Gurrea et al., J Pollut Eff Cont 2018, 6:3
DOI: 10.4172/2375-4397.1000227

Journal of Pollution Effects & Control

Research Article Open Access

T.E.R.R.A. (The Economical Response to Reduce Air Pollution): A New Device for Facilitating the Sequestration and Mineralization of (CO_2)

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Abstract

The atmospheric level of carbon dioxide (CO_2) has exceeded the 400 parts per million (ppm) threshold in 2015, making global warming a concerning cause for alarm. To lessen carbon emissions, Nickel Nanoparticles (NiNPs) have been proven to catalyze mineral carbonation in a Calcium Hydroxide ($Ca(OH)_2$) solution (Bhaduri Siller). This is seen as a viable method to reduce CO_2 in the atmosphere. However, this technology has not yet been applied in an actual device. T.E.R.R.A. (The Economical Response to Reduce Air Pollution) is a device which utilizes the catalytic effect of NiNPs and uses it to lessen the carbon emissions of vehicles. When tested in a laboratory setting, it was found that T.E.R.R.A. was able to sequester 82.5% of CO_2 bubbled through. With lower pressure and improvements in the design, it is projected that higher amounts of CO_2 may be sequestered by T.E.R.R.A., deeming it a feasible device for the mitigation of carbon emissions.

Keywords: Air pollution; Mineralization; Sequestration; Atmosphere; Environment; Climate change

Introduction

Carbon dioxide (CO_2) is a colorless and odorless gas that is composed of two oxygen atoms linked to a single carbon atom through a covalent bond. It is a greenhouse gas (GHG), or an atmospheric traces gas that allows solar radiation from the sun to pass through towards the Earth, but is partially opaque to the thermal radiation emitted by the Earth outwards. These proper-ties allow naturally occuring GHGs to regulate the temperature of the Earth. In fact, without these gaseous compounds, the average global temperature would be around 34°C lower than it is today [1,2]. However, human activity has been altering the carbon cycle through increased emissions, thereby altering the regulation of global temperatures.

According to the Intergovernmental Panel on Climate Change (IPCC), CO_2 has been the biggest contributor to climate change in terms of radioactive forcing RF, or the net increase or decrease in the thermal energy that reaches the Earth's surface. This is because; it is the most abundant GHG, constituting 72% of total emissions. In addition, CO_2 lasts longer in the atmosphere compared to other GHGs. When a pulse of CO_2 is emitted into the atmosphere, 40 percent will remain in the atmosphere for 100 years, 20% will remain for 1,000 years, and the nil 10% will remain for 10,000 years [3]. These reasons make CO_2 perhaps the most threatening GHG.

Due to its adverse effects on the environment, scientists have been looking to reduce the concentration of CO_2 in the atmosphere. Research facilities such as the Mauna Loa Observatory in Hawaii have been monitoring the long term activity of atmospheric CO_2 [4]. In September 2016, the Observatory showed that the atmospheric CO_2 level permanently surpassed the 400 parts per million (ppm) thresholds. This is the highest concentration of atmospheric CO_2 in the last million years (Scripps Institution of Oceanography, 2016). Additionally, the number is projected to reach 450 ppm by 2034, which is considered a high risk level [5].

An atmospheric CO_2 level below 450 ppm is needed in order to have a 50% chance to stabilize global temperatures at less than two degrees celsius above the pre-industrial period. The biggest human activity that emits CO_2 is the burning of fossil fuels for energy and transportation. When fossil fuels burn, carbon and hydrogen react with oxygen in the air to produce CO_2 and water. In particular, the burning of fossil fuels for transportation is the second largest CO_2 contributor. In the U.S., 32% of total CO_2 emissions in 2015 came from the transportation sector. Despite the fuel economy of modern day vehicles reaching a record low, an average passenger vehicle still emits about 4.7 metric tons of CO_2 per year [6]. Aside from this, reliance on vehicles for transportation is increasing as industrialization and modernization reach far areas.

The intake of CO_2 contributes to air pollution and promotes the spread of respiratory diseases. According to the World Health Organization, approximately seven million people worldwide die annually from air pollution, over six million of which were recorded in Asia in 2016. Aside from this, CO_2 contributes to global warming, the phenomenon that causes the gradual increase in the Earth's temperature. It is caused primarily by GHGs which ultimately cause the greenhouse effect: the trapping and redirecting of heat back into Earth.

