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Process modelling and multi-objective optimisation of solid sorbent-based direct air capture

Toluleke E. Akinola[a], Meihong Wang[a]*

[a] The University of Sheffield, School of Chemical, Materials and Biological Engineering, Sheffield S1 3JD, South Yorkshire, United Kingdom
* Corresponding Author: meihong.wang@sheffield.ac.uk

ABSTRACT

Direct Air Capture (DAC) is recognised as a critical climate mitigation technology necessary for achieving global net-zero emissions by balancing difficult-to-avoid emissions. Despite its importance, the commercial deployment of DAC technology is currently challenged by the substantial energy demands and resultant extremely high operational costs associated with handling the low atmospheric CO_2 concentration. This study aims to address these two challenges through dynamic process modelling, simulation and rigorous multi-objective optimisation of a solid sorbent-based temperature vacuum swing adsorption (S-TVSA) cycle. The system utilises an advanced amine-functionalized sorbent, selected for its favourable low-temperature regeneration kinetics.A technical performance assessment was conducted using a first-principle mathematical model to accurately simulate mass and heat transfer in the adsorbent beds. To improve system viability, a multi-objective optimisation with NSGA-II in MATLAB was performed, maximising CO_2 productivity and minimising energy demand. The optimisation targeted key parameters like adsorption/desorption time, temperature, and pressure. The Pareto frontier shows trade-offs, identifying optimal points that minimise W_{eq} and energy use. These results provide a pathway to reduce energy costs and enhance the economic viability of solid-sorbent DAC technology.

Keywords: Direct air capture, amine-functionalised sorbent, process modelling, performance evaluation, Process optimisation, temperature vacuum swing adsorption

1. INTRODUCTION

A significant driver of climate change is the rapidly rising atmospheric concentration of greenhouse gases including CO_2 [1]. Atmospheric CO_2 levels have increased from 280 ppm in the pre-industrial era to 425 ppm in 2025 [2]. As a result, global temperatures have risen, posing a threat to the environment. To limit the temperature rise to 1.5–2°C, achieving net-zero CO_2 emissions by 2050 is essential [3].

Solid-based direct air capture (DAC) technology plays a key role in meeting net-zero targets [4]. However, the diluted atmospheric CO_2 concentration poses challenges for meeting energy demand and for the cost of CO_2 extraction, as large volumes of air must be processed to extract a significant amount of CO_2 [1, 4, 5].

Previous studies [1, 5] demonstrated that a steam-assisted temperature-vacuum swing adsorption (S-TVSA) process can achieve high-purity CO_2 (≥ 95%) from ambient air. In S-TVSA, CO_2 is initially adsorbed onto an adsorbent under ambient conditions. Subsequently, the pressure is reduced, and steam is used to increase the temperature, releasing CO_2. The process alternates between adsorption and regeneration to capture CO_2. Designing these processes is complex due to the nonlinear mass and heat transfer within the adsorbent column [19]. Operational decisions, such as cycle step durations, temperatures, pressures, and flow rates, directly influence performance metrics. This study aims to optimise these operational parameters to maximise productivity, reduce energy consumption, and ensure high CO_2 purity. This study utilises 3-aminopropylmethyldiethoxysilane-functionalized nanofibrilated cellulose (APDES-NFC), an advanced amine- functionalized sorbent due to its (i) favourable low temperature kinetics and (ii) reliable performance in the presence of moisture [1, 5].

2. METHODOLOGY

2.1 Model development

2.1.1 Adsorption Equilibrium

The co-adsorption of CO_2 and H_2O on the APDES-NFC surface was modelled using a combination of the modified Toth isotherm for CO_2 and the Guggenheim-Anderson-de Boer (GAB) isotherm for H_2O. The modified Toth isotherm model is defined as:

$$q_{co_2}(T, p_{co_2}, q_{H_2O}) = n_s(T, q_{H_2O}) \frac{b(T, q_{H_2O}).\, p_{co_2}}{[1 + (b(T, q_{H_2O}).\, p_{co_2})^{t(T)}]^{1/t(T)}} \quad (1)$$

$$n_s(T, q_{H_2O}) = \left[n_{s_0} \exp\left[\chi \left(1 - \frac{T}{T_0} \right) \right] \right] . \left[\frac{1}{1 - \gamma_{q_{H_2O}}} \right] \quad (2)$$

