CASE REPORT

Experimental study on waste heat recovery characteristics of inorganic ceramic membrane flue gas

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ABSTRACT

The wet saturated flue gas discharged by coal-fired utility boilers leads to a large amount of low-temperature waste heat loss. Inorganic ceramic membrane is acid-base resistant and has strong chemical stability. It is an ideal material for recovering low-temperature waste heat from flue gas. The experiment of waste heat recovery of flue gas was carried out with inorganic ceramic membrane as the core, and the characteristic parameters of low-temperature flue gas at the tail of the boiler were analyzed; taking 316 L stainless steel as the comparative object, the strengthening effect of inorganic ceramic film on improving heat recovery power and composite heat transfer coefficient was discussed. The results show that the waste heat recovery of flue gas is mainly the evaporation latent heat recovery of water, accounting for about 90%; circulating water is used as cooling medium, and the waste heat recovery capacity of flue gas is stronger; compared with circulating water, when air is used as the cooling medium, the effect of inorganic ceramic membrane flue gas waste heat recovery is more significant, and the enhancement coefficient is as high as 9; increasing the flue gas flow is helpful to improve the heat recovery power and composite heat transfer coefficient; at the same time, inorganic ceramic membrane can also recover condensate with high water quality. The results of this paper can provide a reference for the application of inorganic ceramic membrane in flue gas waste heat recovery.

Keywords: Ceramic Membrane; Waste Heat of Flue Gas; Latent Heat; Heat Recovery Power; Composite Heat Transfer Coefficient

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1. Introduction

For a long time, traditional coal-fired power stations, as the main force of China's power supply, have provided strong support for the high-quality social development^[1]. At the same time, coal-fired power stations also consume a large amount of coal resources, and the boiler tail flue gas emissions of unsaturated or wet saturated flue gas lead to huge low-temperature waste heat loss^[2]. Taking a 330 MW Unit Boiler of a coal-fired power plant as an example, the outlet flue gas temperature of its wet desulfurization device is 55 °C, and the low-temperature waste heat loss caused by the discharge of wet saturated flue gas is 484 Gj/h, which is equivalent to a waste of 16 t/h standard coal according to the calorific value, and about 80% of the heat is lost in the form of water vaporization latent heat.

At present, the waste heat recovery of wet saturated flue gas at the tail of coal-fired power station boiler mainly includes condensation method, membrane separation method and absorption method. Indirect condensation is the most common method of condensation^[3]. The cooling medium and wet saturated flue gas flow on both sides of the

corrosion-resistant material. The two are not in direct contact, and the flue gas temperature is reduced through heat conduction of the heat exchange material to realize the recovery of flue gas waste heat. The commonly used corrosion-resistant heat exchange materials are mainly fluoroplastics. Xiong et al.^[4] installed a two-stage fluoroplastic heat exchanger at the tail of the wet desulfurization tower of 660 MW coal-fired unit to recover 92 MW of flue gas waste heat. Compared with indirect condensation, direct contact condensation can help reduce the consumption of corrosion-resistant materials by adding a spray tower in the flue at the tail of the boiler^[5]. In addition, direct contact spray towers are usually used in combination with absorption heat pumps. Zhu et al.^[6] used absorption heat pump to provide cold source and direct contact spray tower to recover flue gas waste heat, which can improve the heating capacity of the boiler by 12%. Membrane separation method is based on the selective transportation of flue gas moisture by porous materials to realize the recovery of flue gas waste heat and recover condensate with high water quality^[7,8]. Membrane materials used in membrane separation method can be divided into organic membrane and inorganic membrane. Organic membrane materials are easy to be corroded in acidic environment, resulting in membrane surface defects^[9], while inorganic membrane materials have good chemical stability, acid resistance, alkali resistance, high temperature resistance and organic solvent resistance^[10], and are ideal membrane materials for waste heat recovery of boiler tail gas in coal-fired power stations. Wang et al.[11] used hydrophilic inorganic nanoceramic membrane to recover flue gas waste heat, and discussed the effects of flue gas temperature, flue gas flow rate and cooling medium on heat recovery characteristics. The absorption method is based on the fact that the dehumidification solution has a low saturated water vapor pressure, such as lithium bromide, lithium chloride, calcium chloride, triethylene glycol, etc. By forming a pressure difference with the partial pressure of water vapor in the flue gas, the water vapor in the flue gas is forced into the dehumidification solution^[12,13] to release the latent heat of vaporization. Wang et al.^[10] used lithium bromide solution as dehumidification solution to establish an open absorption heat pump flue gas waste heat recovery system model, and found that water vapor absorption and regeneration are very important for system operation.

