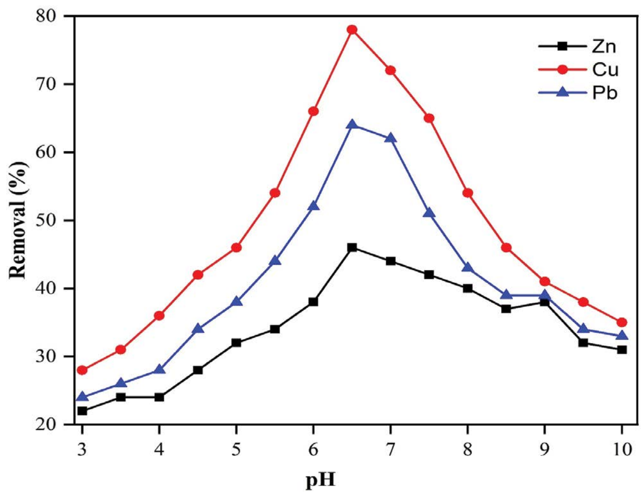

Fig. 2. Relationship between pH and ions removal.

in an orbital shaker Allerga 64 R Avanti™ 30 (BECKMAN COULTER, USA) centrifuge with a speed of 80 rpm in a water bath having a temperature at 25°C. It was separated by filtration and the solution was analyzed by UV spectrometer and FAAS and compared with the control setup. A 20 g of each powdered, slightly crushed, and uncrushed eggshells were added into separate syringes which were sandwiched by cotton wool from both sides. The cotton was secured and tied by a mesh. 20 mL of 100 mg/L Pb(II), Cu(II), and Zn(II) aqueous solutions were pumped through each syringe. Two sizes were considered for each batch to test. We should underline that the spectrophotometry

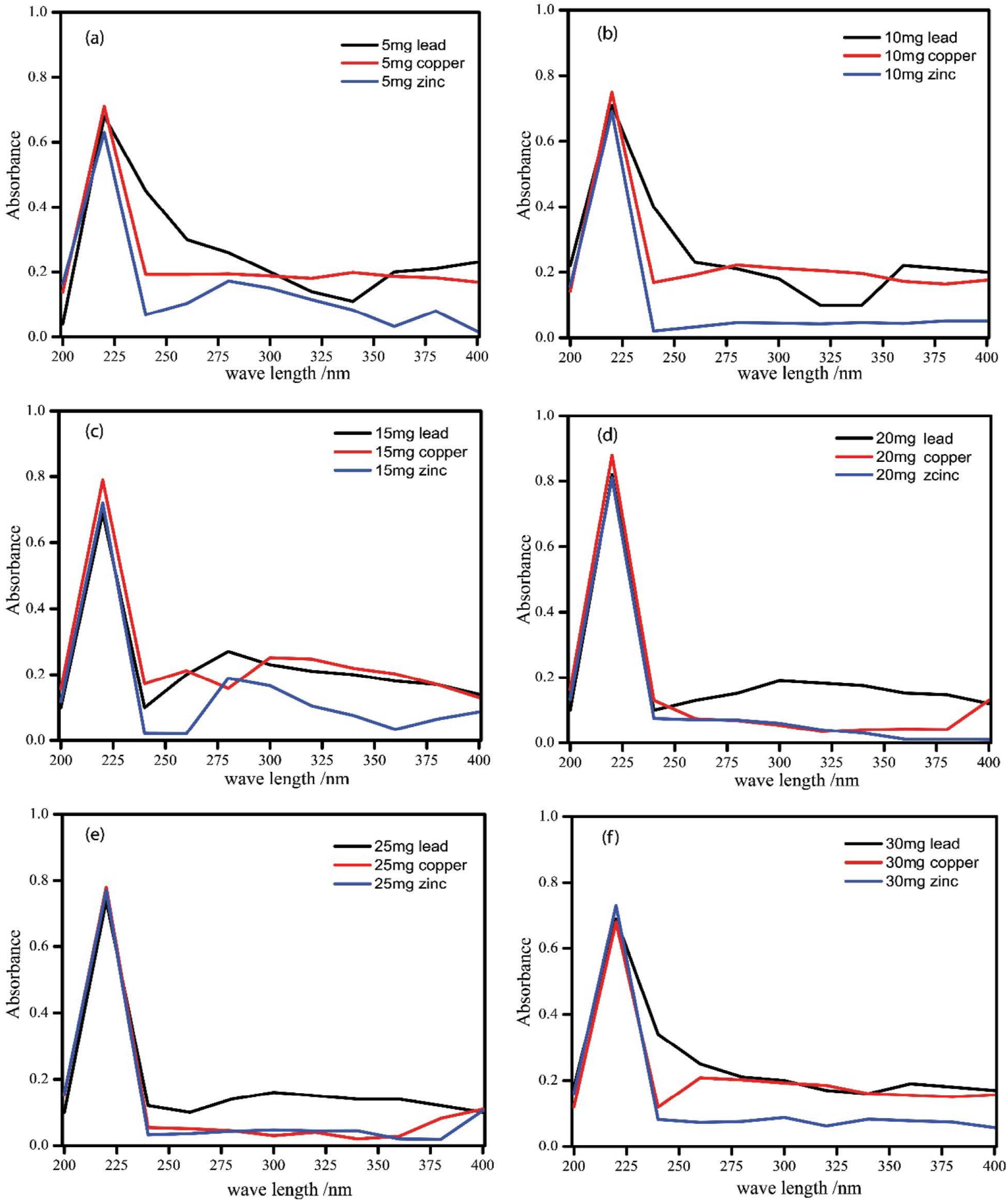

Fig. 3. UV-visible absorbance spectra of an aqueous solution of lead, copper, and zinc.

