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"Surface Properties of Mixed oxides of Transition metals"

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Abstract:-

All the samples of the three systems selected for the present study are expected to show the variying catalytic activities. It is known to us that the catalytic activities of the samples would be mainly the functional properties of the surfaces. Thus in the specific condition in which the samples are prepared, the excess surface oxygen (E.S.O) must play an important role in controlling and characterising the surface heterogeneity of the catalyst, a vital factor contributing towards the catalytic reactivity. Sahay etal have observed that acidity, excess surface oxygen and activity for hydrogen peroxide decomposition are directly proportional to each other. Madhok [2] carried out an extensive investigation on oriental adsorption of normal aliphatic alcohols on pervoskite type oxides. He reperted that the change in E.S.O with the decomposition temperature of the catalyst should be related with the catalytic activity.

(Keyword – E.S.O, surface area, acidity, catalytic activity).

Introduction : -

All the samples of the three system namely $Laco_{1-x}Ti_xO_3$, $La_{1-x}Sr_xCoO_3$ and $La_{1-x}Sr_xCo_{1-x}Ti_xO_3$ having different values of x are expected to show the varying catalytic activation. It seem probable that the catalytic activities of the samples would be related to the surface properties. Thus, in the specific condition in which the samples are prepared, the excess surface oxygen must play an important role in controlling and Characterising the surface heterogeneity of the catalyst, a vital factor contributing towards the catalytic reactivity sahay etal' have observed that acidity, excess surface oxygen and activity for hydrogen peroxide decomposition are directly proportional to each other, Madhok[2] carried out extensive investigation on oriental adsorption of normal aliphatic alcohols on perovskite type oxides. He reported that the change in excess surface oxygen with the decomposition temperature of the catalyst should be related with the catalytic

activity. Recently M.N. Rai[3] estimated the excess surface oxygen of nickel oxide prepared from nickel nitrate with different precipitants. He has explained very clearly the role of E.S.O in imparting the activities to the sample for the decomposition of H_2O_2. Recently Bhagat and Saha [4] determined the E.S.O of non stoichiometric oxides of yttrium , Praseodymium and gadolinium and tried to correlate the excess surface oxygen with the catalytic reactivity .

Materials and Method of Preparation:-

Lanthanum oxalates of purity 99.9 % was obtained from Indian Rare earths Limited, Udyogmandal, Kerala State. These were used as such for the sample preparation cobalt oxalate, Nickel oxalate, stoncium carbonate, Titanium oxide and urea all obtained from sarabhai Chemicals Ltd, and were used without further purification. All the samples of the three system were prepared by the oxalate method reported by Raccah and Good enough.

Results and Discussion

The results on the measurements of excess surface oxygen, acidity and surface area of various samples have been recorded in table 3.1. The plates 3.1, 3.2 and 3.3 represent the change in excess surface oxygen, acidity and surface area with the increasing value of substituents.

From table 3.1 it appears that the values of excess surface oxygen of the different samples of the system $La_{1-x}{}^{Sr}{}_x Co_{1-x} Ti_x O_3$ are greater in comparison to that obtained in case of the different samples of the system $La\ co_{1-x} Ti_x O_3$ and $La_{1-x}{}^{Sr}{}_x Co\ 0_3$. But the values of E.S.O of the different samples of the system $La\ Co_{1-x} Ti\ O_3$ is comparable with the values of excess surface oxygen of the different samples of the system $La_{1-x}{}^{Sr}{}_x Co\ 0_3$. From Table 3.1., it is also clear that the excess surface oxygen has the increasing trend with the rise in the quantity of substituents with the introduction of more and more amount of substituentions of higher valence state are produced in greater concentration and thereby pushing up the oxygen partially to act as excess surface oxygen.

Thus the excess surface oxygen plays the key role enhancing the catalytic activity, the samples of the system $La_{1-x}Sr_xCo_{1-x}Ti_xO_3$ would show the greater activities since these all Possess the high

TABLE - 3.1

SURFACE PROPERTIES OF THE SAMPLES

Sample	Value of X	Excess surface oxygen per 100 gm of the samples X 10^{-2}	Acidity M equivalent/ gm^{-1}	Surface area m^2/gm
$LaCo_{1-x}Ti_xO_3$				
	0.1	16.85	130.50	6.85
	0.2	17.25	135.40	7.25
	0.3	18.42	142.50	8.42
	0.4	20.05	160.65	10.05
	0.5	22.36	162.15	12.36
$La_{1-x}Sr_xCo_{1-x}Ti_xO_3$				
	0.1	25.15	95.40	7.90
	0.2	26.17	100.25	10.20
	0.3	28.04	103.45	11.45
	0.4	29.50	106.82	13.05
	0.5	29.95	110.35	15.25
$La_{1-x}Sr_xCoO_3$				
	0.1	12.25	125.40	5.45
	0.2	13.45	128.80	6.75
	0.3	14.70	130.75	8.25
	0.4	15.85	133.05	9.15
	0.5	17.08	135.35	11.05

magnitude of excess surface oxygen, Consequently it be come imperative to measure the catalytic activity of different samples to ascertain whether the excess surface oxygen adds to the activities.