Thus, carbon capture and storage (CCS) is considered a crucial strategy for meeting CO_2 emission reduction targets, such as that set by the UNFCCC. Present methods include catalytic converters, scrubbing, and cleaning of fuels before combustion. Another method of sequestration is through mineral carbonation, which involves bubbling the gas into an aqueous solution of divalent metal. The end product of this process is inert solid carbonates. Although mineral carbonation is a permanent and safe option for sequestration, many claim it is unfeasible because of its slow reaction rate.

In order to hasten the reaction involved in sequestration, a catalyst has been identified by scientists Baduri and Siller [1]. These are Nickel Nanoparticles, which range below 1,000 nanometers. The use of NiNPs

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Received April 05, 2018; **Accepted** July 17, 2018; **Published** July 23, 2018

Citation: Gurrea LK, Peregrino FR, Regalado J, Salvador EE (2018) T.E.R.R.A. (The Economical Response to Reduce Air Pollution): A New Device for Facilitating the Sequestration and Mineralization of (CO_2). J Pollut Eff Cont 6: 227. doi: 10.4172/2375-4397.1000227

as a catalyst in the rate limiting step of reversible hydration in the process mineral carbonation has been proven to hasten the sequestration of CO_2 by three folds. This study aims to design and evaluate a device using NiNPs which sequesters and mineralizes CO_2. Thus, a device is proposed which utilizes NiNPs in the mineral carbonation of CO_2. The proposed device shall be known as T.E.R.R.A. or The Economical Response to Reduce Air Pollution [7].

Methods

The project involved two phases

1. Phase 1: The Design Phase, and
2. Phase 2: The Construction and Evaluation Phase.

The first phase is the preparation phase which involves theoretically optimizing the device, and later constructing it. The second phase will be the evaluation phase which will evaluate the efficiency of the device through laboratory experimentation.

Materials and Equipment

All glassware's borrowed such as volumetric flasks, Erlenmeyer flask, graduated cylinders, beakers, and etc. are readily available at the Philippine Science High School Chemistry Stockroom. All reagents used for the reactions are of analytical grade. The synthesis of NiNPs necessitated nickel chloride hexahydrate ($NiCl_26H_2O$) as precursor material, sodium borohydride ($NaBH_4$) as reducing agent and sodium citrate dihydrate ($C_6H_5NaO_72H_2O$) as capping agent. For the sequestration and mineralization process, calcium hydroxide ($Ca(OH)_2$), H_2SO_4, and nitric acid (HNO_3) was utilized [8]. All these chemicals are also readily available at the Philippine Science High School Chemistry Stockroom except for the $NiCl_26H_2O$ and H_2SO_4 which was purchased from Patagonian Enterprises in Jaro, Iloilo City. Materials which include distilled water, hoses, hose clamps, spare parts, and CO_2 tanks were bought from local stores. Equipment such as analytical balance, pH meter, hot plate, magnetic stirrer, filter paper, iron stand and ring, cork borer, oven and centrifuge, were readily available at the PSHS Laboratory Stockroom. The Transmission Electron Microscope (TEM) was also available at the South-east Asian Fisheries Development Center (SEAFDEC), Tigbauan, Iloilo.

Device design

A design was conceived through the aid of a professional mechanical engineer, Edgar Allan Vargas. The engineer was able to contribute mostly to the practicality of the device design rather than the chemical design behind the device. The extent of his contribution was mostly centered with regards to its feasibility to manufacture, its practicality, and changes in dimensions to facilitate the welding of stainless steel tube. Further-more, he also gave valuable information in, the drafting of the device design, the purchasing of materials and the construction of the device. Numerous revisions in the design have been submitted for review and for further more changes. These are all listed and explained in the results and discussion section [9].

This design involves the utilization of 3 bubbling chambers. As CO_2 is bubbled through the solution, it reacts in the first chamber with $Ca(OH)_2$ to initiate the carbon mineralization process and is then converted in $CaCO_3$. However, since not all CO_2 is expected to react, additional chambers have been added to allow the escaping CO_2 to be sequestered and react again with the $Ca(OH)_2$ filled chambers. To further understand this process, an air flow diagram visualizing the movement of CO_2 can be seen.

