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A Bootstrapped Neural Network Prediction Model Applied to a Dye Biodegradation Rate in Textile Industrial Process

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Abstract

This work proposes a bootstrapped neural network prediction model in order to estimate the biodegradation of a dye usually applied in textile industrial process. The bootstrapped neural network is used to generate designed data sets to improve the value of statistical information by estimating the underlying relationship between the input and output spaces, in an intrinsic experiment applied to the chemical process. *Peseudomonas oleovorans* was used in biodegradation of reactive Black 5. Results show a brief comparison between the information estimated by the proposed approach and the experimental data, with a remarkable coefficient of correlation between real and predicted values for the biodegradation rate, of above 0.99. Dye concentration and pH had not interference on biodegradation index values. A value above 90% of biodegradation of Blue Remazol is achieved at 1.000-1.841 mL/10 mL of microorganism concentration and 1.000-2.000 g/100 mL of glucose concentration, on the experimental condition studied.

Keywords: bootstrap; neural network; textile dye; biodegradation; simulation.

1 Introduction

The treatment of effluents on the textile industries has become a very important problem. There are over than 100,000 commercially available dyes with more than 7 x10^7 tons of dyestuff produced annually worldwide (Robinson *et al.* 2001; Akhtar *et al.* 2005). These dyes are widely used in a number of industries, such as textiles, food, cosmetics and paper printing, with the textile industry being the largest consumer of dyes (Pandey *et al.* 2007).

In Brazil, there are 5,000 textile industries distributed like this: 11% are large companies; 21% are small companies; and 68% are micro companies. The Brazilian textile sector occupies the 5th place in direct jobs and the 6th in turnover. The production of dye in Brazil reaches 26,500 per year (Ulson de Souza *et al.* 2007; Silveira *et al.* 2009b). Moreover, it is estimated that in the coloring process, at least 20% of textile dyes are discharges into effluents, due to the losses during the process for fixing the colors to the fibers.

Removing these components from industrial waste is one of the major environmental problems faced by the textile sector, and the non-treatment of these effluents may cause serious risks to the environment and, consequently, to the whole productive chain. Therefore, the development of effluent treatment technologies has been a aim of the great interest in present time due to the increase environment conscience and severe law. Also, the companies, at implementing the new and efficient system of effluent treatment, show a proactive and compromised posture facing the environmental problems by measuring and eliminating its negative externalities, not causing impacts coming from its production process; at becoming sustainable there is no water consumption and discharge of polluting effluent.

Due to this growing uneasiness, many works have been done in order to find efficient approaches to simulate different process for degradation of azo dyes (Aleboyeh et al., 2008; Balan et al., 1999; Guimarães et al., 2008; Guimarães and Silva, 2007; Mohajerani et al., 2011; Zarei et al., 2010; Daneshvar, et al., 2006;

Soleymania et al., 2011; Ceylan, 2008). In this context, the use of artificial neural network models is widespread, presenting good results in the prediction of degradation rate under different conditions.

However, the use of neural networks is limited by the size of the experimental data set. Neural network experiments are conducted using a division of the available data into training, selection, and test sets (Panchal et al, 2011). However, unfortunately, the determination of constant values from experimental procedure has many limitations. In such cases, the initial collected data set is most of the time very small, the magnitude of variable effects is sometimes ambiguous and data gathering costs are high. So, the division of the data into multiple subsets is very unfortunate as we usually have less data than we would ideally desire even for a single subset.

An alternative to overcome these problems is to use a resampling approach as, for example, the bootstrap method which is based on an imitation of an probabilistic process using the information supplied by a given small set of random samples. Many works have shown the suitability of the bootstrapping technique for estimating objectives out of sample by redrawing small subsets (Efron, 1979; Efron and Gong, 1983), but the use of this approach in estimation of dye biodegradation rate is practically nonexistent (Prado et al., 2011).

Mainly in the area of business, the coupling between bootstrap and artificial neural networks has producing some improved prediction models (Franke and Neumann, 2000; Lajbcygier and Connor, 1997). In this approach, the bootstrap method is used to create several designed data sets, which are used to train different neural networks. This approach allows identifying the distribution of a statistical estimation to the construction of confidence intervals for the values that are been predicted (dye biodegradation rate in this case). Of course, decisions based upon a number of experiments with different subsets can produce results much more reliable and can at least mitigate the sampling bias, which is an important problem in the neural network approaches and it is an inevitable consequence of the limited number of samples that it is possible to record in a real experiment. (Franke and Neumann, 2000; Lajbcygier and Connor, 1997).

