



EFFECTIVENESS OF METALLIZED-GRAPHITE  
HEAT EXCHANGER

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**ABSTRACT**  
Oleum ( 98% concentrated sulphuric acid) is diluted with water for the production of superphosphate fertilizers. Cooling of the diluted acid is carried out in a heat exchanger made of graphite. Experimental models of heat exchangers simulating an industrial one were prepared from metallized graphite by impregnating synthetic graphite blocks with molten lead, copper and polytetrafluoroethylene (PTFE). The effectiveness of these samples were investigated.

Results obtained show that in a cross-flow heat exchanger, the effectiveness increases with the increase in the heat transfer surface area, the density of the graphite material and the flow velocity of the coolant. The properties of the impregnants such as the specific gravity, thermal conductivity, mass up-take and chemical stability determine the feasibility of the impregnant.

INTRODUCTION

Carbon is one of the most highly corrosion resistant materials being inert to almost all chemicals and conditions except those of a highly oxidizing nature. The thermal characteristics of carbon have been fully discussed (1,2). The thermal conductivity of one form of deposited carbon was found by Brown (3) to be as much as 40 % greater than that of copper. In case of polycrystalline graphite blocks, as used in the construction of heat exchangers, the thermal conductivity is generally 1.1 - 1.5 times greater in the direction of pressure of formation than it is in the transverse direction. The first block-type graphite heat exchanger appeared in the market early in 1950, (4). As it was not convenient to drill long narrow holes through solid blocks, these monoblocks heat exchangers were made by cementing together suitably grooved graphite plates(5). Graphite material was selected in construction of heat exchangers due to its anti-corrosion properties(6). The use of sea-

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water as a coolant was reported(7). In some cases , sodium dichromate salt was added to the coolant in fertilizer plants (8), as well as organophosphate compounds(9). The effectiveness of heat exchangers was studied(10) , the value of which amounts to 80-85 % for graphite material (11).

The aim of the present work was to investigate the effect of some impregnants with metallic nature on the effectiveness of metallized graphite heat exchanger for cooling oleum diluted with water. The parameters studied were the velocity of the coolant , the percent mass up-take of the impregnant and the heat transfer surface area. Pipes as heat exchangers were also studied.

#### EXPERIMENTAL

The experimental part of this paper includes the preparation of the graphite heat exchanger models , the sulphuric acid-water mix and the water coolant. In the industrial plant , the technical data were however obtained . Measurements of the properties of graphite , temperature and heat exchanger effectiveness were determined applying the standard methods. Details are given in the Appendix.

#### RESULTS

Figure 1 shows the effect of the graphite density(of the material used in the construction of the heat exchanger ) on the heat exchange coefficient. It is seen that this coefficient gradually increases with the corresponding increase in the graphite density . Synthetic graphite heat exchanger is more conductive than that prepared from natural graphite. Figure 2 shows the effect of impregnating the synthetic graphite with molten copper , carbon , lead and polytetrafluoroethylene (PTFE) impregnants , on the heat exchange coefficient. It can be seen that the values of this coefficient increase with the impregnant uptake up to 16 % by mass. Such effect decreases in the order molten copper > carbon > molten lead > PTFE polymer. Figure 3 illustrates the effect of flow velocity of the water coolant on the heat exchange coefficient. With synthetic graphite impregnated with 16 % (by mass) carbon or PTFE , increasing the flow velocity of the water coolant from 0.5 to 3.0 m/s results in a corresponding increase of the heat exchange coefficient from 0.46 to 0.84 kW /m<sup>2</sup>/°C respectively.

