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Synthesis And Charecterization of Magnesium Aluminate Spinel By Solution Combustion Method Using Variation of Fuel to Oxidiser Ratio and Mixed Fuels

KEYWORDS: solution combustin synthesis,MgAl2O4 Spinel ,XRay-diffraction,SEM.

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ABSTRACT *This paper is a study of Nano sized MgAl2O4 (Magnesium Alumminate) spinel powders which were synthesized by muffle furnace assisted Solution Com -bustion Synthesisusing various mixtures of urea, glycine as fuel. It was seen that Mg(NO3)2.6H2O and Al(NO3)3.9H2O show different behavior with respect to urea and glycine . Also the variation of fuel to oxidizer (F/O) ratio was studied . The study was carried out for leaner to richer fuel ratios.In the case of MgAl2O4 combustion synthesis results were achieved when fuel mixture (urea and glycine) were used. The useof fuel mixtures allowed the formation of pure, nano-crystalline MgAl2O4 directly from the combustion reaction.The effect of fuel mixture ratios was investigated by variation of the ratios in which the fuel was taken . The later product was characterized by XRay-diffraction analysis .The nanoscaled images where taken by SEM (Scanning Electron Microscope) of the powder being produced.*

Introduction

Magnesium aluminate spinel ($MgAl_2O_4$) is one of the widely used materials[1-3].various method like sol-gel, co-precipitation etc have being used to produce oxides In the MgO–Al_2O_3 system the only compound that forms as a result of the reaction between the two oxides is $MgAl_2O_4$, called spinel. Magnesium Aluminate has a low density (3.58 g/cm3) . Due to its significant properties – high melting point, 2135 °C, low thermal conductivity, low thermal expansion coefficient, high mechanical strength both at room temperature and elevated temperature, good thermal shock resistance and good chemical insipidness [1,4]. This is the result that all these properties of $MgAl_2O_4$ are successfully applied in various applications, like refractory bricks manufacturing for the cement and steel industry transparent windows , good resistance against chemicals and better strength at extreme high temperatures , military applications (armour materials, domes), humidity sensors, catalysts support, dentistry and nuclear technique [1]. The synthesis of $MgAl_2O_4$ with specific characteristics such as chemical homogeneity, high purity, low particle size and uniform size distribution depends considerably on preparation methods. As such, magnesium aluminate has been synthesized by various methods such as sol–gel , solid state, spray drying , co-precipitation , and freeze-drying. However, most of these methods are either complex or expensive which lower preparation of the nano-sized materials in a large scale as compared to the combustion or sol–gel synthesis [4-10]. Moreover, other disadvantages are the necessity of high temperature, inhomogeneity, and low surface area of the nano-sized products [4]. In this work, Here we have used the fuel to oxidizer variation for formation of Magnesium Aluminate ($MgAl_2O_4$) powder. In addition, fuel mixtures were also used to prepare $MgAl_2O_4$ and other oxide powders, e.g. $MgAl_2O_4$ from urea and glycine . Mixtures of urea and glycine were worked as fuel to synthesize nano scale $MgAl_2O_4$ powders via a solution combustion process. Moreover, the effects of urea and glycine addition on characteristics (e.g. particles size and specific surface area) of the powders were synthesized [2] . The further characterization was carried out by XRay-diffraction and SEM examinations.

Experimental procedure

A. Solution Combustion Synthesis (SCS) of Magnesium Aluminate ($MgAl_2O_4$) with Various Fuel to Oxidisers Ratios:

Analytical grade Aluminum Nitrate nonahydrate ($Al(NO_3)_3{\cdot}9H_2O$), urea ($CO(NH_2)_2$), Glycine (NH_2CH_2COOH) were used as oxidizer and fuel respectively for SCS. These reactants are mixed in the required molar ratios with required amount as shown in Table.1 in a minimum volume of deionized water to obtain clear aqueous solutions. The solution is then kept in furnace preheated to 500°C. First thermal dehydration (at 100°C) takes place forming viscous solution, at 200°C the viscous liquids swelled and gets auto ignited, with the rapid evolution of a large volume of gases to produce voluminous powders. The nature of ignition depended on the fuel-to-oxidant ratio. The auto ignition of the precursor containing fuel-to-oxidant ratio according to the concept of propellant chemistry and fuel-rich ratio was found to be more violent compared to the fuel-deficient precursor. Because the time for which the auto ignition exists is rather small (typically 5 s), Under the equilibrium conditions the standard reaction equation in this systems can be represented for all trivalent nitrates - urea combustion (Where M=trivalent cation) for different fuel to oxidizer ratio as

For all Magnesium nitrate, Aluminium nitrate, urea, glycine combustion for different F/O ratios equation can be written as.

$$Mg(NO_3)_2.6H_2O + 2Al(NO_3)_3.9H_2O + 6.66ø\ NH_2CONH_2 \rightarrow MgAl_2O+(4+6.66ø)N_2+6.66øCO_2+(24+13.32ø)H_2O+$$

$$(10-10ø)O_2 \qquad (1)$$

$$Mg(NO_3)_2.6H_2O+2Al(NO_3)_3.9H_2O+4.44øNH_2CH2COOH \rightarrow MgAl_2O_4+(4+2.22ø)N_2+8.88øCO_2+(24+11.1ø)H_2O+$$

$$(10-10ø)O_2 \qquad (2)$$

Here the Ø is the fuel to oxidizer equivalence ratio (not the urea to nitrate molar ratio) given by the formula