In this work, the bootstrap method has been applied in a neural network context in order to create improved dye biodegradation prediction model. *Peseudomonas oleovorans* was used in biodegradation of reactive Black 5 and a comparison between the predicted results of the designed model and experimental data was conducted to validate the proposed numerical approach.

2 Material and Methods

2.1 The Proposed Bootstrapped Neural Network prediction model

The bootstrap method is based on an imitation of the probabilistic process using the information from a given small set of random samples. Given an original (small) dataset, the method estimates the standard error of some parameter of interest using the samples as an approximation of our population. Specifically, it takes samples with replacement from the original dataset to approximate samples from the population. A simple algorithm to illustrate the bootstrap method can be defined as the next three steps (Johnson, 2002):

(1) Sample values with replacement from the original data and compute the parameter of interest p.

(2) Repeat step (1) a moderate to large number of times, B, to come up with "bootstrap" estimates p1, p2,...,pB.

(3) Use the standard deviation of the B estimates in step (2) to estimate the probability distributions describing the prediction process, the standard error, and the confidence interval.

Here, the original data set obtained from the experimental design (Table 1) was used to generate thirty bootstrapped data set for training feed-forward neural networks. The artificial neural network maybe defined as a set of mathematical methods and computational algorithms designed to simulate the

process of information handle and the knowledge acquisition on the human brain. Once inspired from the human brain biology, the neural network has some basic elements such as artificial neurons, synapses, neural weights, transfer functions, and so on (Haykin, 1999). The artificial neurons are grouped into layers and connected in parallel. The first layer of the neural network is called the input layer and it has a number of neurons equal the number of independent variable. Similarly, the last is the output layer and its neurons return the correspondent values of the dependent variables. Between the input and output layer, one or more hidden layers can be located, according to the problem.

In this paper, the bootstrap method has been used to generate thirty dataset to the neural network training. In this phase it was used neural networks with two hidden layers, 5 neurons in both layers, and bootstrapped training sets with 100 samples with replacement. This configuration of the neural networks was reached experimentally for values in the range 1-5, 2-15 and 20-100 for number of layers, number of neurons in hidden layers and bootstrap sample size, respectively. This neural network configuration is shown in Fig. 1.

The number of neural networks was chosen in order to ensure a statistical estimation precision of about 8% with 95% of confidence. Also, the tangent sigmoid was used as the transfer function, gradient descent with momentum back-propagation training with 0.2 of learning rate (Haykin, 1999).

After the training, the neural networks were used to generate new data sets that were used to improve the original set. This second step allows determining all the output values for dye biodegradation. The mean value of the output from the 30 bootstrapped neural networks was used as a predicted value of response biodegradation rate. The resulting prediction based upon a number of experiments with different subset can produce results much more reliable and can at least mitigate the sampling bias, which is an important problem in the neural network approaches and it is an inevitable consequence of the limited number of samples that it is possible to record in a real experiment. The whole prediction process is illustrated in Fig. 2.