To sum up, the technology of condensing flue gas waste heat recovery is mature and reliable, but seeking corrosion-resistant materials with high heat transfer coefficient is the key to restrict its wide promotion; based on the hygroscopic characteristics of dehumidification solution, absorption method has high waste heat recovery capacity, and the regeneration of water leads to its complex system structure and high cost; the system structure of membrane separation method is similar to that of condensation method. It not only has high heat transfer coefficient of convective heat transfer and phase change heat transfer, but also can recover condensate water with high quality^[14,15].

In this paper, inorganic porous ceramic membrane is used to carry out the experiment of deep recovery of flue gas waste heat with air or circulating water as cooling medium; analyze the heat transfer models under different cooling media, and take 316 L stainless steel as the reference object to explore the strengthening effect of inorganic ceramic membrane on the performance of flue gas waste heat recovery.

2. Experiment content

2.1 Experimental system

The experimental system of deep recovery of waste heat from flue gas with inorganic ceramic membrane is shown in **Figure 1**.

In **Figure 1(a)**, the simulated flue gas continuously and stably supplied by the air compressor is heated and humidified by a constant temperature water bath to simulate the real flue gas. The metal tube rotameter measures the volume flow of the simulated dry flue gas. The simulated flue gas is on the shell side of the membrane module, and the circulating water cooling medium is on the tube side of the membrane module. The simulated flue gas and circulating water realize countercurrent heat exchange. The circulating water in the membrane is in a negative pressure state, and

semiconductor refrigeration is used to maintain the temperature of the circulating water cooling medium constant. At the same time, the circulating water cooling medium pipeline is equipped with a water tank, and the upper water collecting opening of the water tank is used to collect the water recovered by the inorganic ceramic membrane.



(a) Circulating water

(1)—air compressor;
 (2)—data acquisition computer;
 (3)—constant temperature water bath;
 (4)—inertia gas-liquid separator;
 (5)—membrane module;
 (6)—diaphragm water pump;
 (7)—water chiller;
 (8)—circulating water tank;
 (9)—measuring cylinder;
 a—metal tube rotameter;
 b—thermocouple;
 c—pressure gauge;
 d—turbine flowmeter.



(b) Air

1—air compressor; 22—data acquisition computer; 33—constant temperature water bath; 43—inertia gas-liquid separator; 55 —membrane module; 66—centrifugal fan; 77—water chiller; 88—condenser; 99—condensate collector; a—metal tube rotameter; b—thermocouple; c—pressure gauge; d—glass tube rotameter.

Figure 1. Schematic diagram of the inorganic ceramic membrane recovery waste heat system.

Table 1. Experimental parameters						
Project	Parameter	Project	Parameter			
Flue gas	56 57	Flue gas	5 0 15			
temperature/°C	30~37	$flow/(L \cdot min^{-1})$	5, 9, 15			
Air	12	Air	0, 1, 5, 10,			
temperature/°C	12	$flow/(L \cdot min^{-1})$	15			
circulating		Circulating				
water	18	water	15~145			
temperature/°C		$flow/(L \cdot h^{-1})$				

Table 1 shows the selection of experimentaloperating parameters. The cooling media arecirculating water and air respectively.

2.2 Experimental materials

The heat exchange materials in the membrane module are 316 L stainless steel and inorganic

ceramic membrane, both of which are tubular structures. 316 L stainless steel is heat transfer and non mass transfer material, while inorganic ceramic membrane is heat transfer and mass transfer material. **Figure 2** shows the SEM (scanning electron microscope) image of the inorganic ceramic film sample. Compare **Figures 2(a)** and (b), at the same magnification (\times 300) the pore diameter of the outer facial mask of the lower ceramic membrane is much smaller than that of the inner facial mask, while **Figure 2(c)** shows that the cross section of the ceramic membrane has an obvious layered structure, so the inorganic ceramic membrane used in this experiment is an outer coating structure.