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$$\frac{Fuel}{oxidiser}Ratio = \frac{\sum all\ oxidising\ and\ redusing\ elements\ in\ fuel}{\sum all\ oxidising\ and\ redusing\ elements\ in\ oxidizer}$$

B.Solution Combustion Synthesis of $MgAl_2O_4$ BY MIXEDFUELS

$MgAl_2O_4$ synthesis has been considered to study the effect of nature of fuel, decomposition temperature difference of fuel and oxidizer, furnace temperature (in case of multi cationic nitrate fuel system) on SCS powders. The $MgAl_2O_4$ was prepared by using Magnesium nitrate, Aluminium nitrate, as oxidizers and urea, glycine as fuels and coded as in Table 6. Effect of mixed fuel also was studied by using reactants from Table 6. The reactants are mixed in distilled water in molar ratio and mass as shown in Table 6. These solutions, then kept in a furnace pre heated to 500°C. The combustion reaction characteristics are shown in table. Under the equilibrium conditions the standard reaction equation in these fuels- nitrate systems for preparation of $MgAl_2O_4$ by MIXED fuels can be represented as below

$Mg(NO_3)_2.6H_2O + 2Al(NO_3)_3.9H_2O + 6.6NH_2CONH_2 \rightarrow MgAl_2O_4 + 6.6CO_2 + 28.2H_2O + 10.6N_2$

$Mg(NO_3)_2.6H_2O + 2Al(NO_3)_3.9H_2O + 4.4NH_2CH_2COOH \rightarrow MgAl2O4 + 8.8CO_2 + 26H_2O + 6.2N_2$

$Mg(NO_3)_2.6H_2O + 2Al(NO_3)_3.9H_2O + 5NH_2CONH_2 + 1.11NH_2CH_2COOH$

$\rightarrow MgAl_2O_4 + 7.22CO_2 + 27.51H_2O + 9.55N_2$

$Mg(NO_3)_2.6H_2O + 2Al(NO_3)_3.9H_2O + 3NH_2CONH_2 + 2.44NH2CH2COOH$

$\rightarrow MgAl_2O_4 + 7.88CO_2 + 27.88H_2O + 8.22N_2$

Structural, Compositional and Micro-structural Analysis.
X-ray diffraction (XRD)
X-ray diffraction studies were carried out for phase confirmation and for calculating crystallite size of the milled samples, using D8-Advance-Bruker machine with Cu-K_α (wavelength of Cu-K_α (λ) ~1.5406 Å) radiations for all the measurements. Ni filter was used to attenuate K_β lines. The crystallite size of powders was calculated using Scherrer's formula where an assumption was made that the particle is spherical in shape The Scherrer's formula gives

$$\beta\theta = \frac{k'\lambda}{d.COS\theta} \qquad (3)$$

where, β_θ is the full width at half maximum (FWHM) of diffracted peaks in degrees, L stands for the liner dimension of particles in meters, θ refers to Bragg's angle in degrees and K' is the shape factor, generally known as a numerical constant and evaluated as 0.93 and depends on shape of crystallites. From this expression, it is clearly seen that diffracted beam gets broadened as the size of crystallite reduces. Silicon was used as an external standard for correction due to instrumental broadening.

Scanning electron microscope (SEM)
The surface morphology of powders and pellet samples was studied using scanning electron microscope a Cambridge, stereoscan-240. (ESEM –FEI, Quanta 200) and elemental compositional analysis was done using the attached energy dispersed analysis of X-rays (EDAX). The grain size of the chosen materials was seen clearly using FEG-SEM .All the samples studied were ceramic oxide insulator materials (non conductors).Grain size was estimated using linear intercept method.

Thermodynamic Modeling
For comparing exothermicity of different fuels in reaction with the nitrates, enthalpy and adiabatic flame temperature of the reactions between urea and aluminum nitrate were calculated in the conditions:- Stoichiometric, Fuel lean and Fuel rich. Using the thermodynamic data for the various reactants and products listed in Table 2.2, the enthalpy of combustion and the theoretical adiabatic flame temperatures as a function of F/O ratio were approximately calculated by the following equations: using Mathematica software

$$\Delta H^\circ = (\sum n\Delta H^\circ)_{products} - (\sum n\Delta H^\circ)_{reactant} \qquad (4)$$

Here ΔH° is the enthalpy of combustion reaction, Cp is the heat capacity of products.