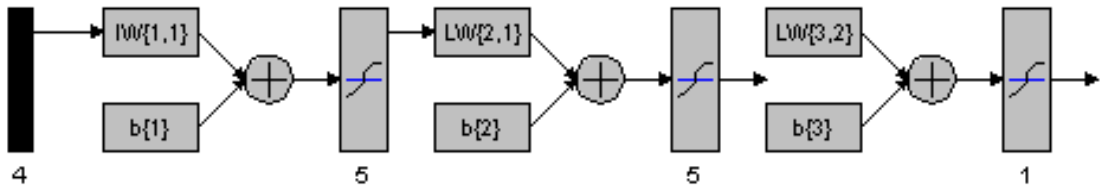

Figure 1: The neural network configuration

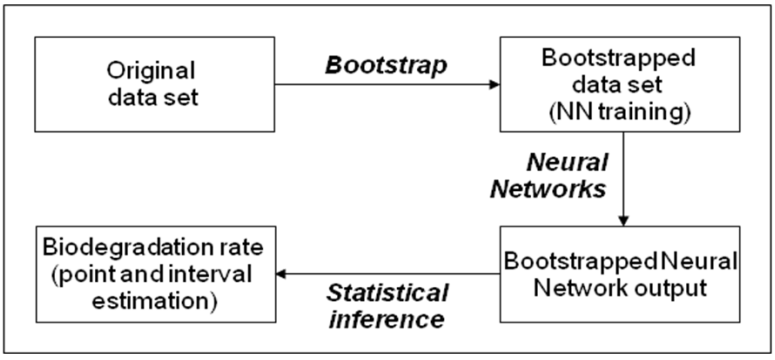

Figure 2: The proposed bootstrapped neural network approach

2.2 The Technique for Effluent Treatment

The treatment of effluents on the textile industries gets more complicated with the common use of several other chemicals with diverse composition, like, for instance: moisturizers, colorants, electrolytes, dispersers, pH controllers, stabilizers, among others, during the coloring process.

The main techniques showed in the literature for the effluent treatment are: adsorption, precipitation, biological and chemistry degradation, electrochemistry and photochemistry, and other. Currently, the most popular methods of color removal from wastewater involve physical and chemical processes that can be costly and usually include the formation of a concentrated sludge that creates a secondary, highly significant disposal problem (Chang and Kuo, 2000).

Although integrated chemical methods seem to be feasible for the treatment of such wastewater, biological methods should be used preferably considering cost and technical advantages (Yu *et al.,* 2010). Environmental biotechnology is constantly expanding efforts in the biological treatment of dye-contaminated wastewater. Although numerous microorganisms are capable of decolorizing dyes, only a few are able to mineralize these compounds into CO_2 and H_2O (Junghanns *et al.* 2008). Under aerobic conditions, azo dyes are not easily metabolized by bacteria (Robinson *et al.* 2001); however, under anaerobic conditions, several bacterial strains, including *Pseudomonas oleovorans*, can enzymatically reduce the azo bonds in the dye molecule to produce colorless by-products (Silveira *et al.* 2009b).

Due to their toxicity and low biodegradability of azo dyes, it is suggested to take advantage of more effective treatment methods such as advanced oxidation processes (AOPs) for destruction of these compounds in wastewater (Kim et al., 2004; Mohajerani et al., 2011). Several research on dye degradation have done, to improve the textile effluent quality (Aleboyeh et al., 2008; Guimarães et al., 2008; Guimarães and Silva, 2007; Mohajerani et al., 2011; Zarei et al., 2010). However, the main cost of Fenton reaction process with H_2O_2 is a disadvantage. Other problem, is that the higher addition of Fe^{2+} resulted in a brown turbidity that causes there combination of hydroxyl radicals and Fe^{2+} reacts with hydroxyl radicals as a scavenger and there is a need to remove this contaminant from the effluent, which raise the cost of the process (Kim et al., 2004).

2.3 Material

The microorganism was obtained from the Brazilian Collection of Industrial and Environmental Microorganisms, State University of Campinas, Brazil, previously identified as *Peseudomonas oleovorans* (CMAI 703). The Reactive Black 5 was provided by the Department of Chemical Engineering from Federal University of Sergipe. Glucose, potassium phosphate mono and di basic were provided by Merck (Darmstadt, Germany).

2.4 Design and Experimental System

An experimental design 2^{4-1} as shown in Table 1 was drawn up to study the effect of dye ($\mathbf{C_{dye}}$, mg/L), glucose ($\mathbf{C_{glucose}}$, g/100 mL), microorganism ($\mathbf{C_{mo}}$, mL/L) concentrations and pH (**pH**), in initial time, on dye biodegradation (%Biod). The experimental studies were carried out in a 250 mL conical flasks containing mineral medium and inoculated with *Peseudomonas oleovorans* at 180 rpm, temperature and pressure at home (27°C and 1 atm) in an orbital shaker, for seven days. After which time, samples were centrifuged and its dye biodegradation were estimated of 4 in 4 h, by a spectrophotometer at 600 nm (Silveira et al., 2009b). When a sample is digested, COD material in that sample is oxidized by the dichromate ion. The result is the change in chromium from the hexavalent (VI) to the trivalent (III) state. Both chromium species exhibit a color and absorb light in the visible region of the spectrum. In the 400 nm region the dichromate ion (Cr2072-) absorbs strongly while the chromic ion (Cr3+) absorbs much less. In the 600 nm region it is the chromic ion that absorbs strongly and the dichromate ion has nearly zero absorption. (Standard Method 5220 D.). The codification of the variables is:

