Adsorption of phosphate by Zn/Cr layered double hydroxides

By

Karin Hansson
Abstract

The concentrations of phosphorous, chromium and zinc are increasing in the water due to human activities. High levels of these elements have adverse effects on humans and other organisms. Thus, methods for removal of these substances from wastewaters have gained an increased interest.

Layered double hydroxides, LDHs, are a group of inorganic clays. Due to its feature as an effective anion adsorbent it is a possible method for removal of phosphate. A further advantage of using LDHs in wastewater treatment processes is that it can be synthesized from heavy metals recovered from plating wastewater.

Based on the adsorption and ion exchange theories, this thesis studies the possibilities of using a Zn/Cr LDH in order to remove phosphate. The Zn/Cr LDH was synthesized by co-precipitation of zinc and chromium. The removal rate of phosphate due to different Zn/Cr molar ratios and pH are investigated and discussed. Preliminary research of the removal mechanism by the isothermal model and X-ray diffraction spectrum is also performed.

The results showed that the most efficient Zn/Cr ratio for phosphate adsorption was 4:1, whereas the optimum pH was 3. When comparing the XRD spectrum to the standard spectrum of a Zn/Cr LDH, the prepared adsorbent demonstrated similar peaks, indicating that the adsorbent indeed was a Zn/Cr LDH.

Keywords: phosphorus pollution, wastewater treatment, layered double hydroxides, adsorption
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1 Introduction

1.1 Heavy metal pollution
Heavy metals released from industrial activities are an emerging problem. One of the main sources of heavy metals such as zinc and chrome is the industry of electroplating and metal finishing. The undesirable and hazardous effects of increased levels of heavy metals are many and treatment of the wastewater from these industries is necessary. However, the present methods for removal of these toxic substances produce large amounts of sludge that is in need of further treatment processes [1]. Because of this, more efficient methods for water treatment are required.

1.2 The phosphate problem of today
The concentration of phosphates in the sea and in lakes has increased during the last century mainly due to fertilizers and leakage from industries [2]. A consequence of high phosphate concentrations in the water is eutrophication, which causes disorders in the ecosystems like algal blooms. In severe cases, high phosphate levels in lakes can lead to hypoxia i.e. oxygen depletion [3]. Eutrophication is however not the only problem concerning phosphate. As all living cells consume phosphate, the phosphate demand is high. We are currently exhausting the sources of phosphate in a rate that will lead to a total lack of phosphate in 50-100 years [4].

1.3 Removal of phosphates
The eutrophication and the phosphate depletion have increased the demand in sustainable techniques for both removal and recover of phosphate from wastewaters. There are currently two main techniques for removal of phosphates from wastewater; biological and chemical treatment [5].

1.3.1 Biological treatment
The biological method consists of an active sludge treatment process, where phosphate adsorbing bacteria are formed. The phosphate rich sludge can then be recovered, leaving the wastewater with a phosphate concentration of about 5% [5]. However, there are several shortcomings with this method. The method is expensive and as it depends on bacteria, certain conditions must be employed in order to avoid situations such as floc formation or toxic chocks [6].

1.3.2 Chemical treatment
The chemical method causes the phosphate to precipitate by utilization of aluminium or iron salts [7]. Even though this method is highly efficient, it demands accurate and variable chemical dosing due to the variations in both quality and quantity of the wastewater. This method also produces a considerable amount of sludge, which requires further treatment [7].

1.3.3 Adsorption
Due to the shortcomings of the conventional wastewater treatment methods, there is still a need for an economical, efficient and sustainable way to remove and recover phosphate. The interest in adsorbents e.g. layered double hydroxides, LDHs as a possible way of removing and recover phosphate has increased lately. The advantage by using LDHs in water treatment plants is mainly their economical benefits but also that the hydroxides can consist of natural occurring and abundant minerals. Recent studies show that these minerals can be recovered from plating wastewaters [8]. Hence, LDHs can be used both for removal and recover of metals such as zinc and chromate, but also oxyanions like phosphate. Furthermore, LDHs has a higher anion exchange than the conventional methods.
1.4 Layered Double Hydroxides

1.4.1 Structure

Layered double hydroxides are a class of nanostructured anionic clays. The structure of LDHs can be described as two-dimensional with positive charged sheets consisting of octahedral units. These sheets are balanced by negatively charged interlayers. A common feature is hydrogen bonded water molecules occupying the free space in the interlayer regions [9]. The interlayers bond weakly, and can therefore be used to capture both organic and inorganic anions like phosphate.