Table1.Relevant thermodynamics data of reactants and products

Compound	ΔH_f (kcal. mol^{-1})	c_p (cal.$mol^{-1}.K^{-1}$)
$MgAl_2O_4$	−547.38	-
$Al(NO_3)_3$· ©	-857.59	-
$Zn(NO_3)_2 \cdot 6H_2O$ ©	-551.30	-
NH_2CONH_2 ©	-79.71	-
NH_2CH_2COOH ©	-126.22	-
CH_3CHNH_2COOH ©	-100.26	-
Ca $(NO_3)_3 \cdot 4H_2O$ ©	-509.64	-
Mg $(NO_3)_3 \cdot 6H_2O$ ©	-624.59	-
SiO_2	-217.75	-
$CaMgSi_2O_6$ ©	28.8	-
NH_4NO_3	-87.37	-
Al_2O_3 ©	−399.09	52.3+ 0.00774T 33.3 22.08 + 0.0089T
ZnO ©	-83.24	11.40 + 0.00145*T* – 182400/*T*²
CO_2 (g)	-94.051	10.34 + 0.00274*T* – 195500/*T*²
NO_2 (g)	7.93	8.8
N_2 (g)	0	6.50 + 0.0010*T*
H_2O (g)	-57.796	8.22 + 0.00015*T* + 0.00000134*T*²
H_2O (l)	-68.38	-
O_2 (g)	0	8.27 + 0.000258*T* – 18770/*T*²
©:crystalline; (g):gas; (l):liquid; (*T*):absolute temperature;		

RESULTS AND DISCUSSIONS
Solution Combustion Synthesis (SCS) of Magnesium Aluminate ($MgAl_2O_4$) with Various F/O Ratios:

Considering the reaction of nitrates with the urea fuel the

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obtained results are ,

For fuel lean Ø<1, three compositions of fuel lean are Ø=0.4,0.8 and chemical equations are respectively (60%,20% fuel lean)

Ø=0.4,fuel lean

$Mg(NO3)2.6H2O+2Al\ (NO_3)_3.9H_2O + 2.66NH_2CONH_2$

→ $MgAl_2O_4+5N_2+2.66CO_2+29.32H_2O+6O_2$

Ø=0.8,fuel lean

$Mg(NO_3)_2.6H_2O + 2Al\ (NO_3)_3.9H_2O + 5.33NH_2CONH_2$

→$MgAl_2O_4+9.33N_2+5.33CO_2+34.66H_2O+2O_2$

Fuel stoichiometric Ø=1

$Mg(NO_3)_2.6H_20 + 2Al\ (NO_3)_3.9H_2O + 6.66NH_2CONH_2$

→ $MgAl_2O_4+10.66N_2+6.66CO_2+37.32H_2O$

Fuel rich Ø>1 Ø =1.2, 1.4, 1.6 (20%, 40%, 60% fuel rich) reaction equations are

$Mg(NO_3)_2.6H_20 + 2Al\ (NO_3)_3.9H_2O + 8NH_2CONH_2$

→$MgAl_2O_4+12\ N_2+8\ CO_2+40H_2O+2O_2$

$Mg(NO_3)_2.6H_20 + 2Al\ (NO_3)_3.9H_2O + 9.33NH_2CONH_2$

→ $MgAl_2O_4+13.33N_2+9.33CO_2+42.66H_2O+4O_2$

$Mg(NO_3)_2.6H_20 + 2Al\ (NO_3)_3.9H_2O + 10.66NH_2CONH_2$

→$MgAl_2O_4+14.6N_2+10.66CO_2+45.32\ H_2O+6O_2$

The above equations are for complete combustion equations formed by assuming extra oxygen needed in fuel rich case is supplied by atmospheric air.

Table 2 . The required molar ratios with required amount of F/O ratios as shown below for Urea as fuel.

Fuel to oxidizer ratio	Urea-nitrate molar ratio	$Mg(NO_3)_2 6H_2O$ (g)	$Al(NO_3)_3.9H_2O$ (g)	Urea $CO(NH_2)_2$ (g)	Rect-ion type	Colour of powder
0.4	1	5.12	15	3.2	No flame	white
0.8	1.5	5.12	15	6.4	No flame	white
1.0	2.5	5.12	15	8	flame	white
1.2	3.0	5.12	15	9.6	flame	white
1.4	3.5	5.12	15	11.20	flame	white
1.6	4	5.12	15	12.8	flame	white

From equation (1) for Magnesium nitrate , Aluminium nitrate ,urea combustion can be written in terms of Fuel-oxidizer ratio (ø) as

$Mg(NO_3)_2.6H_2O + 2Al(NO_3)_3.9H_2O + 6.66ø\ NH_2CONH_2$ → $MgAl_2O+(4+6.66ø)N_2+6.66øCO_2+(24+13.32ø)H_2O+(10-10ø)O_2$

$\Delta H_{(Products)} = \Delta H_{(MgAl2O4)} + (4+6.66ø) * \Delta H_{(N2)} + 6.66ø * \Delta H_{(CO2)} +$

$(24+13.32ø) * \Delta H_{(H2O)} + (10-10\ ø)) * \Delta H_{(O2)}$

$\Delta H_{(Products)} = -547.38+(4+6.66ø)*0+6.66ø*(-94.051)+(24+13.32ø)*(-57.796)+(10-10\ ø)*0$

$= -547.38-626.37\ ø-1387.104-769.84\ ø$

$\Delta H_{(Products)} = -1934.484-1396.21\ ø$

$\Delta H_{(Reactants)} = \Delta H_{(Mg(NO3)2.6H2O)} + 2* \Delta H_{(Al\ (NO3)3.9H2O)} + 6.66* \Delta H_{(NH2CONH2)}$

$\Delta H_{(Reactants)} = -624.59 + 2*(-857.59) + 6$

$\Delta H_{(Reaction)} = (-1934.484-1396.21\ ø)-(-2339.77-530.8686\ ø) + 6.66\ ø\ *(-79.71)$