(a) outer surface (b) inner surface (c) cross section Figure 2. SEM images of the inorganic ceramic membrane sample.

Figure 3 shows EDS (energy dispersive spectrometer) spectra of inorganic ceramic membrane samples. Qualitative analysis of Figure 3 shows that PT element is introduced into the sample after spraying gold, and the other two main elements are oxygen and aluminum. It can be seen that the main component of inorganic ceramic film in the experiment is Al₂O₃.

The inorganic ceramic membrane was tested and analyzed by mercury intrusion to understand its pore structure (Figure 4). It can be seen from Figure 4 that with the decrease of the pore size of the inorganic ceramic membrane, there are two peaks of mercury intrusion. The first peak appears in the support layer and the second peak appears in the separation layer. See Table 2 for the specific structural parameters of membrane components of flue gas waste heat recovery system.



Figure 3. EDS spectra of the inorganic ceramic membrane sample.



Figure 4. EDS spectra of the inorganic ceramic membrane sample.

 Table 2. Structural parameters of the inorganic ceramic membrane modules

Project	Ceramic film	Stainless steel	Assembly housing
Coating	Topcoat		
Material Science	Al_2O_3	316 L	316 L
Aperture/nm	200		
Length/mm	790	790	800
Outer diameter/mm	12	12	22
Inner diameter/mm	8	10	20
Porosity/%	33		
Outer membrane area/cm ²	297.67	297.67	
Film area/cm ²	198.45	248.06	
Flow section/cm ²	0.50	0.50	2.01

2.3 Data collection

There are many types of data acquisition instruments in the experimental system, which are used to measure gas flow, liquid flow, gas temperature, liquid temperature, etc. The specific instrument parameters are shown in **Table 3**. All data acquisition instruments output 4~20 ma current signals, which are read and stored by the multi-function data acquisition box.

Name	Model	Range	Accuracy			
Metal tube rotameter	CGYL-LZ-25	0~30 L/min	1.00			
Turbine flowmeter	LWGY-4-C-10	0~150 L/h	1.00			
Glass rotameter	LZB-6W	0~15 L/min	1.50			
Temperature transmitter	SWB-B	0~100 °C	0.25			
Pressure transmitter	CGYL-202	-50~50 kPa	0.25			

3. Evaluation criteria for recovery characteristics

3.1 Heat transfer model

Figure 5 shows the counter current distribution of the fluid inside and outside the inorganic ceramic membrane. Inorganic ceramic membrane is different from 316 L stainless steel. Flue gas moisture can enter the ceramic membrane tube across the membrane, so that the liquid film cannot be formed on the flue gas side or the liquid film is thin, which helps to reduce the heat transfer resistance on the flue gas side. The shell side of membrane module is simulated flue gas, and the heat transfer process of its outer surface is mainly

convective heat transfer and phase change heat transfer. When the cooling medium on the tube side of the membrane module is air, the moisture in the smoke enters the membrane from the outside of the membrane. Due to the low moisture content of the air, the moisture changes with the air circulation, so the heat transfer process on the inner surface is mainly convective heat transfer and phase change heat transfer. When the cooling medium on the tube side of the membrane module is circulating water, the flue gas moisture entering the membrane across the membrane will not change phase. At this time, the heat transfer process on the inner surface is mainly convective heat transfer.



Figure 5. Countercurrent distribution of fluid inside and outside the inorganic ceramic membrane.

3.2 Flue gas heat recovery index

The deep recovery of waste heat from flue gas at the tail of coal-fired utility boilers is mainly divided into two types: flue gas sensible heat and vaporization latent heat. The characteristics of deep recovery of flue gas waste heat by inorganic ceramic membrane were analyzed based on the change of flue gas waste heat. The latent heat of vaporization of flue gas moisture accounts for a large proportion, so the recovery ratio coefficient of flue gas moisture latent heat is defined ψ :

$$\frac{Q_{s}}{V_{g}\rho_{g}[(d_{T_{i}^{g}}-d_{T_{b}^{g}})(T_{i,g}-T_{o,g})c_{p,w}+1\ 000(T_{i,g}-T_{o,g})c_{p,g}]}{6\times10^{7}}$$
(1)

$$Q_{1} = \frac{V_{g}\rho_{g}\left(d_{T_{1}^{g}} - d_{T_{o}}\right)r}{6 \times 10^{4}}$$
(2)

$$\psi = \frac{Q_1}{Q} \times 100\% = \frac{Q_1}{Q_1 + Q_s} \times 100\%$$

(3)