$$b(T, q_{H_2O}) = \left[b_0 \exp\left[\frac{\Delta H_0}{RT_0} \left(\frac{T}{T_0} - 1 \right) \right] \right] . \left[1 + \beta_{q_{H_2O}} \right] \quad (3)$$

$$t(T) = t_0 + \alpha \left(1 - \frac{T_0}{T} \right) \quad (4)$$

Where q_{co_2} is specific adsorbed amount of CO_2 (mol/kg), n_s is the saturation capacity of CO_2 (mol/kg), p_{co_2} is CO_2 partial pressure (kPa), b_0 is the adsorption equilibrium constant (kPa). The GAB isotherm model is defined as:

$$q_{H_2O}(x) = c_m \frac{c_G K_{ads} x}{(1 - K_{ads} x)(1 + (c_G - 1) K_{ads} x)} \quad (5)$$

Table 1 GAB and Modified Toth isotherm fitted parameters [1, 5]

GAB isotherm	
c_m (mol/kg)	36.4800
c_G	0.1489
K_{ads}	0.5751
Modified Toth isotherm	
n_{s0} (mol/kg)	2.3800
b_0(kPa^{-1})	70.7000
t_0	0.4148
T_0 (K)	296.0000
ΔH_0 (kJ/mol)	-57.0470
α	-1.6060
χ	0.0000
γ (kg/mol)	0.0061
β (kg/mol)	28.907

2.1.2 Model development of fixed-bed adsorption

The key assumptions are as follows:

- 1-D model in axial direction
- Solid and gas phases are in thermal equilibrium
- Mass transfer rate described using the linear driving force (LDF) model
- Pressure drop was described using Ergun equation
- Heat and mass transfer coefficients were assumed to be constant
- Heat capacities and heat of adsorption were assumed to be constant
- Only CO_2 and H_2O are adsorbed by the solid adsorbent.

The mass and energy balance of the fixed-bed adsorption column is described as follows:

Mass Balance

$$\epsilon_t \frac{\partial c_i}{\partial t} + \frac{\partial}{\partial z}(v c_i) + (1 - \epsilon_b)\rho_p \frac{\partial q_i}{\partial t} = 0 \quad i = 1, \ldots, n \quad (6)$$

LDF

$$\frac{\partial q_i}{\partial t} = k_i (q_i^* - q_i) \;\; i = 1, \ldots, n \quad (7)$$

Energy balance

$$\left(\epsilon_t C_g + \rho_b C_s + \rho_b C_{ads}\right) \frac{\partial T}{\partial t} - \epsilon_t \frac{\partial P}{\partial t} + v C_g \frac{\partial T}{\partial z} + \rho_b \sum_{j=1}^{n} \Delta H_j \frac{\partial q_j}{\partial t} = -\frac{4 h_L}{d_{in}} (T - T_w) \quad (8)$$

Wall Energy Balance

$$\frac{\partial T_w}{\partial t} = \frac{2}{C_w (r_{out}^2 - r_{in}^2)} [h_L r_{in} (T - T_w) - h_w r_{out} (T_w - T_{amb})] \quad (9)$$

2.2 Process simulation of the TVSA cycle

Figure 1 shows the steam-assisted temperature-vacuum swing adsorption (S-TVSA) cycle. The cycle consists of four steps: (1) adsorption, where a fan blows atmospheric air (mostly N_2 and O_2) at T_L and p_H through a packed column until CO_2 saturation; (2) blowdown, where the inlet is closed, and the column is evacuated to pressure p_L using a vacuum pump, releasing air back into the atmosphere; (3) heating, where heat at T_H is transferred via an external jacket, with the gaseous stream mainly

being air released back; and (4) desorption, with a steam purge providing additional temperature and vacuum swings[1].