A rectangular chamber was chosen for the sake of fabrication. Stainless steel is a relatively hard metal to bend and weld. Also, allowing for other geometrical shapes would have also meant an irregularly shaped device and would have made it more bulkier. Rectangular shapes decreases the total space the device occupies. Hence, for our device a rectangular chamber was chosen. To empty the $Ca(OH)_2$ solution in the device, a removable drain has been placed under the bottom of the device. The design also includes input and output tubes to facilitate the flow of air. Some padding was placed between the roof of the device and the chamber so that it may be sealed airtight. The roof of the device is removable with nuts and bolts holding it together [10].

Construction of device

The device was constructed through local stainless steel fabricators. Negotiations with M.Estomo Fabrication Stainless Works was set with the total cost for fabrication (labor and materials) is P8,000. The final image of the device can be seen below.

Synthesis of nanoparticles

Nickel nanoparticles were synthesized simultaneously during the construction of the device. This step is in preparation for experimentally evaluating the device. The chemical reduction method will be applied in the synthesis of NiNPs. First, 7.132 g of nickel chloride hexahydrate ($NiCl_26H_2O$) was dissolved in 30 mL distilled water in a 250 mL beaker. After which, 16 g of sodium citrate dihydrate ($C_6H_5NaO_72H_2O$) was added to the solution to act as a capping agent. The prepared solution was then placed in a water bath where it was heated (at 40°C) and magnetically stirred (at 400 rpm) for one hour [11].

During the one hour stirring time, a separate solution of 2.27 g sodium borohydride ($NaBH_4$) was prepared to act as a reducing agent. This concentration corresponds to 2: 1 molar ratio of $NaBH_4$ to $NiCl_26H_2O$ (Nayak et al.). The $NaBH_4$ solution was kept for temporary storage in a sealed 50 mL volumetric flask at room temperature while waiting for the $C_6H_5NaO_7$ and $NiCl_26H_2O$.

After an hour of stirring, the $NaBH_2$ solution was then added drop wise to the $NiCl_26H_2O$ and sodium citrate solution for 10 minutes while continuously stirring at 400 rpm (at 180˚C). The solution turned black, which indicated the reduction of the nickel ions. After the addition of $NaBH_4$, the temperature of the hot plate was decreased to 80˚C and left constant for two hours to allow the reaction to complete.

The resulting solution was filtered using filter paper and alternately washed thrice with distilled water and ethanol. To prevent excess moisture that would contaminate the nickel nanoparticles, it was dried using a hot air oven at 80˚Celsius for one hour. The weight of the produced nickel nanoparticles was recorded. Characterization techniques will not be done for the synthesis of the NiNPs since this method has already been confirmed to work from the researchers previous studies [12].

Production of CO_2

To evaluate the device, CO_2 was necessary for the reaction to take place and hence the purchasing of a CO_2 tank. A makeshift regulator was made by using adapters, tees, hose, and hose clamps. A brass adapter for the CO_2 tank was fitted. This was then connected to a stain stainless steel tee. The other 2 junctions of the tee were connected to a pressure gauge and a customized hose nozzle. The customized hose nozzle was made by welding to stain adapter and in to in hose nozzle. Thus, the resulting customized hose nozzle had in socket and in hose. A stain hose was then connected in the hose nozzle and was fastened

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by the hose clamp. Tee on was also wrapped between the adapters to ensure the no gas would leak out.

Using the setup shown below, a water displacement method was done to bubble in 20 L or 39.29 g of CO_2. 20 L of water was sealed in a large plastic bag. The water level plastic bag was then marked, signifying that this was 20 L of water bubbled in. Then, a similar volume of CO_2 corresponding to the marked 20 L was pumped for a period of time of 14.56 seconds. Hence, opening the valve for 14.56 seconds translates to 20 L of CO_2. A plastic bag was chosen so that their will not be any stretch marks which may compromise the volume of the container.