Where: ψ is the proportion coefficient of flue gas moisture latent heat recovery, %; Q, Q_1 , Q_s is the total heat recovery power, latent heat recovery power, sensible heat recovery power, W; $c_{p,w}$ is the constant pressure specific heat capacity of water, taking 4,200 J/(kg·K); $c_{p,g}$ is the constant pressure specific heat capacity of dry flue gas, taking 1,007 J/(kg·K); $T_{i,g}$, $T_{o,g}$ are the inlet and outlet temperature of flue gas of inorganic ceramic membrane module, °C; $d_{T_p^g}$, $d_{T_0^g}$ are the moisture content of wet saturated flue gas at temperature $T_{i,g}$, $T_{o,g}$, g/kg dry flue gas; r is the latent heat of vaporization of water in flue gas, taking 2,257 kJ/kg; V_g is the volume flow measured by metal tube rotameter (151 kPa, 20 °C), l/min; ρ_g is the density of flue gas (151 kPa, 20 °C), taking 1.80 kg/m³.

The temperature difference between the inlet and outlet of the shell side flue gas or tube side cooling medium of the inorganic ceramic membrane module can be used as an intuitive parameter to reflect the heat exchange capacity of the waste heat of the flue gas:

$$\Delta T_{\rm g} = T_{\rm i,g} - T_{\rm og} \tag{4}$$

$$\Delta T_{\rm m} = T_{\rm i,m} - T_{\rm o,m} \tag{5}$$

Where: Δt_{g} , Δt_{m} is the temperature difference between the inlet and outlet of flue gas on the side of inorganic ceramic membrane module and pipe cooling medium, °C; $T_{i,m}$, $T_{o,m}$ is the inlet temperature of the cooling medium on the tube side of the inorganic ceramic membrane module, which is directly measured by the thermocouple, °C.

The driving force of heat transfer for the deep recovery of waste heat from flue gas of inorganic ceramic membrane components is based on the logarithmic average temperature difference, namely:

$$\Delta T = \frac{\left(T_{\rm i,g} - T_{\rm o,m}\right) - \left(T_{\rm og} - T_{\rm i,m}\right)}{\ln\left[\left(T_{\rm i,g} - T_{\rm o,m}\right) / \left(T_{\rm og} - T_{\rm i,m}\right)\right]}$$
(6)

Where: ΔT is the logarithmic average temperature difference of inorganic ceramic membrane module, °C. The inorganic ceramic membrane is of cylindrical structure, and the change of heat transfer coefficient is discussed with the inner surface as the reference plane. Compared with 316 L stainless steel tube, the heat transfer enhancement coefficient of the inorganic ceramic membrane tube is β , namely:

$$k = \frac{Q}{\Delta T A_{\rm i}} = \frac{Q}{2\pi R \Delta T}$$

$$\beta = \frac{k_{\rm c}}{k_{\rm s}}$$
(7)
(7)

Where: k is the composite heat transfer coefficient, W/(m².°C); A_i is the internal surface area, m²; R is the inner radius, m; β is the heat transfer enhancement coefficient of inorganic ceramic film relative to 316 L stainless steel; k_c and k_s are the composite heat transfer coefficients of inorganic ceramic film and 316 L stainless steel respectively, W/(m².°C).

3.3 Flue gas water recovery index

The performance index of water recovery is

mainly the water recovery rate. The water recovery rate of flue gas can be directly measured by using the collector of circulating water tank or condensate collector v, namely:

 $v^{=}$

$$=\frac{3V}{50S\Delta t}$$
(9)

Where: v is the flue gas moisture recovery rate, L/(m·h); s is the contact area between inorganic ceramic membrane and flue gas, taking 297.67 × 10^{-4} m²; Δt is the running time, min; V is amount of water collected by inorganic ceramic membrane within Δt time, mL.