#### DISCUSSION

The graphite heat exchanger is constructed from impervious blocks made by compressing the composite followed by thermal treatment ( 1200-3000°C) and impregnation to seal its porous texture. In its structure , the graphite crystals are oriented with their axes in the direction which connects the two successive sets of the heat exchanger. Any reasonable number of blocks can thus be assembled in an apparatus to give whatever heat exchange surface area may be required, Plate 1 shows this assembly. In the heat exchanger case under discussion , the effective heat transfer surface area is 22.77m<sup>2</sup>.

being constructed from successive six graphite blocks. The two fluids are cross flowing in the heat exchanger. Both fluids are unmixed, the sulphuric acid and the cooling water pass in the axial and radial directions respectively. The gradual increase in the heat exchange coefficient with the increase in the graphite density is in line with the theory of thermal conductivity model based on lattice vibration (12). Dense body imparts increasing density of the energy carriers. This leads to an increase in the heat exchange coefficient.

Synthetic graphite is more conductive than natural graphite. This is in good agreement with the results given by Tyler(13). The thermal properties as well as the anisotropy results from the variable physico-chemical characteristics of the primary materials, the method of shaping and the degree of graphitization. With synthetic graphite material, the heat exchanger body is fully made of crystallized near ideal graphite lattice. This structure is nearly more perfect than that composed of natural graphite powder bound with amorphous carbon bridges.

The effect of filling in the porous texture of the graphite material with impregnant on the heat exchange coefficient can be explained in the light of the fact that solids satisfying the pores are more conductive than the gaseous species. The differences between the four impregnants are anticipated to the variation in their specific thermal conductivity values. The use of molten copper as an impregnant is not recommended due to its reactivity with the sulphuric acid coming into direct contact. It was used only for modelling. Other impregnants are however nonreactive with the flowing sulphuric acid. The effect of flow velocity of the coolant on the heat exchange coefficient may be explained in the following model. As the coolant comes into contact with the graphite heat transfer surface a boundary layer is established, the stability of which depends on the flow characteristics. With laminar flow (low velocity) the boundary layer is stable. In this case, heat transfers by forced convection to the flowing coolant across this layer i.e.  $(T_g - T_w)$  is low. As the flow velocity increases, the turbulence phenomenon is approached whereby the boundary layer gets successively disrupted. Heat transfer is improved because of the increase in the  $(T_g - T_w)$  value. The effect of turbulence finds support from the results reported by Kazachinskaya (11). In line to this, increasing the heat transfer surface area results in a corresponding increase in the rate of heat transfer leading to a subsequent improvement in the heat exchanger effectiveness. Increase of the heat transfer surface area of holes in the graphite heat exchanger could be exercised by modifying the bore profile in the graphite body keeping the wall thickness separating the axial and radial holes constant.

Under the experimental conditions under discussion, the heat exchanger effectiveness can be computed from the following.

$$N = UA / C_{min} \quad [1]$$

and  $C_{min} = \dot{m} C_p \min, C_{max} = \dot{m} C_p \max$

$$1/U = 1/h_i + x/k + 1/h_o \quad [2]$$

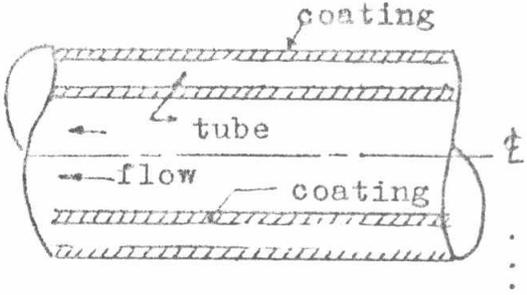
With heat exchanger constructed from pipes , Eq. 2 becomes:

$$\frac{1}{U} = \frac{1}{h_i} + \frac{x_1}{k_1} + \frac{x_2}{k_2} + \frac{x_3}{k_3} + \frac{1}{h_o} \quad [3]$$

where the heat transfer coefficient , h , for the tubes and cross flow is calculated from (16) :

$$Nu = 0.25 Re^{0.6} Pr^{0.38} (Pr_f/Pr_w)^{0.25} , 10^3 Re 2 \times 10^5$$

The thermal conductivity values for the metal-coated tubes were taken as simulated by three resistances connected in series. Vertical and horizontal positioning were also considered in computing the overall thermal conductivity values. Measurements of the heat exchanger effectiveness was computed from the following mathematical relation (16).



$$\epsilon = 1 - \text{Exp} \left[ \frac{\exp(-N C n) - 1}{C n} \right] \quad [4]$$

Table 1 shows the effectiveness values obtained from a mean value of three measurements.