Table 2 Adsorption Column specifications

Column	
Column length L (m)	0.0100
Inner diameter (m)	0.0800
Outer diameter (m)	0.0820
Bed density (kg/m^3)	55.4000
Particle size (m)	0.0075
Wall heat capacity (J/kg/K)	513.0000
Solid heat capacity (J/kg/K)	2070.0000
Fluid Heat transfer coefficient (W/m^2/K)	3.0000
Wall Heat transfer coefficient (W/m^2/K)	26.0000
CO_2 heat of adsorption (kJ/mol)	-57.0000
H_2O heat of adsorption (kJ/mol)	-49.0000
CO_2 Heat capacity (J/K/mol)	42.4600
H_2O Heat capacity (J/K/mol)	73.1000
CO_2 Mass transfer coefficient (1/s)	0.0002
H_2O Mass transfer coefficient (1/s)	0.0020

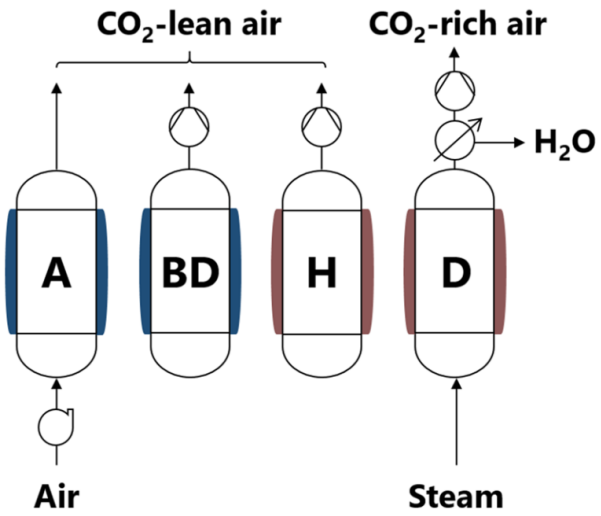

Figure 1. S-TVSA cycle [1]

The boundary conditions for each step is reported in Table 3.

Table 3 S-TVSA boundary conditions[1, 5]

Step	Time (s)	Boundary condition
Adsorption	13, 772	$p\mathrm{I}_{Z=L} = p_H$
		$u\mathrm{I}_{Z=0} = u_{air}$
		$T\mathrm{I}_{Z=0} = T_{air}$
		$y_i\mathrm{I}_{Z=0} = y_{i,air}$
Blowdown	30	$p\mathrm{I}_{Z=L} = p(t)$
		$u\mathrm{I}_{Z=0} = 0$
		$\frac{\partial T}{\partial z}\mathrm{I}_{Z=0} = 0$
		$\frac{\partial y_i}{\partial z}\mathrm{I}_{Z=0} = 0$
Heating	704	$p\mathrm{I}_{Z=L} = p_L$ (0.05 bar)
		$u\mathrm{I}_{Z=0} = 0$
		$\frac{\partial T}{\partial z}\mathrm{I}_{Z=0} = 0$
		$\frac{\partial y_i}{\partial z}\mathrm{I}_{Z=0} = 0$
Desorption	30, 000	$p\mathrm{I}_{Z=L} = p_L$
		$u\mathrm{I}_{Z=0} = u_{steam}$
		$T\mathrm{I}_{Z=0} = T_{steam}$
		$y_i\mathrm{I}_{Z=0} = y_{i,steam}$

2.3 Performance assessment of S-TVSA

2.3.1 Key Performance Indicators

The performance of the adsorption process was assessed using four key indicators: CO_2 Purity, CO_2 productivity, thermal energy use, and electrical energy use. The CO_2 Purity in the desorption step is defined as:

$$Pu_{CO_2}(\mathrm{mol\%}) = 100 \times \frac{n_{CO_2}^{prod}}{n_{CO_2}^{prod} + n_{H_2O}^{prod} + n_{N_2}^{prod}} \quad (10)$$

Where n_i^{prod} is the number of moles of component i produced from the desorption step. CO_2 productivity is defined as the capture rate per unit volume of the adsorption bed over the duration of a complete cycle. The CO_2 productivity is defined as:

$$Pr_{CO_2}(kg/m^3/day) = \frac{m_{CO_2}^{prod}}{V_{bed} \cdot t_{cycle}} \quad (11)$$

Where $m_{CO_2}^{prod}$ is the mass CO_2 produced during the desorption step (kg), V_{bed} is the adsorption bed volume (m^3), and t_{cycle} is the cycle time (s). Thermal energy consumption comprises the jacket heating required during the heating and desorption steps and the heat supplied