Evaluation of device

For added information, the fluid dynamics of the bubbling CO_2 may also be calculated based on the data (calculations shown in appendix). Utilizing, 20 L of CO_2 over 14.56 s, this translates to 1.37 L/s of CO_2 being bubbled into the solution. Using the Bernoulli equation one can also derive its velocity which is equal to 4.84 m/s and its pressure which is 23.24 Pa or 0.003 PSI. This means that only as much as 0.003 PSI back pressure from cars is necessary to facilitate the mineral carbonation process. This shows that 1.37 L/s of gas is very doable in practical as well as experimental situations since it generates only minimal back pressure [13].

Assuming, a 1: 1 ratio (i.e. a 100% efficiency) approximately 39.32 g of CO_2 will necessitate 68.87 g of $Ca(OH)_2$.$Ca(OH)_2$ was set as the excess reactant and spread evenly amongst the 3 chambers and 24 g of CO_2 was added per chamber for a total of 72 g. Furthermore, based on previous studies, an optimal concentration of 30 ppm or 0.09 g of NINP was also added into the solution, evenly spread amongst the 3 chambers.

As a result of reaction of $Ca(OH)_2$ and CO_2, $CaCO_3$ was expected to precipitate at the bottom of the chambers. The contents of each chamber were then drained, filtered and washed, oven-dried, and weighed. The precipitates were washed with distilled water while being filtered to ensure the purity of the $CaCO_3$. This was then oven-dried at 60°C overnight. These were done to determine the amount of $CaCO_3$ precipitate present in the solution.

Results

NiNPs characterization

To determine the the size distribution of the synthesized NiNPs, a Transmission Electron Microscope (TEM) from the Southeast Asian Fisheries Development Center (SEAFDEC) was utilized. The majority of the NiNPs were seen to be well below 50 nanometers (Figure 1) at 200,000X magnification. Using, 600,000X magnification, select samples of the NiNP reach lengths of below 10 nanometers (Figure 2).

Various designs of prototype

The first objective of this project was to come up with a feasible design for the fabrication which utilizes NiNPs. Numerous designs have been conceptualized and all of which have one thing in common, which is the utilization of water and $Ca(OH)_2$ for mineralization of the bubbled CO_2. The very first few designs include the use of a cyclone separator attached to a device with one bubbling chamber (Figure 3). This was to remove the particulate matter in the effluent gas using centrifugal force before proceeding to the bubbling process.

In the preliminaries, a prototype of this design was constructed using 1.5 L PET bottles and tubings and gas containing particulate matter was blown into the device using a vacuum. Through ocular observation the

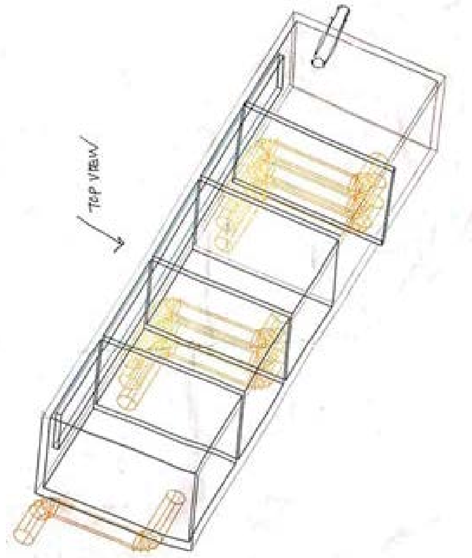

Figure 1: Interior of the proposed design of T.E.R.R.A.

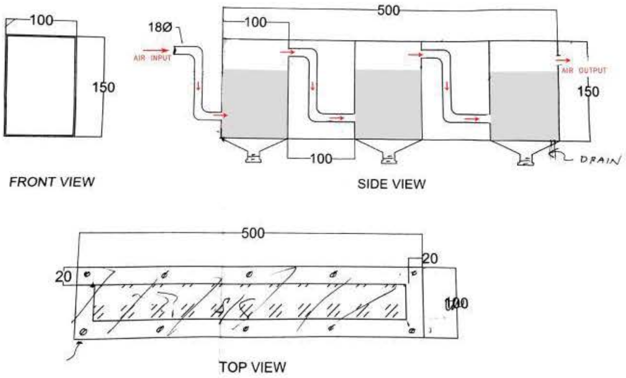

Figure 2: Interior of the proposed design of T.E.R.R.A.

Figure 3: Interior of the proposed design of T.E.R.R.A.