UV-visible technique has been extensively used by many groups [36–38]. Indeed, UV-visible spectroscopic studies provide adequate information regarding the adsorption of visible and ultraviolet radiation which is associated with the excitation of electrons. A similar type of band was observed in the range 200–250 nm was assigned to the $\pi \rightarrow \pi^*$ transitions due to pi (π) electrons in all solutions of metals Pb, Cu, and Zn containing different concentrations with eggshell as in Figs. 3a–f. This band is a general characteristic of metals with eggshell and agrees with the literature studies. The adsorption bands of the metals are assigned to the adsorption based electronic transitions. Metal induced transitions were observed at different concentrations studied. Furthermore, the study of the effects of varying the concentrations, 5, 10, 15, 20, 25, and 30 mg, of metals (lead, copper, and zinc) were carried out using a UV-vis spectrometer and FAAS. The better results obtained at the 20 and 25 mg that were considered as optimal concentrations. Moreover, for all concentrations, it was observed that copper showed significant adsorption followed by lead and zinc.

The efficiency of adsorption can be measured as:

$$R = \left[\frac{\left(C_0 - C_e\right)}{C_0} \right] \times 100 \quad (4)$$

where C_0 and C_e represent the initial and equilibrium contaminant concentrations, respectively. While R is the removal efficiency. The efficiency of all three types of eggshell (powder, crushed, and uncrushed) was analyzed by (FAAS) (Table 1).

The filtration resistance may increase due to solubilization in eggshells. When compared to the same quantity of eggshells with different sizes, a smaller particle size (powder) of eggshell provided higher solubilization and a higher value of filtration resistance. The efficiency of adsorption (R) was decreased with an increase in sorbent diameter and vice versa. During the experiment, the suspensions of water and eggshell were treated by ultra filtration. After the testing of particle size, another test was conducted to determine the effect of concentration on the adsorption efficiency. Four different concentrations with the same amount of eggshell powder were analyzed. It was found that in the sample of eggshells, the adsorption efficiency was increased with an increase in sorbent concentration (Table 2).

The removal efficiency (R) was initially increased with concentration, while it was decreased after certain concentration for all the examined sizes. Furthermore, it was observed that critical concentration lied between 20 and 25 mg/L, and efficiency for Cu and Pb was 82% and 97%, respectively. Similar results have found in the recent literature [42]. Moreover, the effect of temperature on removal efficiency was also analyzed and presented in Table 3. The results showed a slight change in removal efficiency due to a change in temperature between 25°C and 35°C in all three cases. As the temperature increased, the adsorption of eggshell powder was also increased. The faster motion of particles was associated with an increase in temperature. The experiment was performed at different temperatures, best results were obtained at 65°C, hence, it was considered as an optimal condition of temperature. A similar type of result was obtained when the lead was removed from an aqueous solution using waste tire rubber [15]. There was a

Table 1
Eggshell size and adsorption efficiency of Pb(II), Cu(II), and Zn(II)

Batch	Eggshell size (mm)	Revolution per minute	Adsorption efficiency (%)		
			Pb	Cu	Zn
Uncrushed	10	80	65	60	53
	5	80	73	68	59
Slightly crashed	1	80	83	79	67
	0.75	80	88	83	73
Powder	0.5	80	93	89	79
	0.25	80	97	94	83

Table 2
Initial concentration of aqueous solution and adsorption efficiency of Pb(II), Cu(II), and Zn(II)

Serial number	Quantity of eggshell powder (g)	Initial concentration of Pb, Cu, and Zn (mg/L)	Adsorption efficiency (%)		
			Pb	Cu	Zn
1	20	5	82	75	70
2	20	10	85	77	73
3	20	15	88	79	76
4	20	20	92	94	79
5	20	25	84	88	71
6	20	30	94	85	69

Table 3
Temperature and adsorption efficiency of Pb(II), Cu(II), and Zn(II)

Serial number	Quantity of eggshell powder (g)	Temperature (°C)	Adsorption efficiency (%)		
			Pb	Cu	Zn
1	20	50	88	85	80
2	20	55	90	88	83
3	20	60	94	91	87
4	20	65	97	94	89
5	20	70	93	90	81

significant impact of temperature on the removal efficiency of heavy metals Cu(II) and Cd(II) from aqueous solution [34].

4. Conclusion and policy implications

The study was designed to evaluate the efficiency of the eggshell to remove heavy metals from sewage water with an aqueous solution of Pb(II), Cu(II), and Zn(II). A set of experiments was carried out with different fixed conditions of eggshell sizes. Furthermore, the effect of concentration and temperature was also studied. Results revealed that the efficiency to remove contamination was more than 90% with the use of the powdered eggshells. Furthermore, the impact of concentration of aqueous solution had also been tested and found a critical concentration for higher adsorption efficiency which lied between 20 and 25 mg/L. The removal efficiency of Pb(II) was observed 90%, whereas Cu(II) and zinc(II) were 80% at critical concentration. Moreover, the temperature of eggshells also affected the removal efficiency to remove contamination from the wastewater. Specifically, the high temperature significantly influenced the adsorption efficiency of the eggshells. It was observed that the small change in temperature did not influence the adsorption efficiency. When the temperature of eggshells was increased, the adsorption ability of contamination from wastewater was significantly increased. However, the adsorption ability was varied according to the temperatures. Using of eggshells to clean wastewater will significantly improve the economic benefits and produce less environmental pollution. Finally, it is necessary to note that a complementary study should be conducted to improve the existing results by modifying other parameters related to the eggshell. The effects of extending the current study by varying chicken eggshell adsorbents using acids, bases, or some other bio sorbents should be the subject of future work.

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