Results on the acidity measure have been noted in table 3.1. The plate – 3.2 shows the plot of acidity of different sample against the different values of substituents for the system under study. It appears that in case of systems $La_{1-x}Sr_xCo_{1-x}Ti_xO_3$ and $La_{1-x}Sr_xCoO_3$, there are gradual increase in the values of acidity with that

of the increasing values of substituents. However, in case of different samples of the system La Co_{1-x} T_i x O_3 . We find sudden change in the acidity value after x equal to 0.3.

From the parusal of plate. 3.2 and plate 3.1, it becomes obvious that the excess surface oxygen and

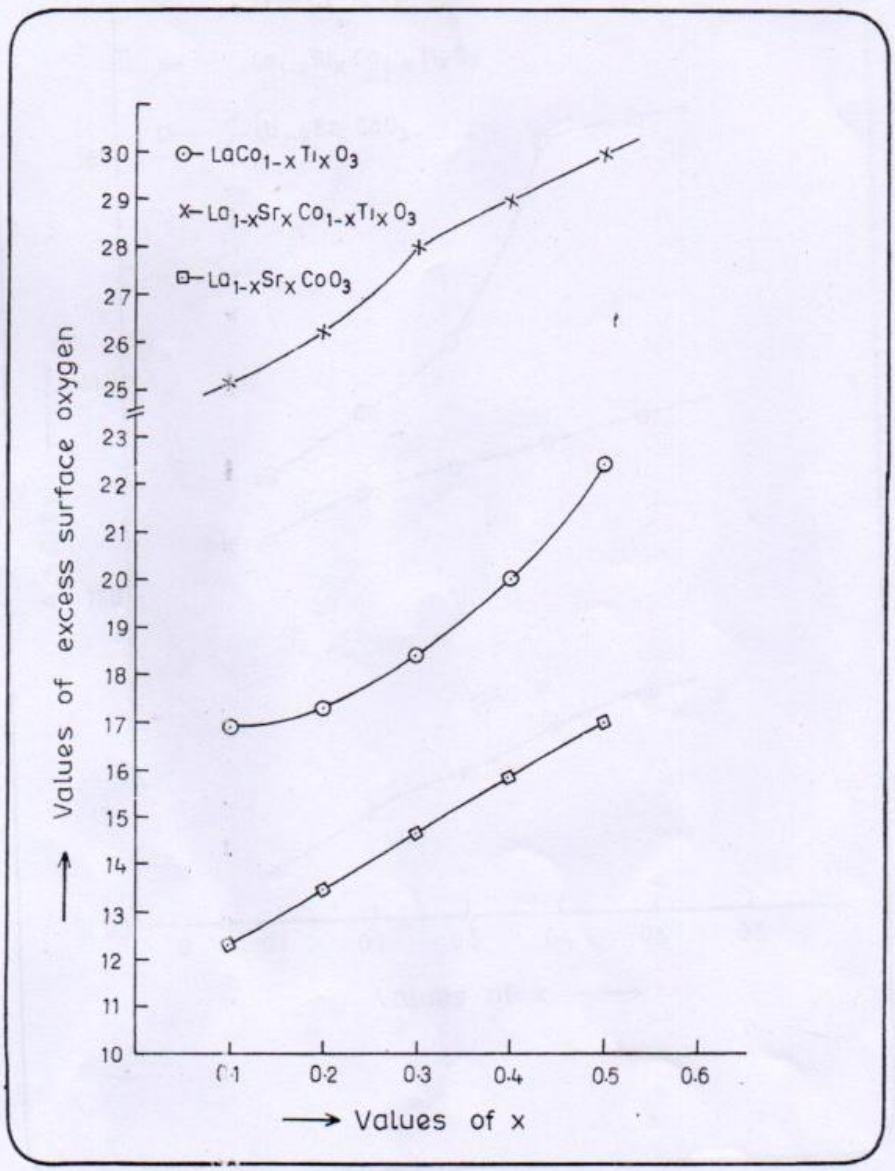

Plate 3.1 Variation of excess surface oxygen with amount of substitute.

