

Investigation of latest techniques in carbon sequestration with emphasis on geological sequestration and its effects

Abstract

In the pursuit of development, man has polluted and exploited many resources provided by mother nature. In these pollutions, CO₂ pollution has become the most concerning contemporary and sought after problem in the current scenario. Observations indicate that the Carbon concentration levels have exceeded beyond the threshold limits. Among the solutions available currently, Carbon-Capture and Storage (CCS) or Carbon Sequestration (CS) is the best solution considering the cost and efficiency of carbon removal from the atmosphere. In the available Carbon sequestration methods, a geological sequestration is a viable option for long-term sustainable storage of CO₂. This article focusses on latest technologies developed with respect to Geological Sequestration and also on the carbon capture techniques, site selection for Geological sequestration, transport as well as uncertainties and difficulties in the modeling of the involved process. The main objective is to stress the need for these techniques and motivate fellow researchers in this essential and emerging field.

Keywords: greenhouse gas, carbon capture and storage (CCS), geological storage, carbon Sequestration, CO₂ mitigation

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Boindala Sriman Pankaj,¹ Jagadeesh Anmala²

¹Graduate Student, Birla Institute of Technology & Science, India

²Associate Professor, Department of Civil Engineering, Birla Institute of Technology & Science, India

Correspondence: Prof. Jagadeesh Anmala, Associate Professor, Department of Civil Engineering, Birla Institute of Technology & Science, Pilani, Hyderabad Campus, India, Email jagadeesh@hyderabad.bits-pilani.ac.in

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Introduction

Though there has been an increase in the level of Greenhouse Gasses (GHG) from the advent of industrialization, which occurred in the late 17th century, there has been a rapid growth of the release of these gases from the early 20th century. These changes have adversely affected the global climatic conditions. Various Global Climatic models (GCM's) predict that the increase in the concentration of these gases will be much more in the near future because more and more countries are getting developed/industrialized. These emissions are mainly due to the consumption of fossil fuels for energy. Though there are many alternatives suggested for energy production, scientists say that fossil fuels will be the main source for the foreseeable future (Park et al. 2004). The main component of these GHGs, which contribute to the increase in the global climatic temperature, is Carbon-dioxide. Carbon sequestration is one of the major methods to mitigate the effects of fossil fuel emissions.

What is carbon sequestration?

Carbon sequestration is a process in which the atmospheric carbon-di-oxide, carbon monoxide, and other forms of carbon, which are linked to global warming and air pollution are captured and stored in carbon sinks for long-term storage. This occurs naturally through many physical, biological and chemical processes. But these natural processes take a huge amount of time in their cycles and cannot compete with the current carbon generation. Therefore, new techniques are developed which artificially capture the carbon-related gases released by industries and store in various carbon sinks like reservoirs, saline aquifers, aging oil fields, oceans, etc.

Physical processes

Biomass sequestration

This is nothing but reforestation. The trees and plants take carbon-

dioxide from the atmosphere and convert them into biomass as they grow. So, the carbon can be stored as biomass in trees/plants until they decompose or are burnt. The problem with this is that the carbon is stored only for a limited amount of time, roughly say for about 80 to 90 years, and therefore, this cannot be a viable solution for the capture of all the carbon which we release into the atmosphere.

For solving this problem, scientists have come up with various technologies to prevent the carbon escape like bioenergy with carbon capture and storage (BECCS), biochar burial, and biomass burial. Bioenergy with carbon capture is a method in which the produced biomass by the plants is sent to industries to be used as a biofuel to generate bioenergy. The carbon dioxide released with this method is then captured and is stored into the geological formation where the carbon is stored for more than 10000 years with very less probability of escape.¹ Biochar is nothing but a type of charcoal used as a soil amendment/soil conditioner prepared by thermal decomposition of the biomass without oxygen. Based on the temperature, the rate of heating and other physical conditions the amount of biomass converted to biochar can vary from 20% to 50%. Other by-products can be used as biofuel.

Ocean storage

The pressures at the ocean bottom are large to contain CO₂ in its liquid phase.² At this location, it is easier to get stable and stationary pools of carbon-dioxide. The potential capacity of an ocean is very large and can hold more than a thousand billion tons of carbon dioxide. Ocean storage is not encouraged due to the impact of carbon dioxide on ocean life and its stability primarily.

