International Journal of Scientific Research and Engineering Development-– Volume 5 Issue 5, Sep- Oct 2022

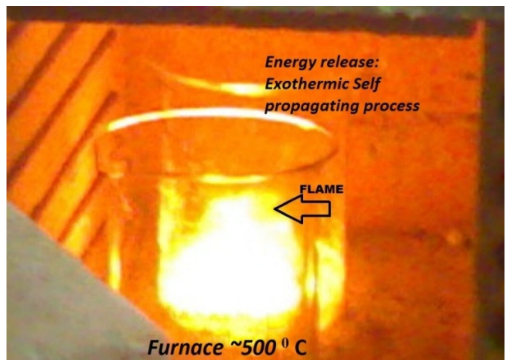
Available at www.ijsred.com

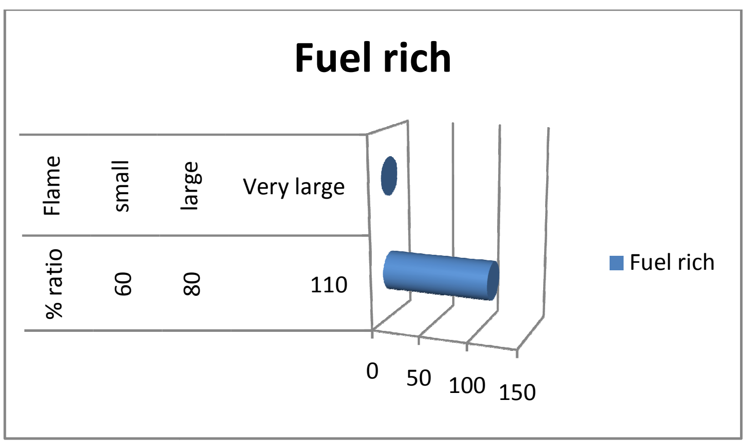
RESEARCH ARTICLE OPEN ACCESS

Comparative Analysis of Solution Combustion Method (Self Propagating High Temperature Synthesis) for Synthesis of Nano-phase Metal Oxides with: Mono & Mixed Fuel Approach

Dr.Amit Sharma,
*Department of Physics, Arni School of Basic Sciences, Horticulture & Agriculture
Arni University, Kathgarh, Indora. 176401
E-mail:vicky2sept@gmail.com

-------------------------------------*****************************----------------------------------





Abstract:

Synthesis of metal oxides by Solution combustion process is widely used approach. This is a economical and simple method for production of various metal oxides, with different properties. Wide range of interest research shown by researchers worldwide to analyze important parameters such as efficiency of different fuels, mixed fuel approach, fuel to oxidizer ratio feasibility of process towards production of different metal oxides. Here comparative analysis of mono and mixed fuel experimental results, with different combinations of fuel to oxidizer ratio is being presented. In each approach data of three samples have been utilized to check out best one from properties or more appropriately application point of view. Flame observation, Scanning electron microscopy and X-ray diffraction were experimental characterization techniques. Exothericity of reaction have fruitful effect to change the properties of product phase respective oxide. Here alumina was target product in nano phase.

Keywords: Metal oxide, SHS, exothermic: self propagating, fuel Oxidizer, ceramic oxides, nano oxides.

---------------------------------------****************************-----------------------------------

1 Introduction:. SHS technology is utilizing less expensive chemical energy instead of the electrical power, thus is simple to perform production mechanism [1-5]. Layer-by-layer heat release that ensures the feasibility to operate with large amount of substance and feasibility of setting up the in-line production [6-15].

Metal oxides are well known for their numerous applications. Metal oxides find their use in manufacturing industry such as tools and parts fabrication. Some most comman applications include spark plugs, tap washers, pump seals, grinding media, abrasion resistant tiles, cutting tools, and wear parts for the textile and paper industries. Researchers from all over the world are working with different methods to produce metal oxides. Commonly used methods for production of metal oxides are sol-gel method, metal matrix method, SHS i.e. self propagating high temperature synthesis or solution combustion [11-15].

Solution combustion or Combustion synthesis (CS) is a excellent, economic and easy mechanism for production of metal oxides. In last few years, a number of new breakthroughs in this field have been observed, notably for production of new catalysts and nano carriers with efficiency better than those for same traditional materials. Alumina (Aluminium oxide) is a widely used metal oxide has many applications in industry (i.e. Abrasive, wear resistant, coating industry etc.) [16-25]Many articles reported synthesis of alumina by SHS (solution combustion) process. Parameter that is most commonly reported in most of papers is effect of fuel to oxidizer ratio on synthesis of metal oxide and properties of product formed [16, 17, 18, 26-40].