4. Result analysis

4.1 Temperature difference between inlet and outlet of flue gas

The cooling media used in this experiment are air and circulating water respectively. Figure 6 shows the influence of the cooling medium flow on the inlet and outlet temperature difference of flue gas. It can be seen from Figure 6 that when air is used as the cooling medium, the temperature difference between the inlet and outlet of inorganic ceramic membrane flue gas is much greater than that of 316 L stainless steel flue gas; when circulating water is used as the cooling medium, the difference between the inlet and outlet temperature difference of inorganic ceramic membrane flue gas and that of 316 L stainless steel flue gas is small, and when the flue gas flow is 15 L/min, the inlet and outlet temperature difference of 316 L stainless steel flue gas is higher than that of 200 nm ceramic membrane flue gas. The temperature difference between inlet and outlet of flue gas with circulating water as cooling medium is higher than that with air as cooling medium; at the same time, whether using air as the cooling medium or circulating water as the cooling medium, the higher the flue gas flow, the smaller the corresponding temperature difference between the inlet and outlet of flue gas.





Figure 7. Change of the temperature difference of inlet and oult flue gas (air) with air flow rate.

When air is used as the cooling medium, the shell side and tube side of the membrane module are both gas, and the specific heat capacity of the gas is small. Figure 7 shows the change of temperature difference between inlet and outlet of flue gas on shell side or air on tube side of inorganic ceramic membrane module with air flow. It can be seen from Figure 7 that when inorganic ceramic film or 316 L stainless steel is used as heat exchange material, with the change of air flow, the temperature difference between the inlet and outlet of air is always higher than that of flue gas, which is consistent with the change of calorific value under the unit temperature difference corresponding to different temperatures (Figure 8). This is because at relatively high temperatures, the moisture content of wet saturated gas is large, and the latent heat of vaporization of water is high, resulting in large changes in the calorific value of gas per unit temperature difference at higher temperatures.



Figure 8. The calorific values of unit temperature difference at different temperatures.

4.2 Waste heat recovery power

The waste heat recovery power of flue gas is mainly affected by flue gas flow, cooling medium and heat exchange materials, and its changes are shown in Figure 9. It can be seen from Figure 9 that when the inorganic ceramic membrane is used as the heat exchange material, the flue gas waste heat recovery power increases significantly with the increase of flue gas flow, and the flue gas waste heat recovery power with circulating water as the cooling medium is generally higher than that with air as the cooling medium. When 316 L stainless steel is used as the heat exchange material and air and circulating water are used as the cooling medium respectively, the waste heat recovery power of flue gas varies greatly, which is higher than the difference between them in Figure 9(a). Comparing Figure 9(a) with Figure 9(b), it can be found that when air is used as the cooling medium, the flue gas waste heat recovery power of inorganic ceramic membrane is higher than that of 316 L stainless steel, while when circulating water is used

as the cooling medium, the difference between the two is small.



Figure 9. Changes of flue gas waste heat recovery power with flue gas flow rate, cooling medium and heat exchange material.

The waste heat recovery of flue gas can be divided into latent heat recovery and sensible heat recovery. **Figure 10** shows the change of flue gas sensible heat (latent heat) recovery power and latent heat proportion of 316 L stainless steel. It can be seen from **Figure 10(a)** that with the increase of flue gas flow or air flow, the flue gas latent heat recovery power increases faster, while the flue gas sensible heat recovery power changes less; the proportion of latent heat increases with the increase of flue gas flow, up to 92%. It can be seen from **Figure 10(b)** that the flue gas latent heat recovery

power also increases with the increase of flue gas flow, and the change of circulating water flow has no obvious law on the latent heat recovery, but the proportion of latent heat is also large. Comparing **Figure 10(a)** with **Figure 10(b)**, it is found that the recovery power of sensible heat or latent heat of 316 L stainless steel with air as the cooling medium is less than that with circulating water as the cooling medium, and the proportion of latent heat with circulating water as the cooling medium is lower than that with air as the cooling medium.



Figure 10. The recovery power of sensible heat and latent heat and the proportion of latent heat (316 L stainless steel).



Figure 11. The recovery power of sensible heat and latent heat and the proportion of latent heat (inorganic ceramic membrane).