Sub-terrain storage

Sub-terrain storage/geological sequestration means to store carbon-di-oxide in depths of the earth like in depleted gas reserves and other fossil fuel reservoirs, un-minable coal mines, etc. Once

the CO₂ is captured it is then subjected to high pressure until the gas enters a supercritical state. This liquefied gas is then pumped to the depths of the earth through pipes. It is believed that the pressure at those depths is sufficient to store the carbon-di-oxide for thousands of years in the liquid state.³

Chemical processes

Mineral carbonation

The mineral carbonation/mineral carbon sequestration is a method in which the captured carbon-dioxide reacts with mineral oxides like Magnesium Oxide and Calcium Oxide and form respective carbonates. This reaction occurs naturally and is also the reason for the weathering of rocks due to their exothermic nature. These can also happen with mineral silicates like magnesium and calcium silicates which produce Silicon-DI-Oxide along with their carbonates. This reaction is also exothermic but comparatively less in its intensity compared to the above reaction. These obtained carbonates can be stored in the earth as landfills. This method of sequestration is known to be permanent without any chances of leakage issues. The main problem with this method is the availability of raw materials and the extraction of these materials is of much concern.

Chemical scrubbers

Chemical scrubbers are natural or artificially made materials to capture CO₂ and separate them from the other gaseous materials. These scrubbers are initially used in rebreather in a diving suit. The researchers are now investigating their use in carbon capture and storage. There are various kinds of scrubbers available in the market which are:

- a. **Amine scrubbers:** Amine scrubbers have been known to man since 1930s and are used for carbon dioxide removal. When amines come in contact with acidic gasses like CO₂ they convert CO₂ to bicarbonate ion which can dissolve in the solution. The main advantage of these scrubbers is that they can release the carbon-di-oxide when the solution is heated which makes the amine solution reusable.
- b. **Zeolites and metal-organic frameworks (MOFs):** Zeolites or MOFs can be used to bind carbon dioxide reversibly. MOFs can be specifically manufactured to adsorb only carbon dioxide. The problem with this method is that they utilize many minerals whose mining may lead to other climatic problems.

Biological processes

The biological processes can be classified into two categories-terrestrial and aquatic. In these processes, the pace of natural carbon sequestration is increased by increasing the population of the photosynthetic organisms via artificial methods. The main problem with these is that the storage is not permanent and the problems caused by artificial indulging in the ecosystem may lead to other harmful consequences.

Carbon capture technologies

There are various stages in the carbon sequestration process out of which carbon capture is the most important and expensive stage. Almost 60 to 70% of the cost of CCS is utilized in carbon capture and the schematic diagrams of its processes could be found from Breeze.⁴ As of now most of the carbon capture techniques are available for stationary sources and very few techniques for remaining sources. These techniques are classified into various types as follows:

Separation with solvents

This process takes place in two stages, the first stage is the absorption of CO₂ by a solution and the second stage is the desorption by pressure or temperature variation or by an electrical impulse. In general, the exhaust gases from the industries or other point sources will be at high temperatures. So, before absorption, first we need to cool down this exhaust to a nominal temperature (normally around $\leq 40-60^{\circ}\text{C}$), then we allow the exhaust to pass through the absorbent chamber. In this stage, the carbon dioxide is absorbed and all the other gases which are inert to the solvent are released out. Then this solvent filled with CO₂ is sent to the desorption chamber where the CO₂ is separated through pressure or temperature variation or by an electrical impulse. After desorption, the solvent is again circulated for absorption and the carbon dioxide is then compressed for easy transport and storage (at pressures around 10–80MPa).

Separation by sorbents

This is similar to the above process, but instead of solvents, we use sorbents like activated carbon, zeolites, aluminophosphates etc. The carbon dioxide is adsorbed and not absorbed, reducing the amount of energy in desorption, but in decreasing the efficiency compared to that of absorption. Though there is a reduction in efficiency, this kind of separation process is also widely used.

Separation through selected membranes

Another method for separation of carbon is through selective membranes. The type of gases it can separate is based on the material of the membrane and the quantity of gas that can be separated is dependent on the pressure change across the membrane. The membranes can either separate the gases or can absorb the desired CO₂ gas.