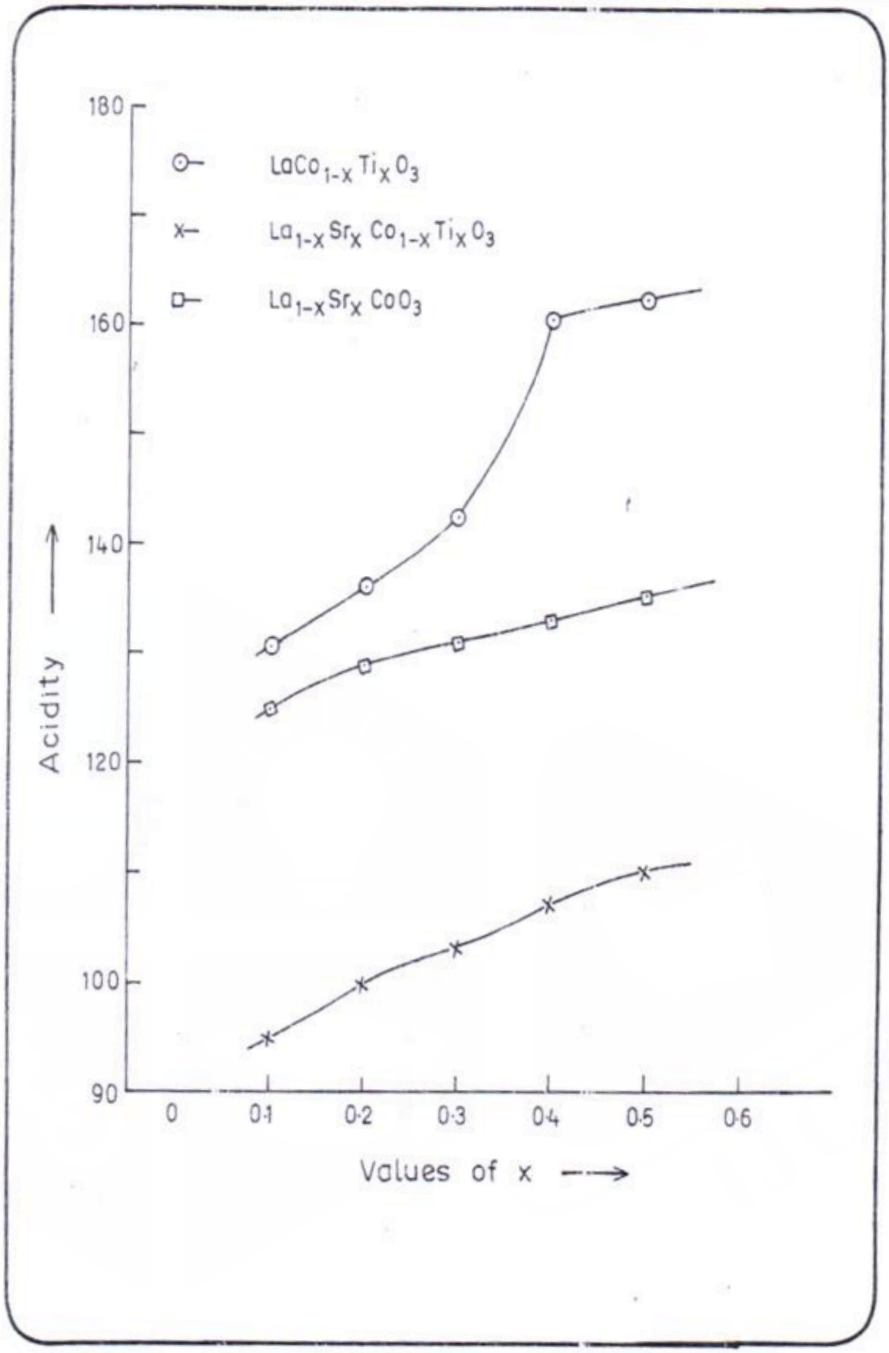

Plate 3.2 Variation of acidity with the amount of substitute.