Glycine, ammonium acetate, ammonium tartarate, urea have been explored as fuels. Different combinations of fuel to oxidizer ratio have been analyzed [15-20] [41]. Here in this paper comparative analysis of mono & mixed fuel approach with different combinations of fuel to oxidizer ratio undertaken. All samples were prepared and characterized for confirmation of change in related properties from application point of view. Fuel to oxidizer ratio effects exothermic property of product phase (crystallinity, particles size and hardness etc.) . Alumina powder was the target material of experiment, with expected nano-phase. Results analyzed are well in agreement with referred literature.

2. Experimental:

2.1 *Material used (chemical composition & specifications*:[16-28]

(a). Urea:

1	Symbol	$CO(NH_2)_2$
2	Oxidizing-reducing valency	+6
3	Solubility in distilled water	1080g/liter at 25^0 C
4	Molecular Weight	60 g/mole

(b). Aluminium Nitrate non-hydrate(GR):

1	Symbol	$Al(NO_3)_3\ 9H_2O$
2	Oxidizing-reducing valency	-15
3	Solubility in distilled water	637g/liter at 25^0 C
4	Molecular Weight	375 g/mole

(c) Glycine:

1	Symbol	$C_2H_5NO_2$
2	Oxidizing-reducing valency	+9
3	Solubility in distilled water	250g/litre at 25^0 C
4	Molecular Weight	75 g/mole

2.2 *Stoichiometric oxidizer to fuel molar ratio*[16-28]:

$Al(NO_3)_3$ (oxidizer)& $CO(NH_2)_2$ (fuel):

Oxidizer/fuel ratio = 15/6 = 2.5 (From formula) i.e. : Stoichiometric aluminium nitrate: urea molar ratio is 1:2.5.

$Al(NO_3)_3$ (oxidizer) & $C_2H_5NO_2$ (fuel) :

Oxidizer/fuel ratio = 15/9 = 1.66 (From formula) i.e. : Stoichiometric aluminium nitrate : Glycine molar ratio is 1:1.66.

2.3 *Precursor Preparation*: Solution prepared after mixing of oxidizer & fuel before heat treatment:

1. Weighing of aluminium nitrate, urea and Glycine as per required quantity/sample, calculated from

Sample No.	Fuel used (MONO Fuel)	Fuel level	Stoichiometric Fuel %
B1	Urea	Fuel lean	60 %

B2	Urea	Fuel lean	80 %
B3	Urea	Fuel Rich	110 %

(a)

Sample No.	**Fuel used (Mixed Fuel)**	**Fuel level**	**Stoichiometric Fuel %**
A1	(Urea+ Glycine)	Fuel lean	30+30=60 %
A2	(Urea+ Glycine)	Fuel lean	40+40=80 %
A3	(Urea+ Glycine)	Fuel Rich	55+55=110 %

(b)

Sample No.	**Fuel used (MONO Fuel)**	**Fuel level**	**Stoichiometric Fuel %**
C1	Glycine	Fuel lean	60 %
C2	Glycine	Fuel lean	80 %
C3	Glycine	Fuel Rich	110 %

(c)

Table 1: (a) Mixed fuel (b) Mono fuel: Urea (c) Mono fuel Glycine (fuel to oxidizer ratio variation percentage).

formula and sample fuel to oxidizer ratio.

2. Solution of each salt mixture with distilled water as per solubility of salts.

3. In last step homogeneous solution of Aluminium nitrate and fuel was prepared separately. Samples with variation from

Stoichiometric value (as per formula of fuel to oxidizer ratio and ratio decided for a sample in proposed work.) were prepared and homogeneously mixed using magnetic stirrer.

The sample solutions were then subjected to heat treatment in electrical furnace. The value temperature was controlled in range of 300^0c -500^0C, until formation of final product i.e. metal oxide.

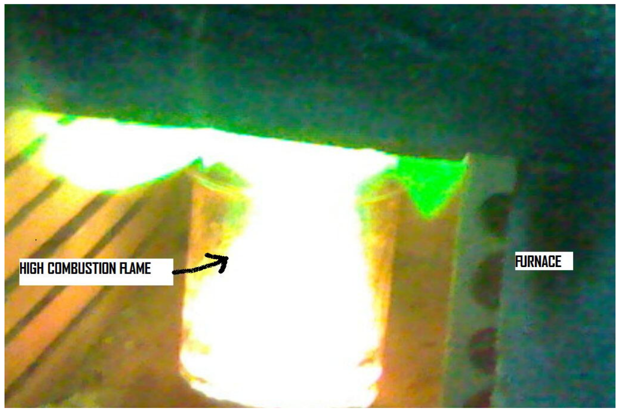
2.4 *Sample preparation for characterization*: After combustion of solution in furnace (Heat treatment) foam like product formed in all samples. These samples were then grounded thoroughly to form a homogeneous powder. Powder samples were used for characterization.