Separation with other technologies

Cryogenic distillation

This process is a bit complicated compared to the other problems, but the yield of carbon dioxide, which we obtain, is high compared to the others mentioned above. In this process, the flue gas is subjected to a controlled, steady increase of pressure and a reduction in temperature is achieved to reach the triple point of the carbon dioxide (216.6K at 5.11 atm P) so that other gas will be in a gaseous phase where CO₂ will be in the triple state and can be easily separated out. The problem with this is that the flue gas should be cleared from H₂O, NO_x, SO_x and O₂ prior to the distillation process and this method is also costly and even energy intensive compared to other methods.

Ionic Liquids

Organic salts which are stable at room temperatures are called ionic liquids. Few of these salts are found to be highly affinitive towards carbon-di-oxide. With the discovery of 1-butyl-3-methyl-imidazolium hexafluorophosphate being able to dissolve carbon-di-oxide in huge amounts up to 0.72-mole fraction,⁵ the popularity of ionic liquids for carbon capture has increased. Another advantage of using ionic liquids is that though high amounts of carbon-di-oxide are being dissolved very least part of the liquid is in the CO₂ phase which makes it ideal for separation. These make them highly potential capture materials for carbon capture.

Enzyme-based system

Carbon capture through enzyme is another latest technique, which tries to mimic the natural process of capturing carbon. Carbonic

Anhydrase (CA) is an enzyme used to convert CO₂ to bicarbonate ion (HCO₃⁻). The hydration rates are as fast as 10⁴ to 10⁶ molecules of carbon-di-oxide per second for every molecule of CA (Nguyen and Codexis 2012; Yong et al. 2017). The rates can be controlled by controlling the pH of the medium. Though this is promising, the enzyme cost and loss of enzyme activity are reducing its popularity.

Geological sequestration

Geological sequestration is nothing but the subterranean storage explained in physical processes of the previous section.

Principles and mechanisms of geological carbon sequestration

Geological sinks for carbon storage

Carbon dioxide is a naturally generated gas, which is stable at normal atmospheric conditions. At 31°C and pressure more than 7.38MPa, carbon dioxide is found to be in the supercritical state. At such critical conditions, carbon-di-oxide fills the usable volume and has a 'liquid-like density' which changes depending on applied or surrounding pressure. Though the CO₂ is soluble in water in general, it is immiscible in water in the supercritical state. In such conditions of temperature and pressure, CO₂ forms a hydrated crystalline structure which is heavier than water. The CO₂ also has a strong affinity to coal, which is greater than methane gas normally found in coal mines. These properties of CO₂ help us in finding appropriate sinks for carbon storage.

Storage in salt caverns

Salt caverns have been used as a storage site since 1940's for many fossil fuels. Use of these storage sites can provide a long-term solution, but the associated cost for the storage is quite high and the disposal of the resulting brine solution is also an environmental hazard.

Oil displacement in reservoirs

The property of CO₂ to reduce the viscosity and capillary pressure of liquid fossil fuels and oils makes it a well-suited substance for enhanced oil recovery (EOR).³ EOR is a process in which the gas is pumped into the oil wells and the residual oil is then displaced due to the interaction of these gases with the oil. About 40% of the residual oil can be displaced with CO₂ in the process of EOR. This method provides us with both the uses-EOR and sequestration. The main problem to use this for sequestration is that the CO₂ storage is very less and the CO₂ obtained from the capture is highly un-economical compared to natural production of CO₂.⁶

CO₂ sequestration in coal mines and the displacement of methane

Carbon dioxide is injected into deep coal mines and mines which are uneconomical to mine CO₂ back. By doing this, the injected carbon dioxide displaces the methane gas present in the mines due to its affinity towards coal being greater than methane.⁷ Though methane is also a greenhouse gas, the use of methane over coal is much effective as methane is a better fuel than coal which reduces the CO₂ emissions. This can be an effective mid or long-term storage method.

Storage in depleted hydrocarbon reservoirs

Hydrocarbon reservoirs are better storage sites compared to the liquid fossil fuel reserves. The main advantage is the presence of existing trapping conditions, which will ensure the separation of

injected carbon dioxide from reaching the surface. This advantage over other sites makes the sequestration much easier and economical.