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device was able to remove the particulate matter from the gas. For the mineralization phase, a large surface area of the bubbling area would be ideal in a small and compact container for it to be a feasible device to be attached on vehicles. However, upon consideration, it was deemed that the design was bulky and due to the addition of a cyclone separator in that design. Hence, this was removed from the design and the project was limited to dealing with CO_2 only. The removal of particulate matter was deemed to be out of the scope of this study and a newer one has been conceived to adjust for a more ergonomic device.

The second design included a smaller but more complex system. It was designed to be attached to the tailpipes of cars. It was also self-sufficient on its own. It basically involves 2 components: an aerator to draw in ambient air and bubble this through a solution of $Ca(OH)_2$ and NiNP. This is then released outside. It uses air stones to use air through the $Ca(OH)_2$ solution and have more surface area for the reaction leading to an increased reaction rate. However, another consideration in this design is the power source for the aerator.

This design was further evolved and instead of an aerator, a customized air pump was fitted to sequester and aerate more air. Hence, the development of the third designs (Figure 4). This was introduced together with a piston and wheel to push and pull air, one way valves, and a DC power supply (battery) powers the said pump.

Shown below is the overview of the third design together with an air flow diagram of how air was supposed to navigate through the device. More components such as the tailpipe clamp were also added in this newer design. However, upon the advice of the research adviser, re-search panelists, and the advice of an engineer. The design was too complex since the device was only an initial prototype. As much as possible, only the main component (bubbling vessel) should be functional since it was only an initial prototype. Another rationale as to why it had to be simplified was because a sequential component leads to an increased risk of dysfunctionality in the device. In this case, for example, as suggested by a research panelist, when the device tailpipe clamp is loose, or when the battery runs out, or when the air pump is jammed, the device would not work. Hence, the design was simplified and ultimately the most recent design was conceived. These involved using the pressure of the gas coming from the tailpipe itself to bubble through the device, thereby, eliminating the need for extra components but increasing back pressure in tailpipes (listed in methods; 23.24 Pa or 0.003 PSI). The design was further improved by adding two extra chambers for a total of three chambers so that the efficiency of sequestration may be more efficient. The rationale for this addition was because the gas which the rest chamber could not sequester would be passed on into the second chamber and then to the third.

There was also other design which was conceived however, they did not utilize NiNPs and were not included because these were not included in the scope of this study. The next objective of this study was to determine the amount of CO_2 sequestered by utilizing $CaCO_3$ as a measurement of success. The amount of CO_2 sequestered per chamber was taken from the recovered precipitate. The weight of precipitate on each chamber was measured individually. The total precipitate in each tank was then measured. 67.6 g of $CaCO_3$ on the rest tank, 84.66 g of $CaCO_3$ on the second tank, and 69.02 g of $CaCO_3$ on the third tank were recovered. Finally, a blank trial (without NiNPs) was also tested and it had a total of 66.76 g of $CaCO_3$ recovered.

Figure 4: Interior of the proposed design of T.E.R.R.A.

The amount of CO_2 sequestered was derived through dimensional analysis of the amount of $CaCO_3$ formed. Of the 39.32 g of CO_2 released, an average of 32.44 g of CO_2 was recovered, approximately, making the average efficiency of the device 82.5%. So far, there have been no researches utilizing mineral carbonation as its method of sequestering CO_2 hence; this cannot be compared to other developmental projects. Through the given data one may also calculate the individual efficiencies of the device. By starting with the $CaCO_3$ sequestered of each tank, one may also calculate the individual efficiencies of each tank.

Chamber 3 had a higher efficiency as compared to the other two. Although chamber 3 may have the high efficiency (65.5% as compared to the other two), the total efficiency is not the sum of individual efficiencies in chambers 1 to 3.