$\Delta H_{(Reactants)} = -2339.77-530.8686\ ø$

$\Delta H_{(Reaction)} = \Delta H_{(Products)} - \Delta H_{(Reactants)}$ (from equation (4))

$\Delta H_{(Reaction)} = 405.286-865.34\ ø$ (5)

By considering the average Cp values of combustion products from

$Cp_{(Products)} = Cp_{(MgAl2O4)} + (4+6.66ø) * Cp_{(N2)} + 6.66ø * Cp_{(CO2)} + (24+13.32ø) * Cp_{(H2O)} + (10-10\ ø) * Cp_{(O2)}$

$Cp_{(Products)} = 42.79+(4+6.66ø)\ x6.961+6.66ø\ x8.87+(24+13.32ø)\ x8.025+(10-10\ ø)x7.01$

$Cp = 440.227+35.33ø$

No of moles of gases/mole product=38+16.64ø (6)

Similarly for glycine as fuel ,the variation of fuel to oxidizer ratio was carried out and the results were obtained as follows

For fuel lean Ø<1, three compositions of fuel lean are Ø=0.4,0.8 and chemical equations are respectively (60%,20% fuel lean)

Ø=0.4,fuel lean

$Mg(NO3)2.6H2O+2Al\ (NO_3)_3.9H_2O + 1.77NH_2CH_2COOH$

→ $MgAl_2O_4+4.88N_2+3.54CO_2+28.42H_2O+6O_2$

Ø=0.8,fuel lean

$Mg(NO_3)_2.6H_2O + 2Al\ (NO_3)_3.9H_2O + 3.55NH_2CH_2COOH$

→$MgAl_2O_4+5.77N_2+7.1CO_2+32.87H_2O+2O_2$

Fuel stoichiometric Ø=1

$Mg(NO_3)_2.6H_20 + 2Al\ (NO_3)_3.9H_2O + 4.44NH_2CH_2COOH$

→ $MgAl_2O_4+6.22N_2+8.88CO_2+35H_2O$

Fuel rich Ø>1 Ø =1.2, 1.4, 1.6 (20%, 40%, 60% fuel rich) reaction equations are,

$Mg(NO_3)_2.6H_20 + 2Al\ (NO_3)_3.9H_2O + 5.33NH_2CH_2COOH$

→$MgAl_2O_4+6.66N_2+10.66CO_2+37.32H_2O+2O_2$

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$Mg(NO_3)_2.6H_20 + 2Al(NO_3)_3.9H_2O + 6.22NH_2CH_2COOH \rightarrow MgAl_2O_4 + 7.11N_2 + 12.44CO_2 + 39.55H_2O + 4O_2$

$Mg(NO_3)_2.6H_20 + 2Al(NO_3)_3.9H_2O + 7.11NH_2CH_2COOH \rightarrow MgAl_2O_4 + 7.55N_2 + 14.22CO_2 + 41.77H_2O + 6O_2$

Table 3 . The required molar ratios with required amount of F/O ratios as shown below for Glycine as fuel .

Fuel to oxidizer ratio	Urea-nitrate molar ratio	Mg $(NO_3)_2$. $6H_2O$ (g)	Al $(NO_3)_3$.$9H_2O$ (g)	Glycine NH_2CH_2COOH (g)	Rection type	Colour of powder
0.4	1	5.12	15	2.66	No flame	White
0.8	1.5	5.12	15	5.32	No flame	Brown-ish white
1.0	2.5	5.12	15	6.66	flame	Grey white
1.2	3.0	5.12	15	7.98	flame	grey
1.4	3.5	5.12	15	9.30	flame	Dark grey
1.6	4	5.12	15	10.64	flame	Black

Standard equation for Magnesium nitrate , Aluminium nitrate ,glycine combustion can be written in terms of Fuel-oxidizer ratio (ø) as

$Mg(NO_3)_2.6H_2O + 2Al(NO_3)_3.9H_2O + 4.44\ ø NH_2CH_2COOH \rightarrow MgAl_2O_4 + (4+2.22ø)N_2 + 8.88ø CO_2 + (24+11.1ø)H_20 + (10-10ø)O_2$

$\Delta H_{(Products)} = \Delta H_{(MgAl2O4)} + (4+2.22ø) * \Delta H_{(N2)} + 8.88ø * \Delta H_{(CO2)} + (24+11.1ø) * \Delta H_{(H2O)} + (10-10ø) * \Delta H_{(O2)}$

$\Delta H_{(Products)} = -547.38 + (4+2.22ø) * 0 + 8.88ø * (-94.051) + (24+11.1ø) * (-57.796) + (10-10ø) * 0 = -547.38 + (-470.255)ø + (-577.96ø) + (-1040.328)$

$\Delta H_{(Products)} = -1934.484 - 1476.7ø$

$\Delta H_{(Reactants)} = \Delta H_{(Mg(NO3)2.6H2O)} + 2 * \Delta H_{(Al(NO3)3.9H2O)} + 4.44 * \Delta H_{(NH2CH2COOH)}$

$\Delta H_{(Reactants)} = -624.59 + 2*(-857.59) + 4.44ø * (-126.22)$

$\Delta H_{(Reactants)} = -2339.77 - 560.41ø$

$\Delta H_{(Reaction)} = \Delta H_{(Products)} - \Delta H_{(Reactants)}$

$\Delta H_{(Reaction)} = (-1934.484 - 1476.7ø) - (-2339.77 - 560.41ø)$

$\Delta H_{(Reaction)} = 405.28 - 916.29ø$ (7)