Figure 11 shows the change of sensible heat (latent heat) recovery power and latent heat proportion of inorganic ceramic membrane flue gas. It can be seen from Figure 11 that when air is used as the cooling medium, the flue gas sensible heat and latent heat recovery power of inorganic ceramic membrane is significantly higher than that of 316 L stainless steel, and the latent heat proportion basically does not change with the change of air flow; when circulating water is used as the cooling medium, the difference between the sensible heat and latent heat recovery power of inorganic ceramic membrane and 316 L stainless

steel flue gas is small, and the proportion of latent heat increases with the increase of flue gas flow, which is relatively stable with the change of circulating water flow.

4.3 Composite heat transfer coefficient

The composite heat transfer coefficient can reflect the strengthening effect of inorganic ceramic film on the deep recovery of flue gas waste heat relative to 316 L stainless steel. **Figure 12** shows the change of composite heat transfer coefficient with the flow of cooling medium.



Figure 12. Change of the compound heat transfer coefficient with cooling medium flow rate.



It can be seen from Figure 12 that with air as the cooling medium, the composite heat transfer coefficient of inorganic ceramic film is much higher than that of 316 L stainless steel, and the enhancement coefficient increases with the increase of air flow; with circulating water as the cooling medium, the composite heat transfer coefficient of inorganic ceramic film is slightly higher than that of 316 L stainless steel, and the enhancement coefficient is also lower than that with air as the cooling medium. Comparing Figure 12(a) with Figure the composite heat transfer 12(b), coefficient of inorganic ceramic film with air as cooling medium is equivalent to that of 316 L stainless steel with circulating water as cooling medium.

4.4 Water recovery rate

There are many ways of convective heat transfer and phase change heat transfer on the shell side of the membrane module. The water vapor in the flue gas forms membrane condensation on the outer surface of the inorganic ceramic membrane tube or 316 L stainless steel tube, which increases the heat transfer resistance on the flue gas side; the permeation and recovery of moisture in flue gas by inorganic ceramic membrane can help to reduce the heat transfer resistance of liquid membrane on the flue gas side, and then strengthen the heat transfer. Figure 13 shows the flue gas moisture recovery rate of inorganic ceramic membrane module. It can be seen from Figure 13 that the water recovery rate with circulating water as the cooling medium is generally higher than that with air as the cooling medium; the water recovery rate of inorganic ceramic membrane module is also higher under larger flue gas flow; **Figure 13(a)** the reclaimed water recovery rate first slightly decreases and then increases with the increase of air flow; **Figure 13(b)** the reclaimed water recovery rate increases with the increase of circulating water flow.

5. Conclusion

In this paper, 316 L stainless steel is used as the comparison object, and air or circulating water is used as the cooling medium to carry out the experiment of flue gas waste heat recovery of inorganic ceramic membrane. The heat transfer characteristics of 316 L stainless steel and inorganic ceramic membrane are analyzed from the perspectives of flue gas inlet and outlet temperature difference, latent or sensible heat recovery power and composite heat transfer coefficient. Based on the composite heat transfer coefficient, the enhancement coefficient is specified to reflect the heat transfer enhancement result of inorganic ceramic membrane, finally, the recovery rate of flue gas water from inorganic ceramic membrane is discussed, and the main conclusions are as follows.

(1) The temperature difference between the inlet and outlet of flue gas is greatly affected by the flue gas flow, and the temperature difference between the inlet and outlet of flue gas with circulating water as the cooling medium is higher than that with air as the cooling medium; under the same flue gas parameters, the temperature difference between the inlet and outlet of inorganic ceramic membrane flue gas is generally higher than that of 316 L stainless steel flue gas; when both

sides of the membrane are gas, the temperature difference between the inlet and outlet of flue gas with high moisture content is lower than that of air.

(2) The waste heat recovery of flue gas is mainly based on latent heat recovery. The inorganic ceramic membrane helps to reduce the thickness of the heat transfer boundary layer on the flue gas side. At the same time, the water inside the membrane changes phase to increase the heat transfer effect, so that the waste heat recovery power of flue gas is higher than that of 316 L stainless steel flue gas. Especially when the cooling medium is air, the strengthening effect is more significant. At this time, the heat transfer enhancement coefficient of the inorganic ceramic membrane is as high as 9.

(3) The recovery of waste heat and water from flue gas by inorganic ceramic membrane is complementary. The recovery rate of flue gas water by inorganic ceramic membrane with circulating water as cooling medium is generally higher than that with air as cooling medium.

Conflict of interest

The authors declared no conflict of interest.

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