3. Results & discussion:

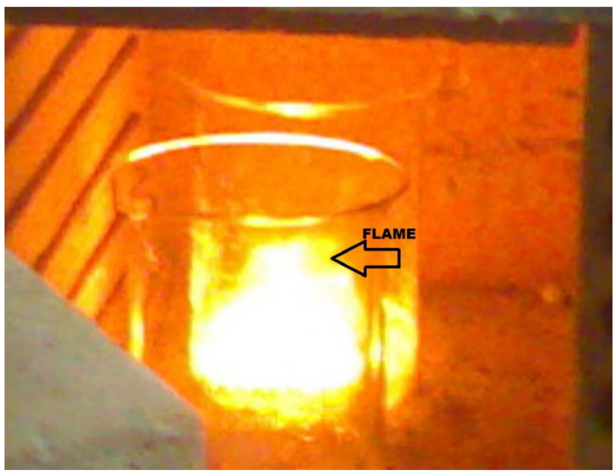
3.1 *Metal oxide formation and Combustion flame observation*: Different colours of metal oxide product phase appeared after combustion process. Also flame content and duration has variation from fuel lean to fuel rich sample. White product formation is indication of complete combustion. Deviation of colour of product phase from white to black/brown may be due to residual carbon content from incomplete combustion reaction. High flame shows highly exothermic reaction and thus sufficient heat for formation of metal oxides.[5-15] Exothermicity of combustion process increases when we proceed from fuel lean to fuel rich sample. N. Sharma et. al. have reported similar behaviour of fuel to oxidizer ratio in production of metal oxide.[41]

Flame observation	
Colour appearance	**Level/Duration**
Brown-Black	Combustion progress with a little self originated flame that remains for about 3 seconds, only observed in some few area of sample.
White (little brownish)	Combustion progress with a self originated flame that remains for about 13 seconds.
White	Combustion progress with a self originated flame that remains for about 16 seconds. Flame raises high inside furnace. As product formed it starts rising inside container until combustion ends.

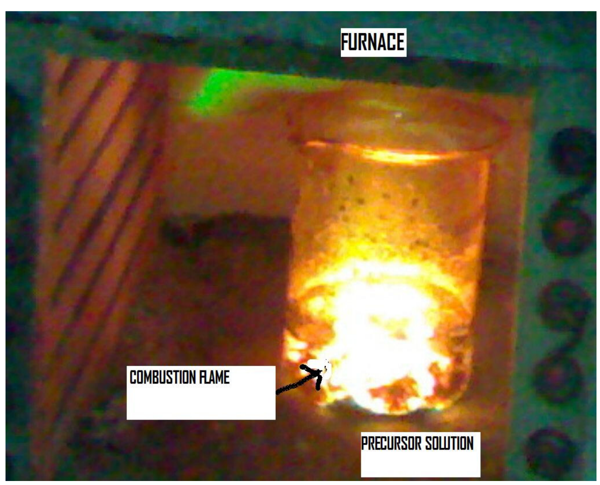
Table 2: Observation of Combustion reaction in furnace.



(a)



(b)



(c)

Figure 1 : Combustion flame during formation of metal oxide.

From literature high flame indicates high exothermic combustion process. Large exothermicity of combustion process results in complete combustion of the reactant material. when the sufficient amount of energy required to achieve crystallization produces in combustion process, crystalline phase forms.

Range of particles size and formation of agglomerates is related to exothermicity of combustion process during production of metal oxide. If small range of particles size and a few agglomerates found in product phase, there will be insufficient heat produced during combustion. All this is related to the amount of flame temperature during combustion process for production of metal oxide. Formation of large particles indicates high combustion flame. [41]

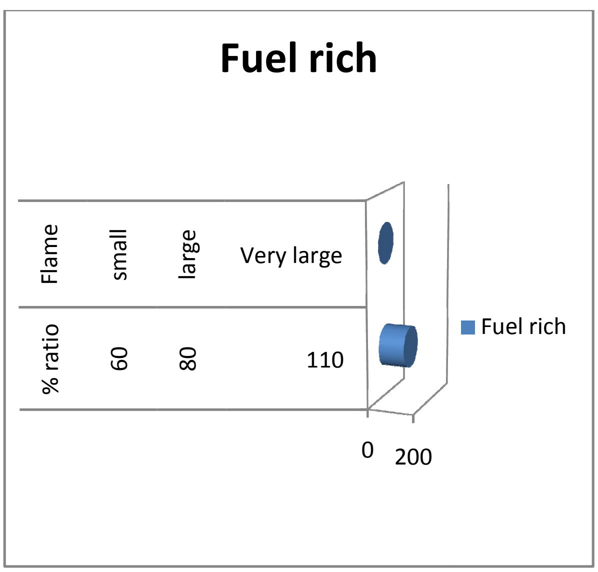


Figure 2: plot of variation in fuel ratio and flame parameter.