Table 1 Effectiveness of The Heat Exchanger For Cooling Sulphuric Acid-Water Mix

Material of the heat exchanger	ε value , %	
	Bore profile	
	10 mm ∅ (round )	10 mm radius (square)
Synthetic graphite , impregnated with :	5 % PTFE	85.2
	12 % PTFE	86.8
	16 % PTFE	88.2
	16 % Lead	88.9
	16 % Copper	89.3

Table 1 Effectiveness of The Heat Exchanger  
(cont) For Cooling Sulphuric Acid-Water Mix

Material of the heat exchanger	ε value , %	
	Bore profile	
	10 mm ∅ (round )	10 mm radius (square)
8 % carbon	85.0	87.3
12 % carbon	86.1	88.0
16 % carbon	86.7	89.7
Natural graphite impregnated with:		
16 % PTFE	84.0	86.2
16 % Lead	84.5	86.7
16 % carbon	85.2	89.0
PTFE-coated Aluminium	85.8	---
PTFE-coated Cu-Ni	83.6	---

### CONCLUSION

This paper presents a concept of the effectiveness of heat exchanger for cooling concentrated sulphuric acid diluted with water. The construction material made from synthetic graphite and impregnated with a suitable impregnant has been found to be the most effective. The heat exchanger effectiveness could be improved by increasing the heat transfer surface area via modifying the bore profile, densifying the graphite body and increasing the coolant velocity.

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## NOMENCLATURE

A	heat transfer surface area , $m^2$
C	heat capacity , kW/K , $C = \dot{m} C_p$
$C_p$	specific heat at constant pressure of fluid , kJ/kgK
d	diameter , m
h	convective heat transfer coefficient , $W/m^2 \cdot ^\circ C$
k	thermal conductivity , $W/m \cdot ^\circ C$
L	length of holes , m
m	mass , kg
$\dot{m}$	mass flow rate of fluid kg/hour
N	number of heat transfer unit of heat exchanger
n	$= N^{0.22}$
Nu	Nusselt number , $Nu = hd/K$
T	temperature , $^\circ C$
U	overall heat transfer coefficient , $W/m^2 \cdot ^\circ C$
$\mu$	viscosity , $kg/m \cdot s$
v	velocity , m/s
x	thickness , mm
Greek letters	
$\rho$	density , $kg/m^3$
$\epsilon$	effectiveness of the heat exchanger , %
$\pi$	constant , $\pi = 3.1416$

Subscripts  
a for acid  
w for water  
i inlet of acid  
o outlet  
g for graphite

#### APPENDIX

The experimental part of this study included the preparation of the materials , measurements of the heat exchange coefficient and heat exchanger effectiveness applying the standard methods. These were:

#### THE MATERIALS

##### CARBON AND GRAPHITE BLOCKS

Carbon and graphite blocks were prepared by hot mixing coke powder and coal tar pitch as binder. The mix was pressed and baked or graphitized. Details of the process is given elsewhere (14). The blocks were drilled with high carbon steel drills having a diameter of 10 mm, and separated by 7.5 mm. Figures 4 and 5 shows schematic diagram of round and rectangle heat exchanger block.

##### PTFE-COATED ALUMINUM OR Cu-Ni

Pipes having an inner diameter of 10 mm and wall thickness of 1mm were separately coated with PTFE ( Polytetrafluoroethylene copolymer) by dipping technique. Details are given elsewhere (15).

##### SULPHURIC ACID AND WATER

Commercial oleum ( 98% conc sulphuric acid ) was obtained in glass containers. This was diluted with tap water flowing in polyethylene tubes with the required mass flow rate. Acid dilution was conducted just at the entrance of the heat exchanger holes. Water for dilution or for cooling was of tap grade. The industrial graphite heat exchanger shown in Plate 1 is supplied with water taken from the Nile, ( 20 °C).