Storage in deep saline aquifers

Beneath the earth's crust, there are many water storage strata called aquifers. Some of these aquifers contain high concentrations of salts making it unusable for consumption or agricultural use. The solubility of carbon dioxide increases with increase in salinity of the water.⁸ Because of this property of carbon dioxide, saline aquifers can be potential storage sinks for CO₂.^{9,10} Due to the deep location of these aquifers the existing pressure present is sufficient enough to reduce the density of carbon dioxide closer to the water which prevents it from escaping out. Though there might be the drift of these waters beneath the earth, the velocity of flow of water is in the order of cm/year. Deep saline aquifers have been proved to be the most potential carbon sequestration sites in all aspects from volume to cost, with almost zero environmental impact.⁷

CO₂ sequestration in geological media and its criteria

There are different criteria in choosing a particular site for geological sequestration. These criteria can be classified into the following and should be considered in order of—geological, hydrodynamic and geothermal, hydrocarbon potential, and economic, political and societal criteria.⁸

Geological criteria: The sites which are technically stable and are not subjected to frequent quakes or volcanoes are considered to be the best sites.

Geothermal criteria: The researchers found out that the temperature gradient is 25°C/km and there will be an increase of 1Mpa pressure with the depth of 100m as we go below the ground.¹¹ This assumption is not always true for all the sites. We need to investigate other geological features like the presence of magma.

Hydrodynamic criteria: We also need to check the presence of water regimes in the storage sites before the injection of CO₂ because some coal sites might contain flowable groundwater which might leak the carbon dioxide.

Trapping mechanisms in geological sequestration

As mentioned above the carbon-di-oxide is mostly injected in a supercritical state because of its density which is more than in its normal state.³ Depending on the available reservoir conditions, CO₂ can be stored in supercritical, liquid or gaseous states. Mostly CO₂ is allowed to freely move laterally and vertically inside the reservoir.¹² Trapping of CO₂ inside the geological formation as a residual gas occurs when the formation water invades or comes in contact with the CO₂ plume.¹⁰ Four kinds of trapping mechanisms occur inside the reservoir but they do not occur at the same time. All these mechanisms are briefly explained below.

I. Hydrodynamic trapping

Structural or hydrodynamic trapping refers to the CO₂ which is trapped in a gaseous or supercritical state under a caprock with low permeability. When the CO₂ is injected into the formation, due to its less density compared to formation fluid it rises up towards the caprock due to buoyancy. Due to the high capillary pressure of caprock compared to the hydrodynamic force of CO₂, the injected CO₂ resides or is trapped inside the reservoir. This is one of the most important

trapping mechanisms as it is a preliminary trapping condition where it ensures that CO₂ is stored until other trapping mechanisms come into action.¹³

The efficiency of this kind of trapping mechanisms of a reservoir/formation can be evaluated by examining the sedimentary basins, the locations of strata having high and low permeability as those control the nature of the flow of fluids in that reservoir.¹² There are many kinds of structural traps which can be used as physical traps for carbon storage among those commonly we can see anticlinal folds and sealed fault blocks. The storage capacity of these reservoirs is determined by the volume of pore spaces. CO₂ can be injected until it reaches a point where the pressure of CO₂ is more than the capillary entry pressure.

II. Residual trapping

Residual or Capillary trapping can be observed mostly when we use saline aquifers or any other formation containing a fluid. When the Carbon dioxide is injected into the formation it displaces the existing fluid in the capillary pores, when the injection is stopped the fluid again tries to occupy the pore spaces. While doing this only the top layers of the pores are replaced trapping most of the Carbon dioxide in its bottom layers. Fluid previously existing in the reservoir acts as a seal in this kind of trapping mechanism.

III. Solubility trapping

Solubility trapping occurs after the above-mentioned trappings. It is a trapping mechanism wherein CO₂ dissolves in the formation liquid. In general, the formation liquid is a brine solution. When the CO₂ is injected into the formation it spreads completely over the surface of the formation liquid. As soon as the CO₂ touches the formation liquid, mass transfer takes place between the formation liquid and CO₂ until it reaches equilibrium. The rate of dissolution of CO₂ in water increases with increase in salinity. This process is very slow compared to the above trapping mechanisms as the molecular transfer rate coefficient is too small. It is assumed that to completely dissolve the CO₂ in the brine liquid in the formation, it takes about 1000 years.¹⁴⁻¹⁶

In the dissolution of CO₂, a process named DDC (dissolution-diffusion-convection) occurs. In this process, when the CO₂ dissolves in the top layer of the brine solution, it becomes denser compared to its original state. This makes the top layer to come down and the bottom layer to go up. This process continues to increase the formation's storage capacity.