By considering the average Cp values of combustion products from

$Cp_{(Products)} = Cp_{(MgAl2O4)} + (4+2.22ø) * Cp_{(N2)} + 8.88ø * Cp_{(CO2)} + (24+11.1ø) * Cp_{(H2O)} + (10-10ø) * Cp_{(O2)}$

$Cp_{(Products)} = 42.79 + (4+2.22ø) \times 6.961 + 8.88ø \times 8.87 + (24+11.1ø) \times 8.025 + (10-10ø) \times 7.01$

$Cp = 333.244 + 113.27ø$

$\Delta Cp = 333.244 + 113.27ø - 98.83ø = 333.244 - 14.44ø$

No of moles of gases/mole product $= 28 + 22.21ø$ (8)

The adiabatic flame temperature was calculate using formula

$T_{ad} = To + (\Delta Hp - \Delta Hr) / C_p$ (9)

Table 4. Effect of Urea – Nitrate F/O ratio on adiabatic flame temperature, enthalpy of reaction, no of moles of gases evolved

Fuel/oxidiser	Enthalpy(ΔH) Kcal/mol	Adiabatic flame temp (T_{ad}) °C	No gases evolved
0.4	59.065	467.71	44.65
0.8	-287.25	961.80	51.31
1.0	-460.06	1266.26	54.64
1.2	-634.1	1521.61	57.96
1.4	-806.98	1737.31	61.29
1.6	-979.79	1922.63	64.62

Table 5. Effect of Glycine– Nitrate F/O ratio on adiabatic flame temperature, enthalpy of reaction, number of moles of gases evolved

Fuel/oxidiser	Enthalpy(ΔH) Kcal/mol	Adiabatic flame temp (T_{ad}) °C	No gases evolved
0.4	38.21	411.55	36.88
0.8	-262.95	1089.23	45.76
1.0	-458.37	1360.45	50.21
1.2	-690	1727.57	54.65
1.4	-879.34	1989.78	59.09
1.6	-1060.07	2203.16	63.53

B.Solution Combustion Synthesis (SCS) of Magnesium Aluminate ($MgAl_2O_4$) with MIXED FUELS:

The $MgAl_2O_4$ was prepared by using Magnesium nitrate, Aluminium nitrate, as oxidizers and urea, glycine as fuels. Effect of mixed fuel also was studied by using variation of fuel mixture as in Table.6. The fuels (reactants) are mixed in deionized water in molar ratio and mass as shown in Table. 6. These solutions, then kept in a furnace pre heated to 500°C. The combustion reaction characteristics are shown in Table.6. Under the equilibrium conditions the standard reaction equation in these fuels- nitrate systems for preparation of $MgAl_2O_4$ by mixed fuels can be represented as below

$Mg(NO_3)_2.6H_2O + 2Al(NO_3)_3.9H_2O + 6.6NH_2CONH_2 \rightarrow MgAl_2O_4 + 6.6CO_2 + 28.2H_2O + 10.6N_2$

$Mg(NO_3)_2.6H_2O + 2Al(NO_3)_3.9H_2O + 4.4NH_2CH_2COOH \rightarrow MgAl2O4 + 8.8CO_2 + 26H_2O + 6.2N_2$

$Mg(NO_3)_2.6H_2O + 2Al(NO_3)_3.9H_2O + 5NH_2CONH_2 + 1.11NH_2CH_2COOH \rightarrow MgAl_2O_4 + 7.22CO_2 + 27.51H_2O + 9.55N_2$

$Mg(NO_3)_2.6H_2O + 2Al(NO_3)_3.9H_2O + 3NH_2CONH_2 + 2.44NH2CH2COOH \rightarrow MgAl_2O_4 + 7.88CO_2 + 27.88H_2O + 8.22N_2$

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Table 6.Amount of fuel and oxidizer, used for the combustion reaction to produce $MgAl_2O_4$ by mixed fuel approach.

	Sample code	Mg (NO3)2 (g)	Al (NO3)3 (g)	Fuel Urea (g)	Fuel glycine (g)	Obtained Powder (g)
A	Mg-5U+1.11G	5.12	15	6	1.66	2.89
B	Mg-3U+2.44G	5.12	15	3.6	3.66	2.76

CHARECTERIZATION RESULTS

X –ray characterization :

Figure 9 shows XRD pattern of the synthesized powder for Mixture of fuels . It shows the formation of pure crystalline $MgAl_2O_4$ phase confirmed by comparing the peaks with standard peaks of (JCPDS card number 75-1796) with little impurity peak of Al_2O_3 (JCPDS card no 82-1468) at 2Θ value of 43.35°. The formation of phase pure crystalline magnesium aluminate is confirmed by comparing the peaks with standard peaks of JCPDS card number for the mixed fuels.The percentage of magnesium aluminate phase increases as deviation goes from stoichiometric to fuel rich and fuel lean. It was also observed that there was a formation of impurities in case of lean mixtures .For fuel rich region even though the enthalpies of reactions are higher than that of fuel lean but the amount of gases produced in the reactions are more than the stoichiometric composition so those gases take the more amount energy. It was noted in Figure 10 that the formation of Mg_3N_4 and MgC_3(JCPDS card no 47-1456) takes place at lower temperature due to incomplete combustion. Figure.9 shows XRD pattern of the as synthesized $MgAl_2O_4$ powder formed by using mixed fuels of urea and glycine maintaining stoichiometric conditionThe furnace temperature maintained was 500°C. But here when taken with aluminium nitrate and using urea and glycine mixed fuel could able to form the Magnesiunm Aluminate. The crystallite size(d) was calculated using Scherer formula (3)