#### THE EXPERIMENTAL CONDITIONS

Table 2 summarizes the conditions of the case studied.

Property	Material		
	Acid	water for	
		dil.	cooling
$\dot{m}$	21000	6000	100 000
$C_p$	3.316	4.178	4.178
$T_i$	160	20	20
$T_o$	70		35

Table 2 cont.

Property	Material		
	Acid	water for	
		dil.	cooling
$\Delta T$	90		15
$T_{av}$	115		27.5
density, $\rho$	1525	997	997
K	0.685	0.616	0.616
Pr	16.94		5.73
$\mu$	$3.5 \times 10^{-3}$		$8.4 \times 10^{-4}$
d	0.10		0.10
No. of blocks	6		6
No. of holes	302		252
v	0.16		1.48
A	22.77		

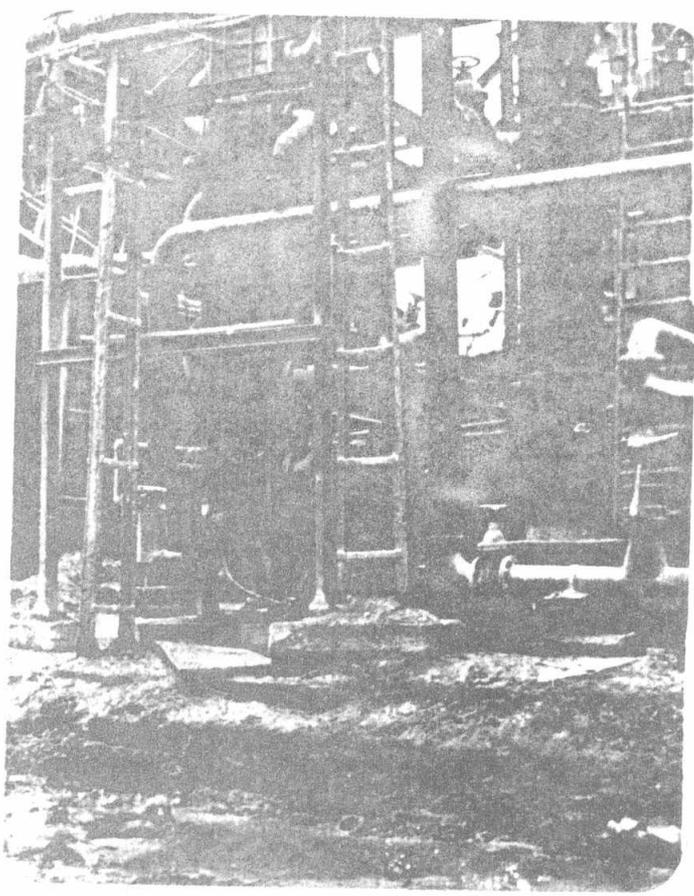


Plate 1 A photograph of the industrial graphite heat exchanger

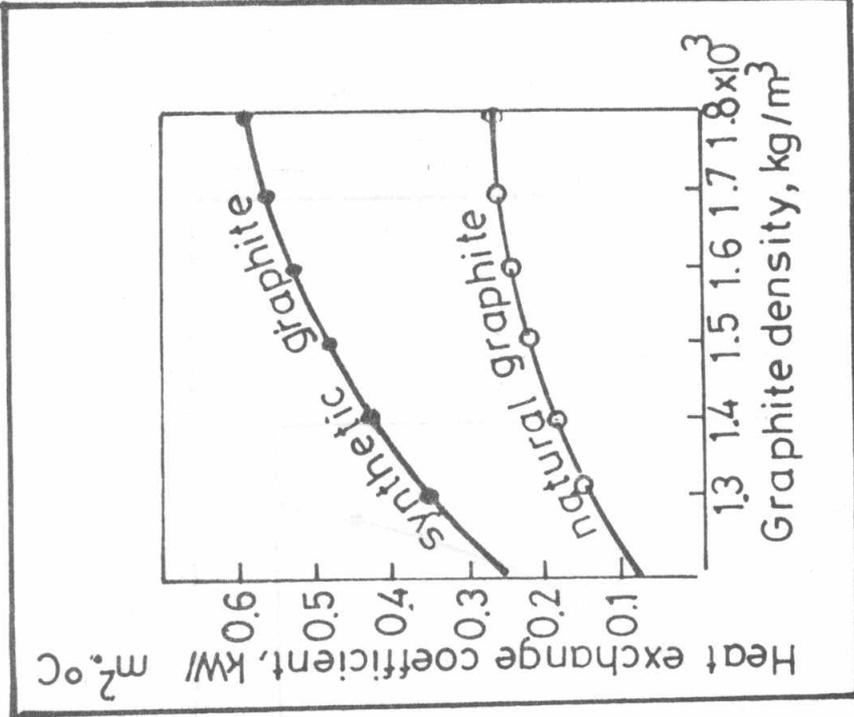


Fig.1 Effect of density of graphite material on heat exchange coefficient.

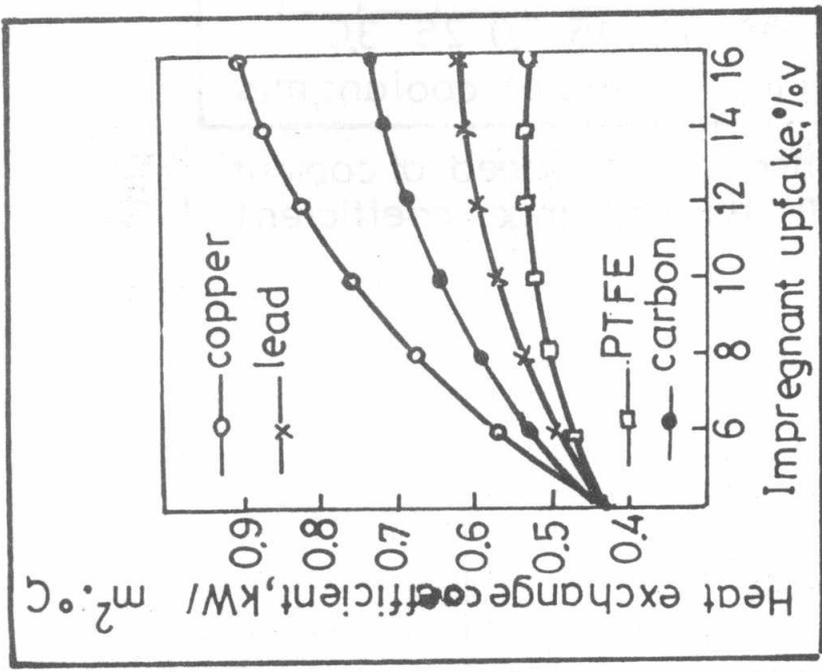


Fig.2 Effect of impregnating graphite ( $\rho=1500 \text{ kg/m}^3$ ) on heat exchange coefficient