$$x_1 = \frac{\left(C_{dye} - 0.1000\right)}{0.0500} \qquad (1)$$

$$x_2 = pH - 7.000 \tag{2}$$

$$x_3 = \frac{(C_{mo} - 1.000)}{0.500} \tag{3}$$

$$x_4 = \frac{(C_{Glucose} - 1.0000)}{0.500} \tag{4}$$

The biodegradation index was given for:

$$\%Bio = \frac{\left(C_{dye}(initial) - C_{dye}(end)\right)}{C_{dye}(Initial)} * 100 \tag{5}$$

3 Results and Discussions

3.1 Experimental Results

Table 1 shows the experimental planning and the experimental results for each assay from this work. Looking at the table, absolute majority of the trials showed a greater than 90% biodegradation of reactive Black 5. Apparently, there was a greater influence of the microorganism content than other factors. However, the glucose concentration cannot be excluded from the important factors, because under its lower concentration a low biodegradation index had been observed. From Table 1, the best result was found at 0.15 g/L of dye, pH 6, 1.5 mL/L of microorganism and 1 g/100 mL of glucose, while a biodegradation index of 96% was achieved.

Table 1: The experimental design to study of the dye biodegradation

Assay	**C_{dye} (g/L)**	**pH**	**C_{mo} (mL/L)**	**$C_{glucose}$ (%)**	**%Biod**
1	0.05	6	0.5	1	60.4628
2	0.05	6	0.5	2	72.9332
3	0.05	8	0.5	2	42.0354
4	0.05	8	0.5	1	78.3236
5	0.15	6	1.5	2	92.6132
6	0.15	6	1.5	1	96.6211
7	0.15	8	1.5	1	90.5308
8	0.15	8	1.5	2	92.5205
9	0.1	7	1	1.5	90.5028
10	0.1	7	1	1.5	90.4448
11	0.1	7	1	1.5	91.3872
12	0.1841	7	1	1.5	89.7986
13	0.0159	7	1	1.5	85.3622
14	0.1	8.682	1	1.5	90.1906
15	0.1	5.318	1	1.5	86.3221
16	0.1	7	1.841	1.5	93.0466
17	0.1	7	0.159	1.5	84.7183
18	0.1	7	1	2.341	66.8290
19	0.1	7	1	0.659	5.7394

C_{dye}, C_{mo} and $C_{glucose}$ are, respectively, the concentrations of dye, microorganism and glucose; %Biod is the biodegradation index

Golob et al. (2005) achieved a significant result for the coagulation and precipitation of the dye, reaching more than 90% removal of reactive Black 5, with $Al_2(SO_4)_3$ with a precipitant agent. However, after the process a high concentration of dissolved solid and chemicals can be observed; which prevents the use of this process because it increases the environment contamination, due to the decrease of the quality of the ecological parameters.

Aleboyeh et al (2008) showed that the initial concentration of the dye and initial pH have strong effects on the decolorization efficiency. And, none of the variables studied in your work could have been neglected in the photochemical decolorization of C.I. Acid Orange 7 solution, which in the best condition 90% has been decolorized. Similar efficiency was also observed by Zarey et al. (2010), in a study of degradation of C.I. Basic Red 46 (BR46) by photoelectro-Fenton (PEF) combined with photocatalytic process. Guimarães and Silva (2007) were also observed degradation efficiency more than 90% of the Acid Orange 52 and Acid Orange 10 and minor than 80% to the Acid Brown 75 and Direct Red 28 dyes.

Kim et al. (2004) had verified an efficiency of removal inferior than 60% was obtained by $FeCl_2$ precipitation reactive yellow 84 and blue 49 and an efficiency of 95% to photo-fenton process.

A lab scale active sludge reactor combined to a membrane separation process was used in the dye removal of denim textile wastewater. An efficiency of 75% and 90% were observed. However, in a biological treatment with yeast of *Pseudomonas pictorum* at 30°C, Balan et al. (1999) observed a degradation of 98% of the phenol for a high glucose concentration. The cell content has been not significant. It demonstrates the importance of the glucose on the *Pseudomanas* yeast in biodegradation process.