![Layered Double Hydroxides Structure](image)

Figure 1: The general structure of LDHs [9].

The general formula to represent LDHs can be written as [9]:

\[
[M_{2+}^{2+}M_{3+}^{3+}(OH)_{2}]^{x+}(A^{-n})_{x/n} \cdot mH_2O
\]

In this formula, \(M_{2+}^{2+}\) and \(M_{3+}^{3+}\) represent divalent and trivalent cations, respectively. In this study, \(M_{2+}^{2+}\) corresponds to \(Zn^{2+}\) and \(M_{3+}^{3+}\) to \(Cr^{3+}\). \(A\) is the interlayer anion of valence \(n\). As the character of \(M_{2+}^{2+}\), \(M_{3+}^{3+}\) and \(A\) can vary to a great extent, there are many possible LDH configurations. The most common interlayer anion is carbonate, but anions such as halides, oxyanions and organic anions may also be accommodated [9].

1.4.2 Synthesis

LDHs occur naturally i.e. hydrotalcite \((Mg_6Al_2(OH)_{16}CO_3 \cdot 4H_2O)\) [10], but they are also relatively simple to synthesize. There are however, several different techniques for this purpose. The most common method is co-precipitation [9]. The idea of co-precipitation is that an aqueous solution containing \(M_{2+}^{2+}\), \(M_{3+}^{3+}\) and the anion that is to be incorporated into the interlayer, precipitates under supersaturation. The precipitation is then followed by a heating and an ageing process. This is to increase the yield and the crystallinity of the material.

The desired interlayer anion can also be incorporated by the ion exchange method. In this...
method, the present interlayer anions are exchanged by another type of anions [9].

Another way of replacing the accommodated anions is through the structural “memory effect”. In this method, water, anions and hydroxyl groups are removed by calcination of the LDHs. This results in mixed metal oxides with the ability to regenerate the LDH-structure when exposed to water and anions. These anions do not need to be the same ones that were incorporated before the calcination [9].

1.4.3 Removal of oxyanions
An interesting application of LDHs is the adsorption of contaminants such as oxyanions like phosphate, arsenate and nitrate. In order to remove contaminants, three different methods have been proposed. These methods are surface adsorption, interlayer anion exchange and reconstruction of calcined LDH precursors by the memory effect [9].

There are several factors contributing to the efficiency of the oxyanion removal by the LDHs. Among these are the temperature, the pH of point of zero charge, the presence of competitive oxyanions, the nature of the cations and the layer charge [9].

1.5 Purpose
The aim of this study is to prepare and examine the sorption behaviour of Zn/Cr layered double hydroxides. The influence of the Zn/Cr ratio, pH and temperature will be analysed and discussed. To examine the structure of the prepared adsorbent, X-ray diffraction will be used.
2 Experimental

2.1 Materials

2.1.1 Chemicals
Table 1: List of the chemicals used during the experiments.

<table>
<thead>
<tr>
<th>Name</th>
<th>Formula</th>
</tr>
</thead>
<tbody>
<tr>
<td>Zinc sulphate heptahydrate</td>
<td>ZnSO₄•7H₂O</td>
</tr>
<tr>
<td>Chromium(III) sulphate</td>
<td>Cr₂SO₄</td>
</tr>
<tr>
<td>Phosphate solution</td>
<td>G</td>
</tr>
<tr>
<td>Ascorbic acid</td>
<td>C₆H₈O₆</td>
</tr>
<tr>
<td>Ammonium molybdate</td>
<td>(NH₄)₆Mo₇O₂₄•4H₂O</td>
</tr>
<tr>
<td>Sodium hydroxide</td>
<td>NaOH</td>
</tr>
<tr>
<td>Sulphuric acid</td>
<td>H₂SO₄</td>
</tr>
</tbody>
</table>