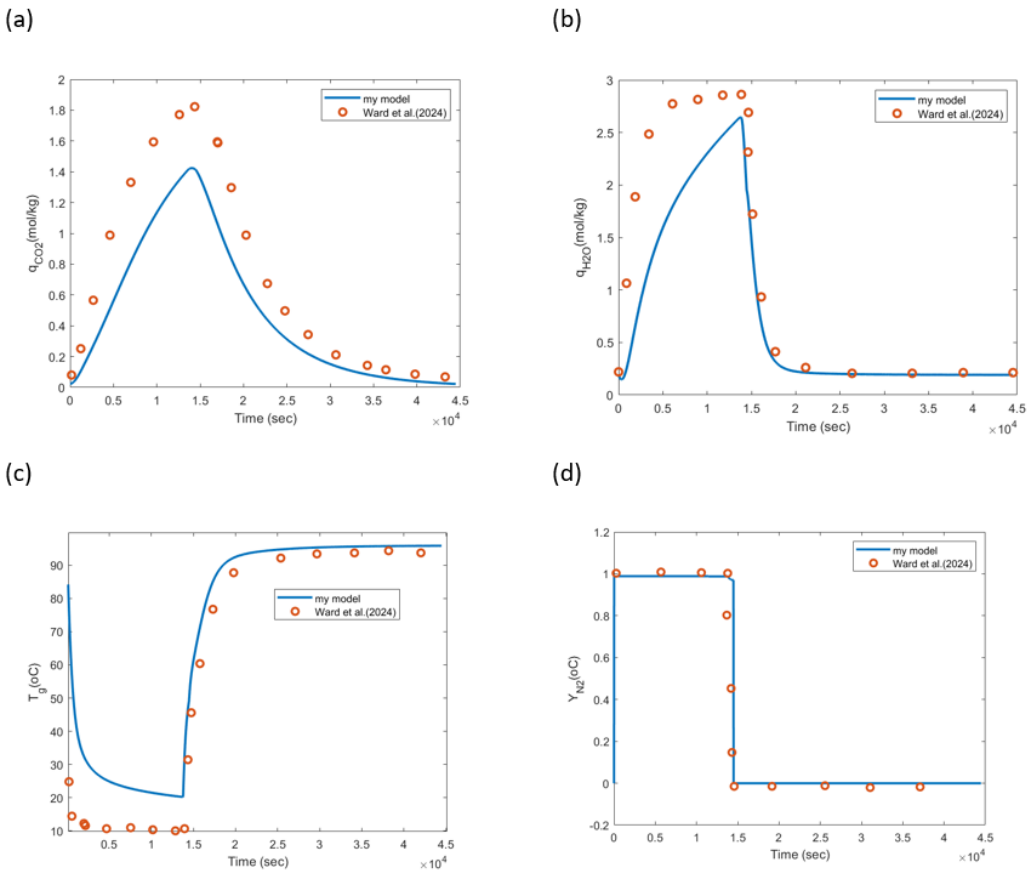

Figure 2: TVSA Cycle performance compared with Ward et al[5]. (a) CO_2 loading (b) H_2O loading (c) gas temperature (T_g)(d) N_2 composition

by the steam during the desorption step. The specific thermal consumption (Q_{therm}) is defined:

$$Q_{therm}(MJ/kg) = \frac{Q_{heat}^{Jacket} + Q_{des}^{jacket} + Q_{des}^{steam}}{m_{CO_2}^{prod}} \quad (12)$$

Where Q_i^{Jacket} is the heat (MJ) required in the jacket for step i, and Q_{des}^{steam} is the heat (MJ) supplied by the stream during the desorption step. The electrical energy consumption includes the energy required to operate the compressor during both adsorption and desorption steps, as well as the energy required for the vacuum pump to create and maintain a vacuum during the blow-down and heating stages. The electrical energy consumption is defined as:

$$E_{elec}(MJ/kg) = \frac{E_{ads} + E_{bd} + E_{heat} + E_{des}}{m_{CO_2}^{prod}} \quad (13)$$

Where E_i is the electrical energy (MJ) consumption of step i. To facilitate representing the total energy duty of the capture process as a single value for process optimisation, we use the concept of specific equivalent work to combine electrical and thermal energy consumption. The specific equivalent work is defined as:

$$W_{eq}(MJ/kg) = E_{elec} + \eta_{turb}\left(1 - \frac{T_L}{T_H}\right) \quad (14)$$

Where η_{turb} is the turbine isentropic efficiency of a hypothetical turbine undertaking the conversion between electrical and thermal work. η_{turb} was assumed to be 75%.