This demonstrates that the *P. oleovorans* are very efficient in the dye removal, and its results are similar than photo-fenton (Aleboyeh et al, 2008; Guimarães and Silva, 2007; Kim et al., 2004; Zarey et al., 2010) and *P. pictorum* treatments (Balan et al., 1999), higher than precipitation (Kim et al., 2004), active sludge and membrane processes (Sahinkaya et al. 2008). According to Balan et al. (1999) and Silveira et al. (2009a), the *Pseudomonas* yeasts, when are adapted to medium, at the end of nutrients they use the dyes as a substrate to survive. This occurs because they have the ability to release several enzymes (catalases) in the reaction, able to catalyze the decomposition of the carbonic and azo links.

And as photo-fenton processes have a high cost with chemicals and also increases the turbidity and the metal concentration in the effluent, it can consider that the biodegradation process using *P. oleovorans* is istill one of the most efficient and cheaper for the effluent treatment from textile industries (Kim et al., 2004).

On the best condition, the ecological parameters were measured and showed in Table 2. It was observed a higher percent of removal for all parameters.

Table 2. Measured parameter to the best experimental conditions

Parameter	Initial	End	% Removal
TOC (mg/L)	4047	2.055	99.95
COD (mg/L)	10150	28.22	99.72
BOD (mg/L)	6394	5.089	99.92

3.2 Bootstrapped Neural Network Prediction Results

For a common simulation by the traditional neural network method a large range of the assays is need. A full factorial design 2^4 has 27 assays (for a full experiment), and it is the maximum reduction in the assay quantity from a 3^4 common experiment, with 243 total assays (with triplicates), without losses statistically significant. A factorial design 2^{4-1} is a fraction from full factorial planning, resulting in 19 assays, as showed in Table 1. So, the original small data set obtained from the experimental design (Table 1) was used to generate thirty bootstrapped data set for training feed-forward neural networks.

After the training, the neural networks were used to evaluate the biodegradation rate from the original input values. This second step allows determining all the output values for dye biodegradation in order to generate new data sets to improve the original set. The mean value of the output from the 30 bootstrapped neural networks was used as a predicted value of response biodegradation rate. The results are numerically presented in Table 3 and illustrated in Fig. 3.

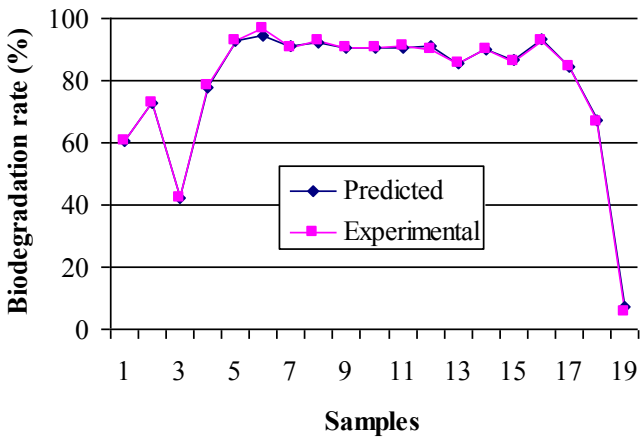

Figure 3: Predicted versus experimental dye biodegradation rate

Table 3: Original experimental data versus bootstrapping neural network prediction

Predicted biodegradation rate (%)	Experimental biodegradation rate (%)
60.3813	60.4628
72.7091	72.9332
41.9781	42.0354
77.9580	78.3236
92.6671	92.6133
94.4711	96.6211
91.3024	90.5308
92.0159	92.5205
90.6902	90.5028
90.6902	90.4448
90.6902	91.3872
90.8631	89.7986
85.5133	85.3622
90.2577	90.1906
86.7679	86.3221
93.5585	93.0466
84.1985	84.7183
66.9789	66.8290
7.3786	5.7394

The bootstrapped neural network approach also allows to calculate the confidence interval for all the predicted values. The 95% confidence intervals for this case are presented in Fig. 4. The prediction of the last sample was omitted for readability.