2.1.2 Instruments
Table 2: List of the instruments used during the experiments.

<table>
<thead>
<tr>
<th>Instrument</th>
<th>Model</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH-meter</td>
<td>SCHOTT Instruments, Lab 850</td>
</tr>
<tr>
<td>UV-spectrophotometer</td>
<td>UV-1800 Shimadzu</td>
</tr>
<tr>
<td>Spectrophotometer</td>
<td>XinMao 721</td>
</tr>
<tr>
<td>Scale</td>
<td>Mettler Toledo</td>
</tr>
<tr>
<td>Centrifuge</td>
<td>Anke TGL-16G</td>
</tr>
<tr>
<td>X-ray diffractometer</td>
<td>Rigaku Ultima IV</td>
</tr>
</tbody>
</table>

2.2 Method

2.2.1 Standard curve drawing
Phosphate standard solution of the following amounts was added to colorimetric tubes; 0.0, 0.05, 0.3, 0.5, 1.0 and 15.0 mL. 50mL distilled water was added to each tube. 1 mL ascorbic acid was then added and after 30 seconds, 2 mL ammonium molybdate were added. Each tube was then mixed. After 15 minutes, the solutions were tested at the maximal wavelength of 650nm in a UV-spectrophotometer. A standard curve could then be drawn by plotting the absorbance as a function of the known phosphate concentrations.

2.2.2 Preparation of the adsorbent
The adsorbents were prepared by co-precipitation according to the following: A fixed amount of zinc sulphate heptahydrate with the concentration of 0.5 mol/L was added four beakers together with different amounts of chromium (III) sulphate solution. This was in order to gain the four different Zn/Cr ratios 1:1, 2:1, 3:1 and 4:1. The beakers were then put into a water bath with a temperature of 80°C. Sodium hydroxide (20%) was added drop by drop until the solutions reached pH 9. The beakers were then kept in the 80°C water bath kettle for 18 hours. After 18 hours, the products were cooled in room temperature. They were then filtered and washed before putted in an 80°C drying oven for 24 hours. The products were then grinded into powder.
2.2.3 Impact test of phosphate removal rate

0.05 g of each adsorbent with different Zn/Cr ratio was put in 150 mL serum bottles. 50 mL phosphate solution (50 mg/L) with pH 7 was added to the bottles. Each sample was made in duplicates.

The solutions were stirred for 24 hours in room temperature by a rocking table. The stirring speed was set to 170 rpm. After 24 hours, 5 mL of each sample were taken out and centrifuged at 8000 rpm for 5 minutes. The liquid supernatant was then retained.

1 mL of each supernatant was taken out and diluted with 24 mL distilled water. 1 mL ascorbic acid was added, and after 30 seconds, 2 mL ammonium molybdate were added. Each sample was then mixed. After 15 minutes, the samples were transferred into cuvettes. The absorbance was then measured with a spectrophotometer. The phosphate concentration in the residuals could be determined with the standard curve.

2.2.4 Impact test of phosphate removal rate with pH

The pH of the phosphate solution was adjusted to 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12 and 13 respectively, with sodium hydroxide and/or sulphuric acid. The different phosphate solutions were then added to 0.05 g of the best adsorbent (4:1). Each sample was made in duplicates.

The solutions were stirred for 24 hours in room temperature by a rocking table. The stirring speed was set to 170 rpm. After 24 hours, the concentrations of phosphate in the different solutions were tested by a spectrophotometer.

2.2.5 Adsorption isotherm experiment

The adsorbent that in the previous experiments showed the highest phosphate adsorption was weighed to 50 mg and put in serum bottles. The pH of the phosphate solution was adjusted to 3. Then 50 mL phosphate solution of different concentrations was added. The phosphate concentrations were: 25, 50, 75, 100, 125, 150, 175 and 200 mg/L respectively. The samples were then stirred for 24 hours at 30°C. After that, the samples were centrifuged according to the previous methods. The obtained liquid supernatant was diluted to fit the standard curve (for dilution factors, see appendix). The samples were then analysed by the UV- spectrophotometer.
3 Results and discussion

3.1 Standard curve

The standard curve can be used in order to determine the concentration of phosphate in a solution with known absorbance.

As the $R^2$ value is close to 1, the linear function displayed in the graph is an accurate linearization.

3.2 Impact test of phosphate removal rate with different Zn/Cr ratios

Figure 3 shows the removal rate of phosphate for the different Zn/Cr ratios. The amount adsorbent of different ratios used in the experiment was 50 mg. 50 mL of phosphate solution with a concentration of 50 mg/L were added to the adsorbents. The initial pH for the experiment was 7 and the reaction time was 24 hours.