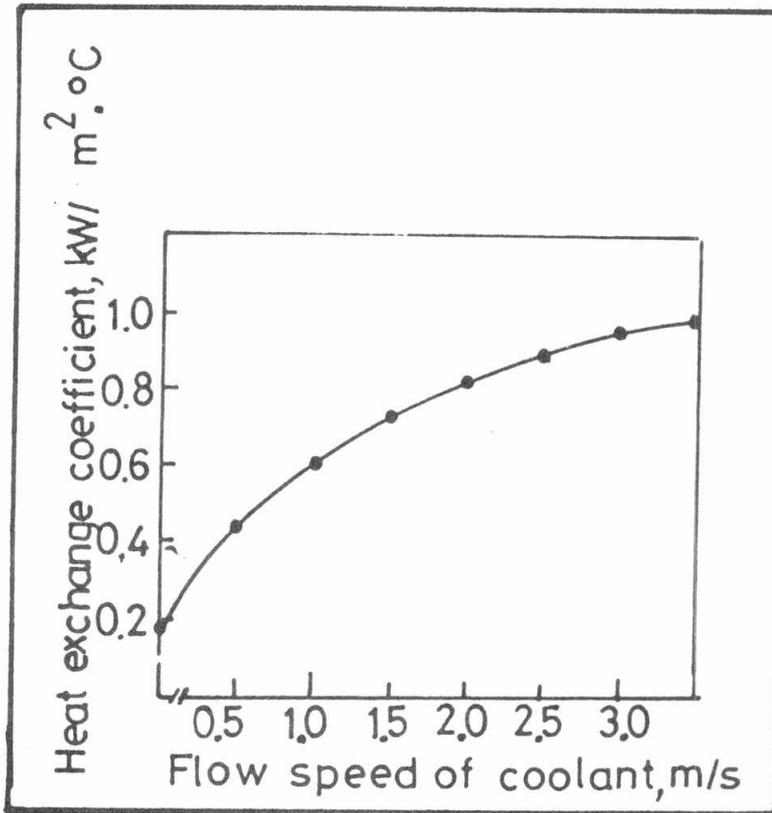


Fig.3 Effect of flow speed of coolant on heat exchange coefficient

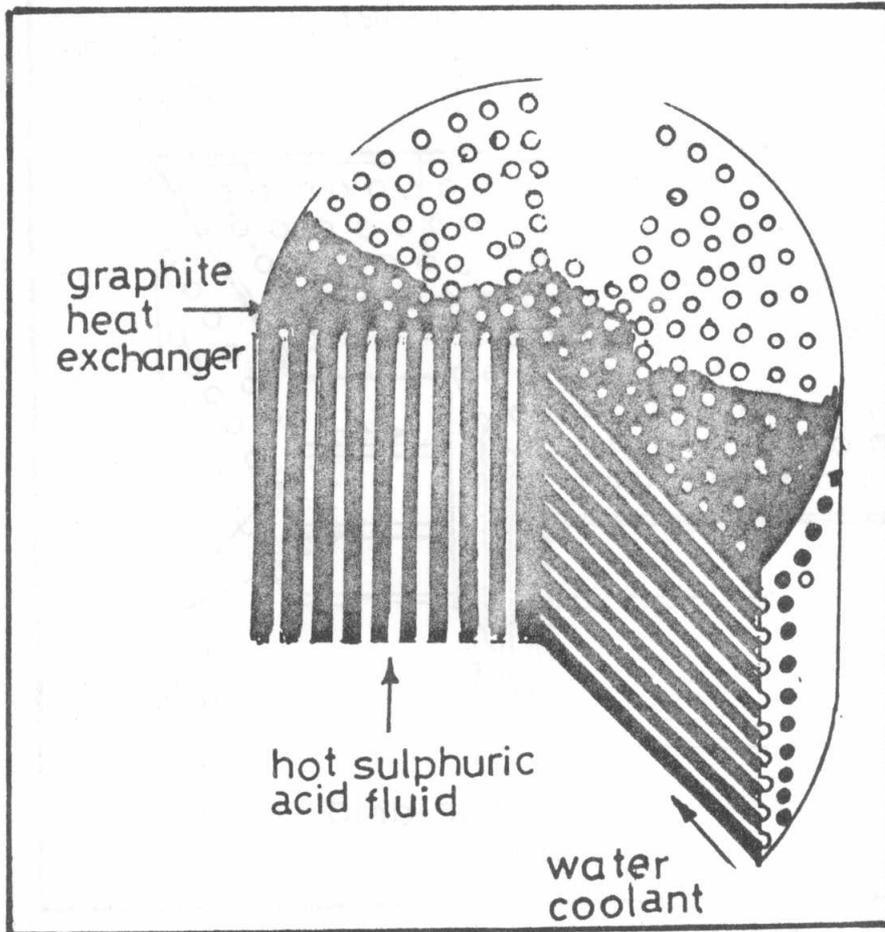