2.3.2 Process optimisation

The solid-based DAC performance was optimised by maximising system productivity and minimising energy use. CO_2 productivity quantifies the system volume required to achieve a specified CO_2 capture rate, thereby affecting the land footprint. Since DAC must be deployed globally for a significant climate impact, high productivity minimises land use. The aim is to minimise energy consumption in a steam-assisted DAC process. The multi-objective optimisation was formulated as follows:

$$\min_{\theta}[-Pr_{CO_2} + \phi, W_{eq} + \phi] \quad (15)$$

$$s.t\ \theta_L \leq \theta \leq \theta_U$$

$$\phi = 0.25 \times [\max(0, 95 - Pu_{CO_2})]^2$$

Where θ is the vector of the operating variables, θ_L and θ_U are the lower and upper operating variable limits (see Table 4). A constraint was imposed on the CO_2 purity, requiring a minimum of 95%. ϕ is the penalty function.

Table 4 Lower and upper limits

Parameter	Lower Limit	Upper Limit
t_{ads}(s)	10, 000.000	20, 000.000
t_{heat}(s)	500.000	1, 500.000
t_{des}(s)	20, 000.000	40, 000.000
p_L(bar)	0.050	0.500
T_{des}(°C)	80.000	100.000
v_s(m/s)	0.001	0.005
v_f(m/s)	0.005	0.010

We deployed the NSGA-II algorithm from MATLAB's Global Optimization Toolbox as the gamultiobj function, running for 100 generations with a population of 140. It was initialized with 140 operating points and their key performance indicators, determined by quasi-random Sobol sampling within bounds in Table 4.

3. RESULTS

3.1 TVSA Cycle performance

The behaviour of the S-TVSA cycle was analysed through CO_2 and H_2O loading profiles in comparision with Ward et al.[5]. Figure 2(a) shows CO_2 loading increasing during the adsorption phase (13, 772 s) at 20 °C and 1 bar.

Figure 2(b) presents H_2O loading, indicating sorbent stability under humid conditions, which is crucial for DAC applications. Thermal dynamics are shown in Figure 2(c), with the gas temperature (T_g) rising to approximately 95 °C during heating and desorption, driven by steam injection and external heating. Figure 2(d) tracks N_2, which evacuates rapidly during blowdown and desorption, maintaining high CO_2 purity.

3.2 Energy consumption

The assessment (Figure 3) shows thermal energy (Q_thermal) as the dominant component of total energy, accounting for jacket heating and steam regeneration (7.04 MJ/kg). Electrical energy (0.37 MJ/kg) primarily powers blowers and vacuum pumps across various phases. The Specific Equivalent Work (W_{eq}) determined is 7.41 MJ/kg.

3.3 Process optimisation

The optimisation results, generated using the NSGA-II algorithm with 100 generations and a population of 140, are summarised in the Pareto Curve (Figure 4). The results demonstrate a clear trade-off between CO_2 productivity and energy consumption. Initial points derived via Sobol sequence sampling within the bounds helped identify an optimal operating window that balances throughput with energy efficiency. The optimal solution yielded a productivity range of 32.6-33.4kg/m^3/day. The corresponding energy requirement for the optimal points ranges from 7.0 to 7.6 MJ/kg.

The multi-objective optimisation reveals a key link between the S-TVSA cycle's physical performance and the economic viability of solid-based DAC. The Pareto front marks the 'best possible' performance. Increasing CO_2 productivity raises specific work (W_{eq}) and reduces adsorbent bed volume (V_{bed}), lowering capital costs and land requirements. The 33.4 kg/m^3/day point boosts productivity and cuts CAPEX; the 7.0 MJ/kg point minimises OPEX by reducing thermal energy ($Q_{thermal}$).

Thermal energy far exceeds electrical energy, making the heat source critical. Waste heat or low-cost geothermal energy for the heating and desorption steps could greatly lower OPEX, favouring high-productivity points.