Interpretation of Results and Findings

Aside from $Ca(OH)_2$, $CaCO_3$ precipitate was also used as a measurement of efficiency, as its production indicates the mineralization of CO_2. Other measurements of the mineralization of CO_2 would be the formation of water, and the depletion of $Ca(OH)_2$. The formation of water was not utilized to measure the mineralization of CO_2 because it would have been more accurate to measure the formation of $CaCO_3$ instead. $Ca(OH)_2$ could not have also been measured since excess amount of $Ca(OH)_2$ was added into the solution. Ideally, when the ratios are 1: 1 $Ca(OH)_2$ could have been measured through the pH of the solution. However, this could still lead to imprecise calculations, because as long as there is still more than enough $Ca(OH)_2$ than its solubility (1 g/L), it will still have effectively the same pH before and after bubbling. This makes calculations imprecise with the use of the pH meter. Hence the only viable choice was the measurement of $CaCO_3$

$$Ca(OH)_2 + CO_2 \rightarrow H_2O + CaCO_3 \quad [1]$$

The product efficiency of the device in sequestering CO_2 was 82.5%. There are several explanations as to how the sequestration did not reach 100 percent ideal. One possibility is a physical law in the construction of the device. The device may possibly not have a completely airtight container and CO_2 may have escaped through the gaps. Another reason may be caused by the speed of the CO_2 gas that was bubbled (1.37 L/s). This air-flow is might have been too fast and gave the CO_2 lesser time to dissolve in the solution. Remembering the basics of chemistry, the surface area of 2 reactants is a key factor and is directly proportional to the reaction speed of both reactants. In conjunction with this, a faster bubbling of CO_2 translates to more CO_2 aerated in a short period of time and hence, the consolidation of the bubbles in the solution, forming one big bubble instead and decrease surface area: volume: ratio. This gives lesser surface area for the CO_2 to react and mineralize. Instead, when CO_2, is aerated slowly, little bubbles are made and surface area:volume:ratio of these miniscule but numerous bubbles are definitely greater than the big bubble. This theory is also supported by the data when comparing the individual efficiencies of the chamber. As it can be seen, there is an increasing efficiency from chamber 1 to

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chamber 3. Since all other variables (pressure, temperature, size of chamber, weight of $Ca(OH)_2$, amount of NiNPs and etc) were kept constant, only the amount of input CO_2 per chamber changed as this entered the solution. This implies that it is the amount of CO_2 over a period of time which could have been responsible for the increasing efficiencies of the chambers. A slower aeration of CO_2 translates to a more efficient mineralization process.

Conclusion

A device design was conceived and constructed. It involved the use of $Ca(OH)_2$ to react with CO_2. This process was halted by the addition of NiNPs. A total of 73.76 g of $CaCO_3$ was formed during the course of the experiment as a result of the reaction between CO_2 and $Ca(OH)_2$. This converts to a total of 32.44 g of CO_2 mineralized out of the 39.32 g or 20 L of CO_2 bubbled. The entire device efficiency was calculated to be 82.5 percent efficiency, with the three individual chambers having 27.39%, 38.20%, and 65.5% efficiency. This study reports the successful development and the construction of a prototype of T.E.R.R.A and the mineralization of CO_2 using Nickel nanoparticles as catalyst. The efficiency of the device was calculated to be 82.5 percent based on the amount of CO_2 sequestered from $CaCO_3$.

Recommendations

This project is only the initiation phase of the development of T.E.R.R.A and will serve as an assessment of its potential as a viable commercial product. It recommended to experimentally determining the amount if CO_2 sequestered at varying CO_2 pressures to simulate the exhaust pressures of vehicles. Through this, a trend can be observed and a mathematical model can be generated from the relationship of the two variables. For improvements on the device, the effect of other variables such as type of gas (NOx and SOx), exhaust temperature and back pressure should be taken into account. Future additions to the feature may also include a particulate matter filter powered by a sustainable power source generated from the vibrations of the vehicle and $CaCO_3$ and NiNPs retrieval apparatus.

Acknowledgements

The completion of this research project was due in large part to the help and support of our research adviser, Mam Erika Eunice Salvador, who guided us every step of the way. We thank Sir Rusty Balcoba, Sir Michael Padernal and Mam Leilani Estilo for attending to our needs and providing us with access to the facilities and apparatus needed for us to complete this research project. Our heartfelt appreciation also goes out to Engr. Edgar Al-lan Vargas whose invaluable advice provided us with the foresight to conduct our experiments. Maam Shena Faith M. Ganela, Ph.D. for providing the necessary work areas in the conduct of our research. We extend our gratitude as well towards the Department of Science and Technology for aiding us financially. We thank our parents who have been supporting us throughout this entire process. We finally thank the Lord almighty, who provides us with all things. This research project was done for his greater glory.

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