IV. Mineral trapping

Mineral trapping implies the permanent containment of CO₂ in stable forms when reacted with the minerals and other organic matter present in the formations. This trapping mechanism takes a very long time to initiate compared to the above-mentioned mechanisms. After the dissolution of CO₂ into the formation liquid, the formation minerals undergo various chemical reactions. Some of these reactions are favorable to us due to the formation of carbonates which is a permanent storage form for CO₂.

Geological sequestration of CO₂ and its transport

A. Modelling transport of CO₂ for GCS (geological carbon sequestration)

The location of carbon capture and storage might not be always in the same location. Sometimes the location of both of these sites might

be far from each other. For this purpose, transportation of carbon or related gases also arises in CSS processes. In general, onshore transportation of CO₂ is done by using pipes or tankers while offshore transport is done by using ships and sometimes by pipes.

Pipe modeling of transport

In general, CO₂ can be transported in solid, liquid or gaseous state. But transporting CO₂ in the supercritical state (at a pressure of 7.36Mpa and temperature of 30°C) is most economical. As CO₂ in this phase has low viscosity and high density, very small diameter pipes can be used. Maintaining constant high pressure in a pipe is reasonably possible, but maintaining a constant temperature throughout is very difficult. The temperature of the material that is being transported depends on the surrounding temperature. It is possible that the surrounding temperature may vary to some extent. The pipes are usually buried underground, and in the soil, the temperature does not vary much. We need to take care of two-phase flow of CO₂ in pipes because that might damage the pumps or other equipment used while transporting through pipes.

Design of pipes systems

The pipe design for CO₂ is almost the same as the pipe design used for transportation of various other fossil fuels. One of the important conditions to be maintained is that the pressure should not drop below the critical pressure (7.36MPa) and the other conditions should be such that there is no two-phase transportation through the pipes. There are various components in the design of pipes for transport, which are as follows:

- Amount of CO₂ to be transported in the supercritical state determines the design of pipe
- Design of compressors, pumps and other machines used to maintain CO₂ in single-phase dense state
- Design of valves and other components for controlling, operating and monitoring the transport.¹⁷

Pipe design: The internal diameter (d) of the pipe is calculated assuming a uniform gas velocity throughout the pipe. A standard velocity equation is used to compute this parameter.

$$d = \sqrt{\frac{4Q}{\pi U}} \quad (1)$$

Where d is the internal diameter of the pipe, Q is the flow rate, U is the uniform velocity. The thickness of the pipe is determined by the maximum pressure the pipe should retain. For this calculation, we use Barlow's formula.¹⁷

$$t = \frac{Pd}{2(S - P)} \quad (2)$$

Where t is the thickness of the pipe, S is the Specific yield of the material of pipe, d is the internal diameter of the pipe, P is the maximum pressure to be retained by the pipe. As there will be head losses while transporting the CO₂, booster pumps are used to maintain the pressure above the critical pressure. The spacing of these is found out by using the Darcy-Weisbach equation. Here, we assume that all the head loss is due to friction and is given by

$$h_f = f \cdot \frac{L U^2}{d 2g} \quad (3)$$

Where f is the coefficient of friction, L is the length of pipe; h_{fs} is the head loss due to friction. By fixing the allowable head loss we can find the spacing of pumps.

B. Estimation of formation site's storage capacity

As mentioned above, though there are various sources where we can store CO_2 . Geological sequestration is found out to be best storage site with huge storage potential and with less risk of environmental contamination. According to Mo et al.¹⁰ the best sites for storage are:

- Exhausted Oil and Gas reserves
- Coal formations
- Saline/ brine formations

Among these saline aquifers have largest storage potential and are considered to be best storage sites. The estimation of storage capacities is given below.