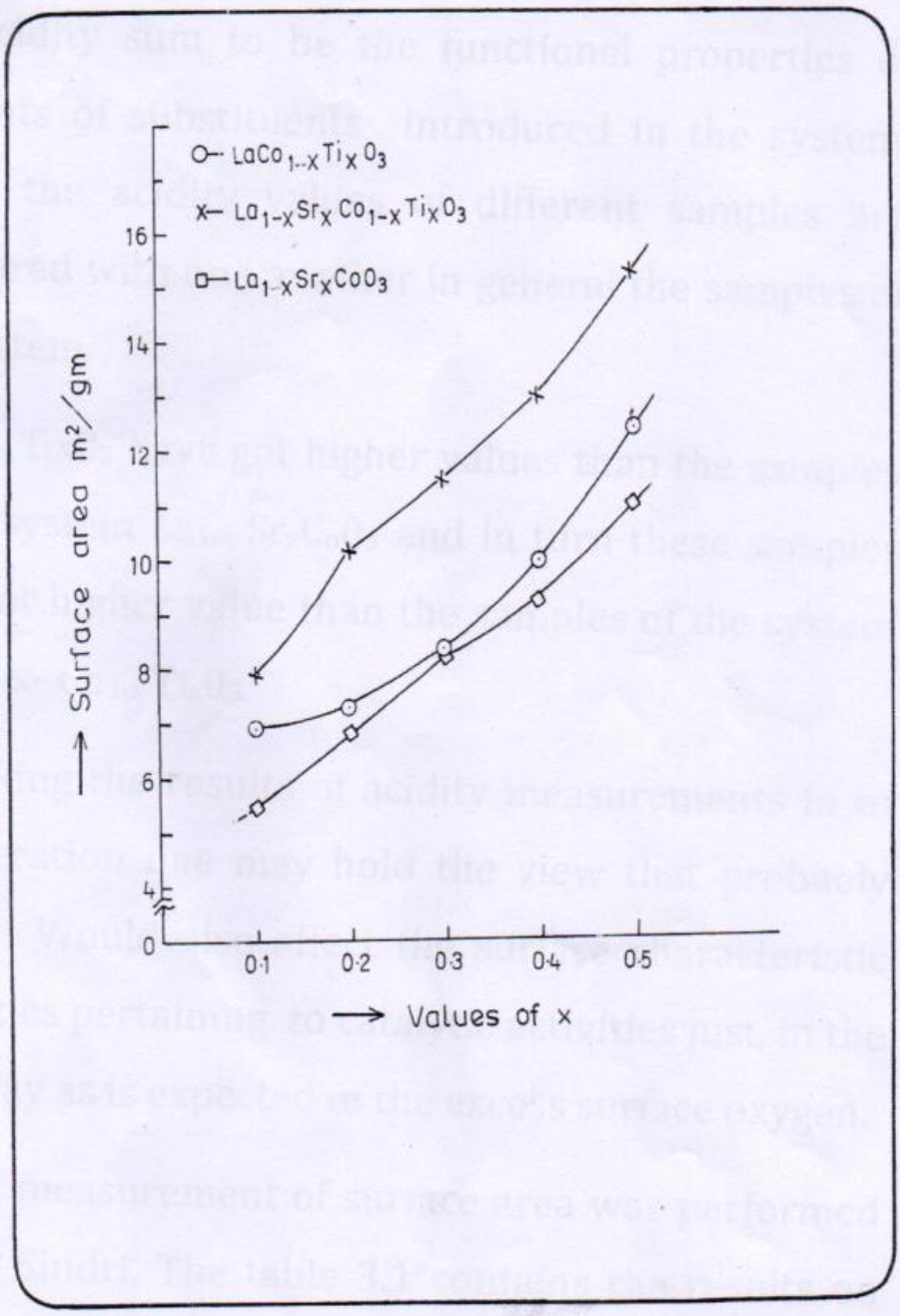

Plate 3.3 Variation of surface area with the amount of substitute.

the acidity sum to be the functional properties of amounts of substituents introduced in the system. When the acidity values of different samples are compared with one another in general the samples of the system.

$LaCo_{1-x}Ti_xO_3$ have got higher values than the samples of the system $La_{1-x}Sr_xC_oO_3$ and in turn these samples have got higher value than the samples of the system $La_{1-x}Sr_x$- $Co_{1-x}Ti_xO_3$.

Taking the results of acidity measurements in to consideration one may hold the view that probably acidity . Would also affect the surface characteristic properties pertaining to catalytic activities just, in the same way as is expected of the excess surface oxygen.

The measurement of surface area was performed in PDIL Sindri. The table 3.1 contains the results on the measurement of surface area of the different samples. Plate 3.3 depicts the variation of surface area with the increasing amount of substituents introduced. It is worth while mention that all the samples particularly with small amount of substituents possess very simple surface area resulting in the adsorption of very small volume of nitrogen. The data presented in table 3.1 are the average of two measurements made for each sample. From the plots of surface area versus the amount of substituents it is interesting to note that the values of

surface area of the different samples of the system $La_{1-x} Sr_x Co_{1-x} Ti_x O_3$ are a bit high in comparision to the surface area values of the samples of the systems $La Co_{1-x} Ti_x O_3$ and $La_{1-x} Sr_x Co O_3$ indicating the samples of the system $La Co_{1-x} Ti_x O_3$ to be catalytically more active. Infect the surface area value is definitely the deciding factor for the degree of catalytic performance. It is also notable that all the samples of the two system $La Co_{1-x} Ti_x O_3$ and $La_{1-x} Sr_x Co O_3$ seem to possess almost the same values of surface area. In view of the trend observed in between surface area and the amount of substituents it is predictable that the samples with the higher values of substituents would be associated with high activity and there would be a gradual form in the activity as the amount of substituents would form.

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