Table 7 . Representing the crystallite size in nano meters

Sr.no	β_θ =full width at half maximum (FWHM)	θ	Cos θ	d (nm)
1	0.334	9.62	0.98	24.3
2	0.246	15.78	0.96	33.4
3	0.250	18.57	0.94	33.5
4	0.266	22.55	0.92	32.2
5	0.294	29.83	0.86	31.2
6	0.30	32.76	0.84	31.1

Scanning Electron Microscope (SEM):

Figure 10 , 11 and 12 show the SEM images of $MgAl_2O_4$ of porous morphology with sphere like agglomerates ranging from 10 to100 nm. The agglomerates are porous due to escaped number of moles of gases released in combustion reaction . Some particles are sintered to form big hard aggregates of 1-5µm. Some isolated 100nm seized particles are also seen in $MgAl_2O_4$ powder. The formation of $MgAl_2O_4$ when used with mixture of fules shows that temperature given was enough to help the formation of the spinel powder. Associated gas evolution results in highly porous structure as the amount of gas increases agglomerates are more likely to break and form the porous structure as seen in the images.

FIGURES AND GRAPHS

Figure 1.Muffle Furnace setup.

Figure 2. Clear aqueous solution of fuel and oxidizer mixture in solution combustion synthesis used.

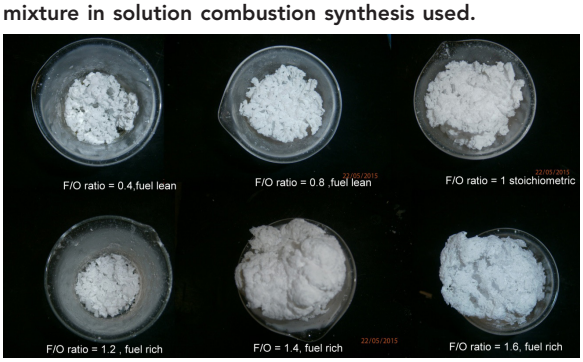

Figure 3. Different powders formed after scs with fuel to oxidizer (F/O) variation when urea was used as fuel.

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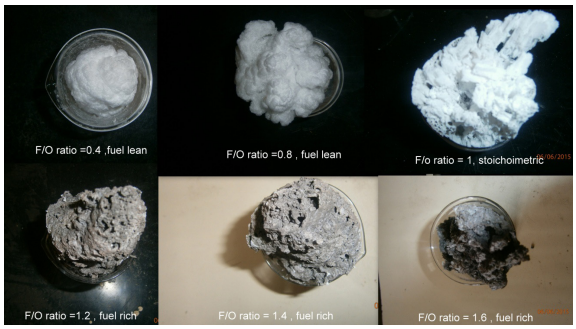
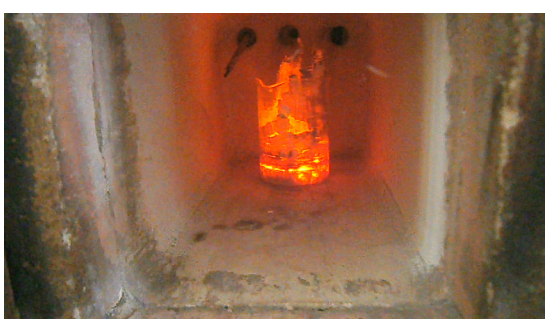


Figure 4 . Different powders formed after scs with fuel to oxidizer (F/O) variation when glycine was used as fuel.



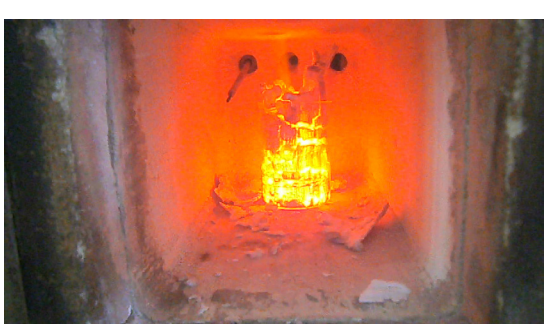


Figure 5. Phases of powder formation during Solution Sombustion Synthesis.