4. CONCLUSION

This study developed and validated a robust fixed-bed model and an S-TVSA cycle for solid-sorbent-based DAC. The process optimisation with NSGA-II revealed that thermal energy is the main factor driving the energy penalty. Key findings include:

- APDES-NFC demonstrates reliable co-adsorption of CO_2 and H_2O, making it suitable for environments with varying humidity.
- $Q_{thermal}$ is the dominant factor in W_{eq}, necessitating advanced regeneration strategies or materials with higher working capacities.
- Optimal solutions achieved CO_2 productivity between 32.6 and 33.4 kg/m^3/day. The corresponding specific equivalent work is between 7.0 to 7.6 MJ/kg. This provides a pathway to balancing the trade-off between plant size (CAPEX) and energy demand (OPEX).
- Findings indicate that solid-sorbent DAC's economic feasibility largely relies on low-carbon, low-cost thermal energy for desorption.

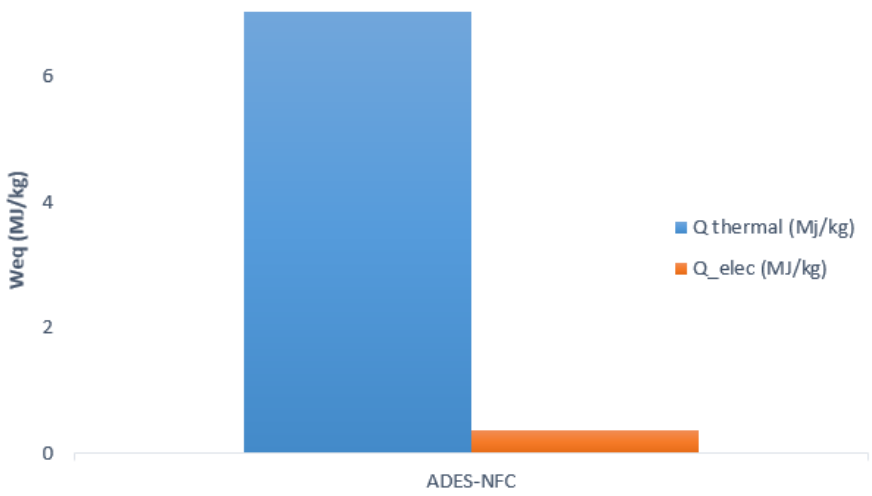

Figure 3. Specific equivalent work

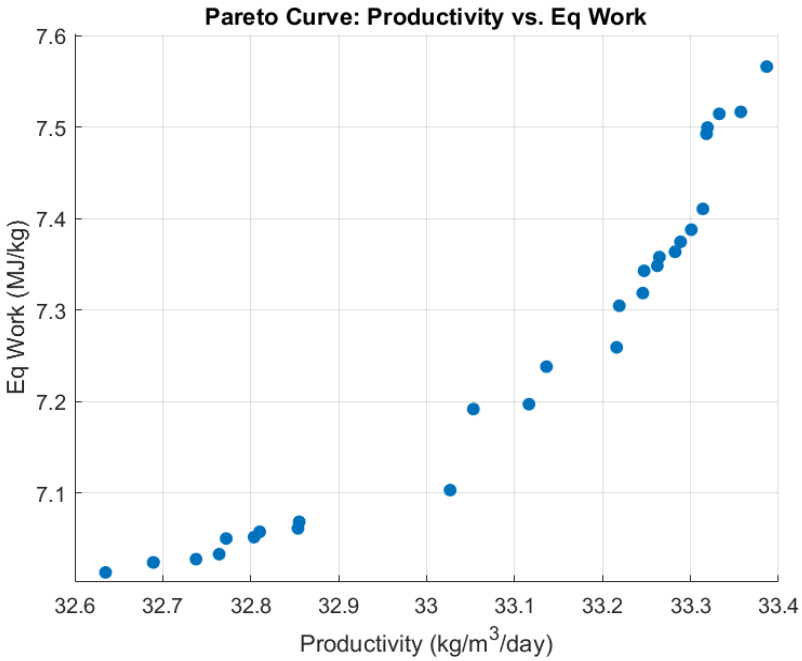

Figure 4. Pareto Curve

Future work will focus on a detailed parametric analysis and the impact of water-enhanced amine-functionalised adsorbents on the DAC performance.

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AUTHOR IDENTIFIERS

Author ORCIDs:
Toluleke E Akinola: 0000-0003-4379-9425
Meihong Wang: 0000-0001-9752-270X

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