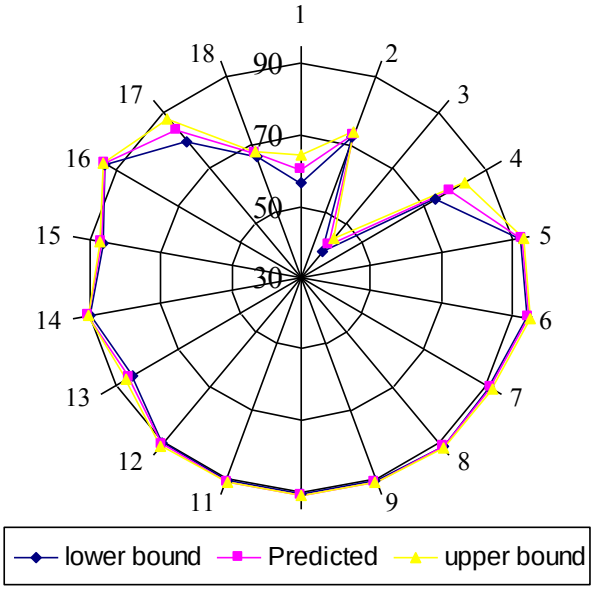

Figure 4: 95% confidence intervals for the predictions

Additionally, the results of the proposed approach were also compared with the experimental data. The performance of a linear regression between the network response and the corresponding target are presented in Fig. 5. The proposed model presented remarkable coefficient of correlation between real and predicted values for the biodegradation rate, above 0.99.

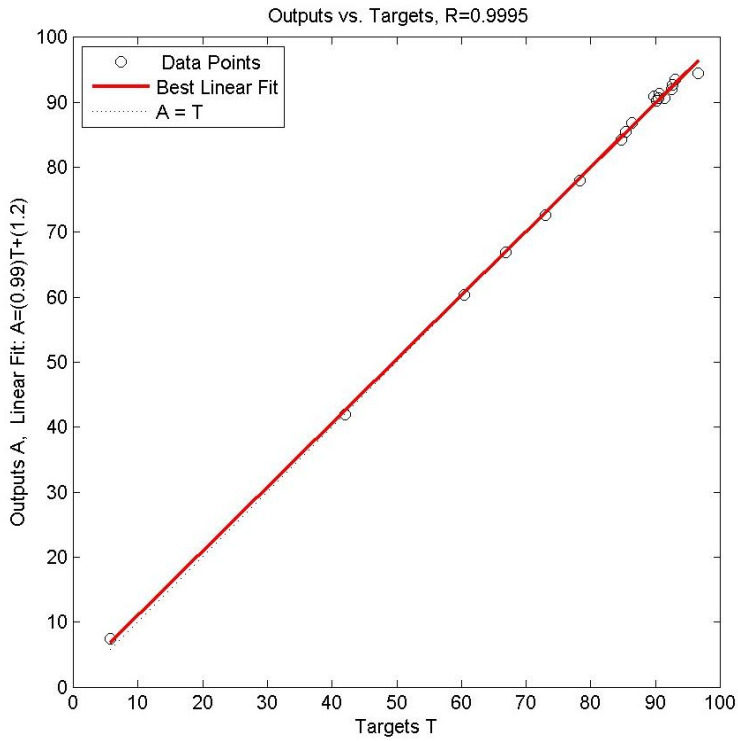

Figure 5: Linear regression results for the bootstrapped neural network approach

4 Conclusion

The bootstrapped neural network prediction model presented a good fit to experimental data. The model shows a remarkable coefficient of correlation between real and predicted values for the biodegradation rate, above 0.99.

The resulting prediction based upon a number of experiments with different dataset can produce results much more reliable, once it allows identifying the distribution of a statistical estimation, calculate the standard error and construct confidence intervals for the values that are being predicted. Moreover, this approach can at least mitigate the sampling bias, which is an important problem in the neural network approaches and it is an inevitable consequence of the limited number of samples that it is possible to record in a real experiment.

As the quality of empirical research depends in part on the quality and suitability of available data, making the search for new approaches to deriving better statistical estimators from constrained data sets is of continuing importance.

The proposed approach can be useful for problems in which the initial collected data set is very small, the magnitude of variable effects is ambiguous and the data gathering costs are high.

Future works will test the power of this technique in simulations results of others experiments.

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