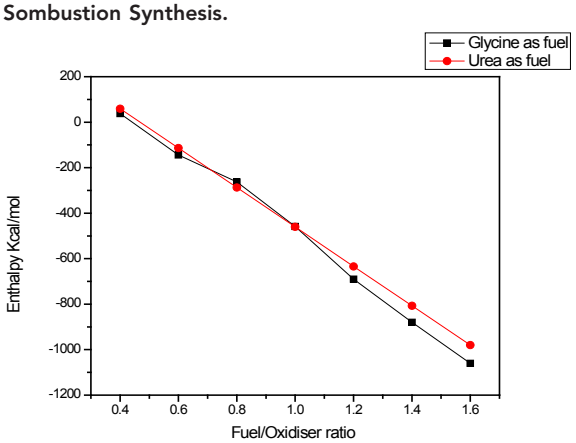


Figure 6. Graph representing the enthalpy vs F/O ratio for

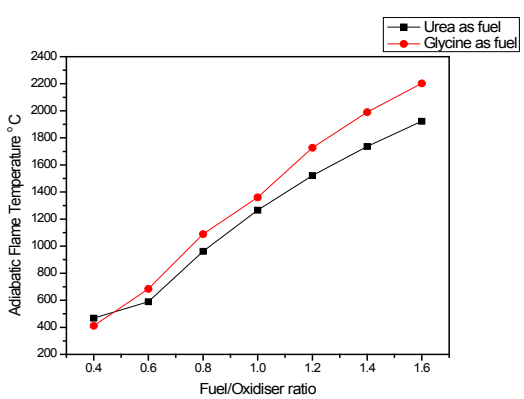


Figure 7. : Graph of fuel /oxidizer ratio on measured flame temperature.

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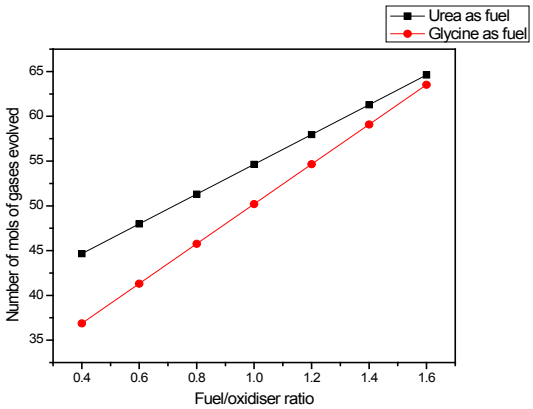

Figure 8. Graph of number of moles of gases released as a function of fuel to oxidizer ratio .

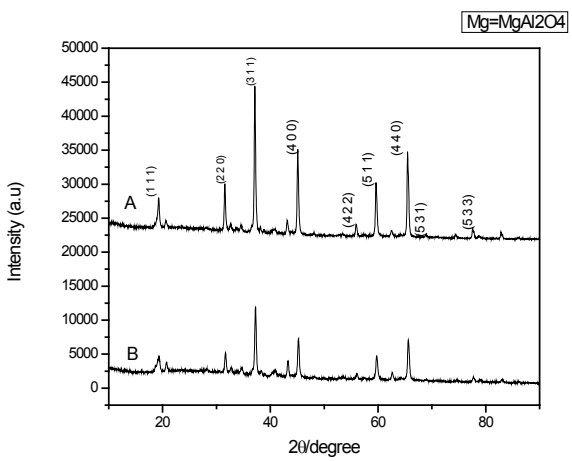

Figure 9. XRD pattern of Magnesium Aluminate powders synthesized by solution combustion synthesis by mixed fuels

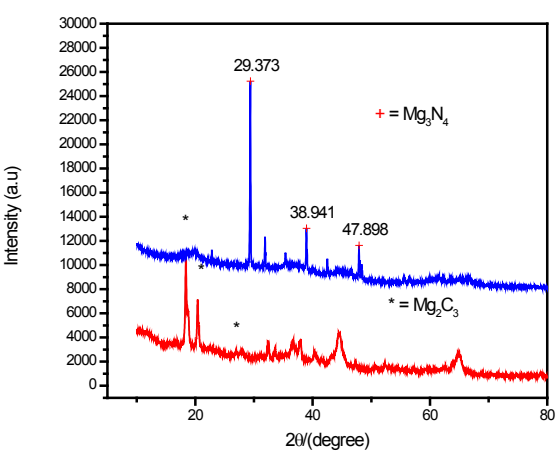

Figure 10. . XRD pattern of powders at lean fuel by solution combustion synthesis

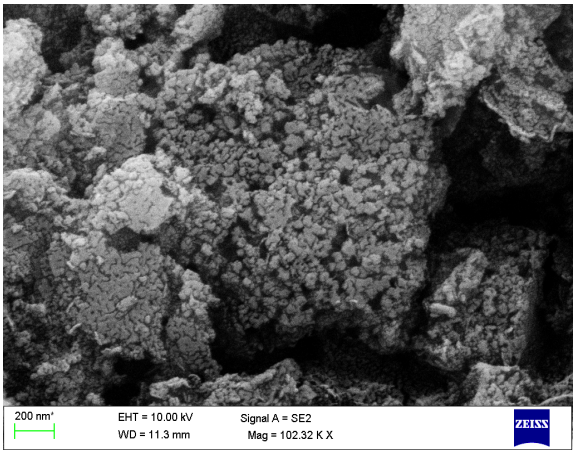

Figure10. SEM image showing nano $MgAl_2O_4$ Spinels

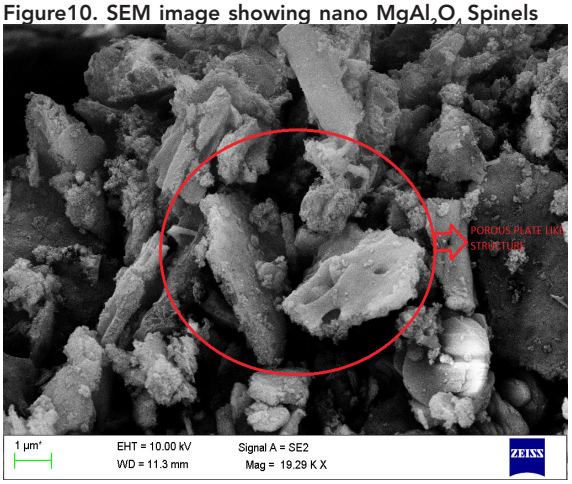

Figure 11. SEM image showing Porousity developed due to escaped gases at fuel rich in variation of F/O ratio.