Fig.4 The X-flow bloc heat exchanger arrangement

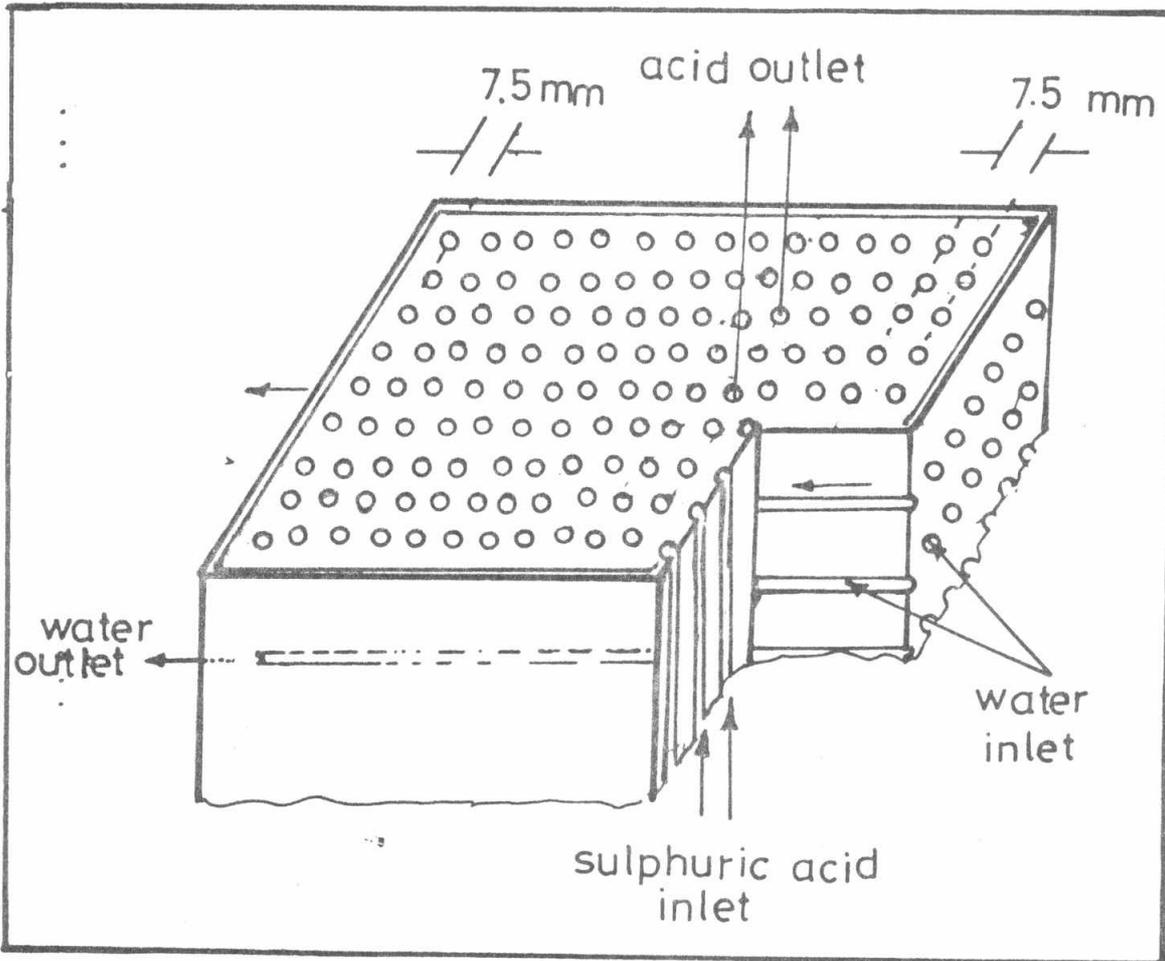


Fig.5 The  $\chi$ - flowbloc heat exchanger arrangement