As can be seen in figure 3, the LDH with a Zn/Cr ratio of 4:1 proved to be the most efficient phosphate adsorbent. The figure also shows that the LDH with a Zn/Cr ratio of 1:1 adsorbed the least. A possible explanation for this behaviour might be that when the Zn/Cr ratio increases, there will be more reaction sites and more space and in the interlayer region for the phosphate to...
occupy. This is due to that the ionic radius of the hydrated zinc ions is smaller than the one of chromate [11]. A small ionic radius corresponds to a small specific surface area which in turn correlates to a higher specific surface energy. A higher Zn/Cr ratio thus denotes more space and reaction sites in the interlayer region for the phosphate.

The removal rate for the 4:1 ratio is close to 90%, which confirms that this ratio indeed is the most efficient phosphate adsorbent. The graph shows that the 3:1 ratio is less efficient than both 2:1 and 4:1. The reason for this behavior is unknown and might be caused by different factors of error. A possible factor of error might be inaccurate measuring of the phosphate solution.

Further experiments to examine the adsorption behavior at higher Zn/Cr ratios are essential in order to find the most effective adsorbent.

### 3.3 Impact test of phosphate removal rate with pH

Figure 4 displays the removal rate of phosphate as a function of pH. 50 mg of the LDH with a Zn/Cr ratio of 4:1 was used for the experiment as it showed to be the most efficient adsorbent in the previous experiment. The concentration of the phosphate solution was 50 mg/L and the reaction time was 24 hours.

![Removal rate vs pH](image.png)

**Figure 4: The removal rate of phosphate as a function of pH.**

As shown in figure 4, the optimum pH is around 3. The reason for this might be that the LDH is partly dissolved in the acid environment below pH 3. Hence, the structure is not suitable for adsorption. A future experiment might be to examine the actual structure of the LHD as a function of different pH values.

When the pH increases, the removal rate decreases. A possible explanation might be that the surface charge of the adsorbent decreases with increasing pH. Below the pH of point of zero charge, the surface becomes negatively charged and consequently it repels the negatively charged phosphate ions.

However, in a future scenario where LDHs are used for removal and recovery of heavy metals and oxyanions, a low pH value in the treatment process is not desirable since the solubility of heavy metals increases with decreasing pH.

### 3.4 Adsorption isotherms

Figure 5 shows the adsorption capacity as a function of the phosphate concentration in the residuals. The unit for the adsorption capacity is mg phosphate per g adsorbent. The pH in the solution before the adsorption was 3. The experiment was performed on 50 mg of the LDH with
a Zn/Cr ratio of 4:1. The temperature during the experiment was approximately 30°C and the reaction time was 24 hours.

![Adsorption isotherm](image1)

**Figure 5:** The adsorption isotherm for phosphate adsorption at 30°C.

According to figure 5, the adsorption capacity seems to increase as the concentration of phosphate in the solution increases. However, the adsorption capacity will probably reach a saturation point when the adsorption capacity remains constant, even though the phosphate concentration increases. The phosphate concentrations that were tested in this experiment were probably too low to reach this point.

The following two graphs (figure 6 and 7) show the adsorption isotherm fitted to the Langmuir and the Freundlich equations.

![Langmuir](image2)

**Figure 6:** The Langmuir adsorption curve.
The $R^2$ values for neither the Langmuir nor the Freundlich curve fitting were satisfying. Consequently, further experiments for the adsorption at higher phosphate concentrations are required in order to decide which adsorption isotherm that is the most applicable.

### 3.5 LDH characterization

Figure 8 displays the X-ray diffraction spectrum of the Zn/Cr LDH before the adsorption of phosphate. The diffraction scatter can reveal information about the structure of the adsorbent.