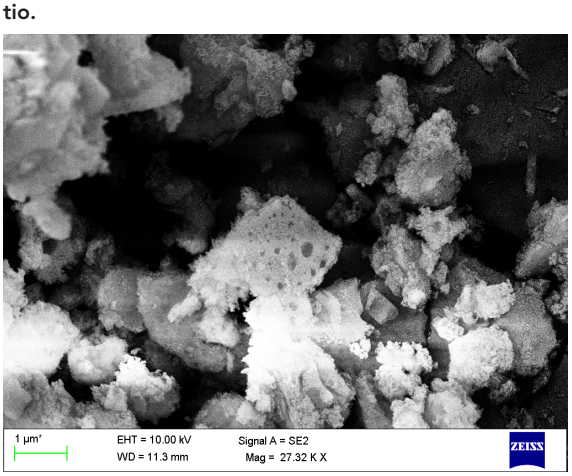

Figure 12. SEM image showing agglomerates of $MgAl_2O_4$ powder.

v.conclusions

This paper is a study on solution combustion synthesis by variation of fuel to oxidizer ratio and by mixed fuels . The end results are promising and the characterization results prove that the formation of Magnesium Aluminate can be carried out at low temperatures (500 °C) using two or more fuels together . More precisely it is a low cost and newer method of producing oxides .

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REFERENCE
[1]Jiahai Bai, Juncheng Liu, ChengfengLi,GuochangLi,Qingyang Du, Mixture of fuels approach for solution combustion synthesis of nanoscale MgAl2O4 powders , Advanced Powder Technology 22 (2011) 72–76 | | [2]F. Tavangarian, R. Emadi, Synthesis and characterization of pure nanocrystalline magnesium aluminatespinel powder. Journal of Alloys and Compounds 489 (2010) 600–604. | | [3]Mostafa Y. Nassar, Ibrahim S. Ahmed, Ihab Samir , A novel synthetic route for magnesium aluminate (MgAl2O4) nanoparticles using sol–gel auto combustion method and their photocatalytic properties ,SpectrochimicaActa Part A: Molecular and Biomolecular Spectroscopy 131 (2014) 329–334. | | [4] M. Rosso , Ceramic and metal matrix composites: route and properties. | | [5] Ping Fua,b,WenzhongLua,c, Wen Leia,c, KeWub, Yong Xub, JiaminWua,c ,Thermal Stability and Microstructure Characterization of MgAl2O4Nanoparticles Synthesized by Reverse Microemulsion Method,Materials Research. 2013; 16(4): 844-849 | | [6] P.Y. Lee1, H. Suematsu1,T. Yano and K. Yatsui , Synthesis and characterization of nanocrystalline MgAl2O4 spinel by polymerized complex method , Journal of Nanoparticle Research (2006) 8:911–917. | | [7] Structural, electrical and dielectric properties of spinel type MgAl2O4 nanocrystalline ceramic particles synthesized by the gel-combustion method, Ceramics International41(2015)3178–3185. | | [8] P.V. MarakkarKutty, SubrataDasgupta ,Low temperature synthesis of nanocrystalline magnesium aluminate spinel by a soft chemical method ,Ceramics International 39(2013)7891–7894 | | | [9] Lim RooiPinga, Abdul-MajeedAzadb, Teng Wan Dunga , Magnesium aluminate (MgAl2O4) spinel produced via self-heat-sustained (SHS) technique , Materials Research Bulletin 36 (2001) 1417–1430. | | [10] Shay Meir, Sergei Kalabukhov, Natasha Froumin, Moshe P. Dariel,wand Nahum Frage ,Synthesis and Densification of Transparent Magnesium Aluminate Spinel by SPS Processing. J. Am. Ceram. Soc., 92 [2] 358–364 (2009). | | | [11] M. M. Rashad ,E. Z. I. Zaki Æ H. El-Shall , A novel approach for synthesis of nanocrystalline MgAl2O4powders by co-precipitation method ,J Mater Sci (2009) 44:2992–2998. | | [12] M.F. Zawrah, H. Hamaad, S. Meky ,Synthesis and characterization of nano MgAl2O4 spinel by the co-precipitated method ,Ceramics International 33 (2007) 969–978. | | [13] L. G. Jacobsohn,M. W. Blair, S. C. Tornga, L. O. Brown, B. L. Bennett, and | R. E. MuenchausenY2O3 :Bi nanophosphor Solution combustion synthesis, structure, | and luminescence , journal of applied physics 104, 124303 _2008. | | [14] C. P˘acurariu,I. Laz˘au,Z. Ecsedi,R. Laz˘au,P. Barvinschi,G. M˘arginean , New synthesis methods of MgAl2O4 spinel , Journal of the European Ceramic Society 27 (2007) 707–710. |