Oil and Gas formations: In the above three storage sites, we know the characteristics of the oil reservoir more. We assume that the volume of CO_2 which can be stored in these sites is equal to or slightly less than the volume of the oil extracted. For this estimation we use the following equation:¹⁸

$$G_{CO_2} = Ah_n \varphi_e (1 - S_{wi}) B \rho_{CO_2std} E_{oil/gas} \quad (4)$$

G_{CO_2} is the storage resource mass estimate of carbon-di-oxide, A is net surface area, h_n is the net aquifer thickness, φ_e is the effective porosity, $(1 - S_{wi})$ original hydrocarbon saturation, B is the gas or oil volume factor, ρ_{CO_2std} is the standard density of carbon-di-oxide, $E_{oil/gas}$ is the coefficient of efficiency of storage of reservoir.

Deep saline/brine formations: According to Bachu¹⁹ saline aquifers have the greatest storage potential among all other known types of geological storage. Not all the saline aquifers are suitable for storage. Aquifers having sufficient pressure to maintain the CO_2 in either liquid or supercritical state are only suitable. Another condition is that the storage formation should have a low permeable caprock preventing CO_2 to escape until the solubility mechanism comes into the picture. The capacity of the aquifer satisfying all these conditions is calculated by using this equation:¹⁸

$$G_{CO_2} = Ah_n \varphi_{tot} \rho E_{saline} \quad (5)$$

G_{CO_2} is the storage resource mass estimate of carbon-di-oxide, A is the total gross area of formation, h_n net aquifer thickness, φ_{tot} is the total porosity, ρ -standard density of carbon-di-oxide, E_{saline} is the factor indicating the efficiency of storage.

Un-economical Coal mines: Un economical or un-minable coal mines are also potential storage sites. The selection criteria for these sites are that the permeability of coal bed should be greater than 1 md. The deeper you go the more permeability decreases. According to IPCC 2005, the depth limit for coal bed storage is around 1300-1400 m below the surface of the earth. The storage capacity of a coal bed can be calculated by equation (6) (Goodman et al. 2011)¹⁷

$$G_{CO_2} = Ah_n C_{s,max} \varphi_{tot} \rho_{CO_2std} E_{coal} \quad (6)$$

G_{CO_2} is the storage resource mass estimate of carbon-di-oxide, A is the area, h formation thickness, $C_{s,max}$ is the maximum absorption capacity of carbon-di-oxide by unit volume coal bed, φ_{tot} is the total porosity, ρ -standard density of carbon-di-oxide, E_{coal} is the efficiency of storage.

C. Modeling of sink performance

Modeling of sink performance is nothing but finding out the behavior of the carbon-dioxide after injection into the reservoir. As we don't know the exact conditions in the reservoir, we rely on numerical results. The behavior of CO_2 will change with time and spatial scale. We can either use a multidimensional model having various parameters and equations for finding out the state of CO_2 to give accurate values or we can use a simple model with many assumptions which are easy to compute and which gives an overall idea of CO_2 in the formation. Based on the first approach, complex 3-dimensional models are generated. In these 3D models, many processes which are expected to occur inside are included and parameters which determine capillary pressure, permeability, dispersion, diffusion, equations of state, the energy of transport and geochemical reactions are also included. Although these kinds of models are accurate, due to data unavailability (and too much effort in obtaining the required data) these models are not always used. However, suitable assumptions are made to simplify these 3-dimensional models to produce results with reasonable accuracy. Some of these simplified models which are widely used are TOUGH2, ECLIPSE, LLNL, STOMP, NUFT. Among those TOUGH2 is mostly used. It assumes Darcy's law to be applicable for all the phases in the system, no chemical reactions, and all phases to be in local chemical and thermal equilibrium.

Conclusion

From the above study, the conclusions can be drawn as follows:

- Artificial Carbon Sequestration should be focused as it is a promising technique to prevent accumulation of greenhouse gases such as CO_2
- The potential coal beds and oil reservoirs should be identified and should be used for carbon sequestration as the additional fuel extraction might motivate industries to contribute towards the carbon sequestration.
- Ocean sequestration has also great potential and should be investigated more thoroughly.
- Geological sequestration is proved to be safe and efficient, so industries should focus on using this method instead of releasing CO_2 into the atmosphere.
- Investigations on economical carbon capture techniques are a potential research area.

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Conflicts of interest

Author declares there is no conflicts of interest.

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