![XRD spectrum before adsorption](image)

**Figure 8:** The XRD spectrum for the Zn/Cr LDH before the adsorption of phosphate.

<table>
<thead>
<tr>
<th>Peak Position (°)</th>
<th>31.76</th>
<th>34.44</th>
<th>36.28</th>
<th>47.6</th>
<th>56.62</th>
<th>62.91</th>
<th>97.97</th>
</tr>
</thead>
</table>

**Table 3:** The positions of the peaks in the XRD spectrum displayed in figure 8.

Figure 9 shows the XRD spectrum of the Zn/Cr LDH after the adsorption of phosphate. The pH of the phosphate solution was 3.
When comparing figure 8 and 9, the peaks from the spectrum before the adsorption still remains after the adsorption. However, there are some new peaks after the adsorption. These are probably due to the change in the structure as a result of adsorption at the surface.

**Table 4: The positions of the peaks in the XRD spectrum displayed in figure 9.**

<table>
<thead>
<tr>
<th></th>
<th>17.41</th>
<th>18.26</th>
<th>19.37</th>
<th>20.12</th>
<th>22.18</th>
<th>22.88</th>
<th>25.70</th>
<th>26.26</th>
<th>31.32</th>
</tr>
</thead>
<tbody>
<tr>
<td>Value</td>
<td>31.74</td>
<td>34.38</td>
<td>36.2</td>
<td>46.79</td>
<td>47.59</td>
<td>56.58</td>
<td>62.91</td>
<td>67.95</td>
<td>69.15</td>
</tr>
</tbody>
</table>

Figure 10 displays the standard spectrogram of the Zn/Cr LDH.

Judging from the standard spectrogram presented in figure 10, the examined adsorbent has indeed several peaks in the same positions as a Zn/Cr LDH. There are yet some additional peaks in both figure 8 and 9, but these might originate from impurities. It is also possible that the spectrums of the LDHs showed in figure 8 and 9 differ from the standard spectrum due to the low pH that might have caused the LDHs to partly dissolve. To further ensure the nature of the adsorbent, a new XRD spectrums that includes the diffraction pattern at lower degrees are required.

**Table 5: The position of the peaks in the standard spectrogram**

<table>
<thead>
<tr>
<th></th>
<th>11.44</th>
<th>23.23</th>
<th>34.76</th>
<th>39.32</th>
<th>46.68</th>
<th>60.64</th>
<th>62.04</th>
</tr>
</thead>
</table>

### 4 Conclusions

The experiments showed that the most efficient Zn/Cr ratio for phosphate adsorption is 4:1. The optimum pH for phosphate adsorption by the Zn/Cr LDH is pH 3. After pH 3, the removal rate decreased as the pH increased. The adsorption isotherm indicated that the removal rate increases with an increased
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concentration of phosphate in the solution. However, this study did only include concentrations below 200 mg phosphate per litre and further studies on the behaviour at higher concentrations are required.

The analysis performed by the X-ray diffractometer showed that the prepared adsorbent had a similar diffraction pattern as the standard spectrum for a Zn/Cr LDH. Additional analyses are necessary to determine the spectrum for degrees below 10.

Even though further experiments are required to find the optimal conditions for phosphate removal by the LDHs, the future looks bright. Additional research also has to be made regarding the recovery from the plating wastewaters of heavy metals necessary for the synthesis of the LDHs.

5 Acknowledgements

The author would like to thank Dr. Tong Ouyang and Changjun Peng at Xiamen University. This project has been financially supported by Thyréns, Xylem and Sveriges Ingenjörer.

6 References


7 Appendix

7.1 Standard curve
Table 6: The standard curve data.

<table>
<thead>
<tr>
<th>concentration mg/L</th>
<th>0.00</th>
<th>0.04</th>
<th>0.08</th>
<th>0.24</th>
<th>0.4</th>
<th>0.8</th>
<th>1.2</th>
</tr>
</thead>
<tbody>
<tr>
<td>absorbance</td>
<td>0.000</td>
<td>0.020</td>
<td>0.038</td>
<td>0.104</td>
<td>0.164</td>
<td>0.321</td>
<td>0.486</td>
</tr>
</tbody>
</table>

7.2 Effect of phosphate removal with different ratio LDHs experiment
Table 7: The absorbance measured for different Zn/Cr ratios.

<table>
<thead>
<tr>
<th>ratio</th>
<th>absorbance 1</th>
<th>absorbance 2</th>
<th>average</th>
<th>Concentration of P in the residuals</th>
<th>Removal rate (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1:1</td>
<td>0.289</td>
<td>0.32</td>
<td>0.3045</td>
<td>18.7594</td>
<td>62,48</td>
</tr>
<tr>
<td>2:1</td>
<td>0.174</td>
<td>0.147</td>
<td>0.1605</td>
<td>9.7728</td>
<td>80,45</td>
</tr>
<tr>
<td>3:1</td>
<td>0.115</td>
<td>0.232</td>
<td>0.1735</td>
<td>10.5841</td>
<td>78,83</td>
</tr>
<tr>
<td>4:1</td>
<td>0.099</td>
<td>0.091</td>
<td>0.095</td>
<td>5.6852</td>
<td>88,63</td>
</tr>
</tbody>
</table>

7.3 Effect of phosphate removal at different pH solution experiment
Table 8: The variations in absorbance, adsorption capacity, removal rate and the pH values after the adsorption due to different pH values.

<table>
<thead>
<tr>
<th>pH (pH value after adsorption)</th>
<th>Absorbance mean</th>
<th>Absorbance mean</th>
<th>Adsorption capacity (mg/g)</th>
<th>Removal rate (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
<td>0,285</td>
<td>0,315</td>
<td>31,5215</td>
<td>63,0429</td>
</tr>
<tr>
<td>3</td>
<td>0,002</td>
<td>0,006</td>
<td>49,9938</td>
<td>99,9875</td>
</tr>
<tr>
<td>4</td>
<td>0,087</td>
<td>0,082</td>
<td>44,9700</td>
<td>89,9401</td>
</tr>
<tr>
<td>5</td>
<td>0,112</td>
<td>0,106</td>
<td>43,4411</td>
<td>86,8822</td>
</tr>
<tr>
<td>6</td>
<td>0,144</td>
<td>0,14</td>
<td>41,3817</td>
<td>82,7634</td>
</tr>
<tr>
<td>7</td>
<td>0,304</td>
<td>0,283</td>
<td>31,9271</td>
<td>63,8542</td>
</tr>
<tr>
<td>8</td>
<td>0,369</td>
<td>0,37</td>
<td>27,1842</td>
<td>54,3684</td>
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<tr>
<td>9</td>
<td>0,44</td>
<td>0,43</td>
<td>23,0966</td>
<td>46,1932</td>
</tr>
<tr>
<td>10</td>
<td>0,414</td>
<td>0,415</td>
<td>24,3759</td>
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<td>11</td>
<td>0,479</td>
<td>0,458</td>
<td>21,0060</td>
<td>42,0120</td>
</tr>
<tr>
<td>12</td>
<td>0,515</td>
<td>0,58</td>
<td>16,0759</td>
<td>32,1518</td>
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<tr>
<td>13</td>
<td>0,59</td>
<td>0,595</td>
<td>13,2676</td>
<td>26,5352</td>
</tr>
</tbody>
</table>
## 7.4 Adsorption isotherm experiment

Table 9: The data from the adsorption isotherm experiment.

<table>
<thead>
<tr>
<th>Phosphate concentration (mg/l)</th>
<th>absorbance 1</th>
<th>absorbance 2</th>
<th>Mean</th>
<th>Dilution factor</th>
<th>Phosphate residuals (mg/l)</th>
<th>Adsorption capacity (mg/g)</th>
<th>Removal rate (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>25</td>
<td>0.103</td>
<td>0.049</td>
<td>0.076</td>
<td>0</td>
<td>0.1800</td>
<td>24.8200</td>
<td>99.2801</td>
</tr>
<tr>
<td>50</td>
<td>0.255</td>
<td>——</td>
<td>0.255</td>
<td>0</td>
<td>0.6268</td>
<td>49.3732</td>
<td>98.7464</td>
</tr>
<tr>
<td>75</td>
<td>0.1</td>
<td>0.118</td>
<td>0.109</td>
<td>25</td>
<td>6.5589</td>
<td>68.4411</td>
<td>91.2548</td>
</tr>
<tr>
<td>100</td>
<td>0.226</td>
<td>0.235</td>
<td>0.230</td>
<td>25</td>
<td>14.1413</td>
<td>85.8587</td>
<td>85.8587</td>
</tr>
<tr>
<td>125</td>
<td>0.597</td>
<td>0.381</td>
<td>0.489</td>
<td>25</td>
<td>30.2733</td>
<td>94.7267</td>
<td>75.7813</td>
</tr>
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