It is observed that when fuel lean samples (A1,A2,B1,B2,C1,C2,) observed there is incomplete combustion, which leads to amorphous product and agglomeration of particles. In fuel rich (A3,B3,C3) sampleS complete or sufficient combustion take place, which leads to crystalline product and agglomeration of particles is less, so may approaches nano-pahse.

Another important observation leads the fact that in case of urea as fuel combustion is more efficient and effective. So urea can be better than glycine, as a fuel during solution combustion reaction. Further the particle morphology and nano-pahse can be confirmed from XRD and SEM analysis and co-related with exiting data of already reported work in literature.

4. Conclusions:

From production process of aluminium oxide (Alumina) and analysis of sample characterization we concluded that Fuel lean sample in lowest range of fuel to oxidizer ratio results in incomplete combustion, thus less exothermicity. Small range of exothermicity during heat treatment process gives rise to amorphous metal oxides. In case of little fuel lean and fuel rich sample trend of variation of exothermicity is as observed from literature. As the fuel to oxidizer ratio increases, crystallinity and exothermicity increases.Microstructure study can confirms the extent of desired heat of combustion, by variation in range of particles size and formation of agglomerates. Overall from above analysis one can easily control parameters like mechanical properties (crystallinity or amorphous phase, hardness etc.), Particle size, and grade of metal oxide. Variation in fuel to oxidizer ratio leads to variation in properties of metal oxide formed (alumina powder in present study).

References:

1. Chyi-Ching Hwang,Tsung-Yung Wu, journal of material science 39(2004) 6111-6115. [12] Mukasyan AS, Epstein P, Dinka P. Solution combustion synthesis of nanomaterials. Proc Comb Inst 2007; 31:1789–95.
2. Roth P. Particle synthesis in flames. Proceed Combust Inst 2007; 31:1773–88.
3. Sun Z, Axelbaum BH, Chao A. A multicomponent sectional model applied to flame synthesis of nanoparticles. Proc Comb Inst 2002; 29:1063–9.
4. Stobierski L, Wegrzyn Z, Lis J, Buck M. SHS synthesis of nanocomposite AlN–SiCpowders. Int J Self-Prop High-Temp Synth 2001; 10:217–28.
5. Bernard F, Gaffet E. Mechanical alloying in SHS research. Int J Self-Prop High- Temp Synth 2001; 10:109–32.
6. Borovinskaya IP, Ignat'eva TI, Vershinnikov VI, Khurtina GG, Sachkova NV. Preparation of ultra-fine boron nitride powders by self-propagating hightemperaturesynthesis. Inorg Mater 2003; 39:588–93.
7. Nersisyan HH, Lee JH, Won CW. SHS for a large-scale synthesis method of transition metal nanopowders. Int J Self-Prop High-Temp Synth 2003; 12:149–58.
8. Nersisyan HH, Lee JH, Won CW. The synthesis of nanostructured molybdenum under self-propagating high-temperature synthesis mode. Mater Chem Phys 2005; 89:283–8.
9. Nersisyan HH, Lee JH, Won CW. A study of tungsten nanopowder formation by self-propagating high-temperature synthesis. Combust Flame 2005; 142:241–8.
10. NersisyanHH, Won HI, Won CW, Lee JH. Study of the combustion synthesis process of nanostructured WC and WC-Co. Mater Chem Phys 2005; 94:153–8.
11. Muenchausen RE, McKigney EA, Jacobsohn LG, Blair MW, Bennett BL, Cooke DW. Science and application of oxyorthosilicate nanophosphors. IEEE Trans NucleSci 2008; 55:1532–5.
12. Song H, Chen D. Combustion synthesis and luminescence properties of SrAl2O4:Eu2+, Dy3+, Tb3+ phosphor. Lumin 2007; 22:554–8.
13. Qiu Z, Zhou Y, Lü M, Zhang A, Ma Q. Combustion synthesis of threedimensionalreticular -structured luminescence SrAl2O4:Eu, Dy nanocrystals. Solid State Sci 2008; 10:629–33.
14. Ekambaram S. Solution combustion synthesis and luminescent properties of perovskite red phosphors with higher CRI and greater lumen output. J Alloys Comp 2005; 390:L7–9.
15. Jin Y, Qin WP, Zhang JS, Wang Y, Cao CY. Synthesis of Gd3PO7:Eu3+ nanospheresvia a facile combustion method and optical properties. J Solid State Chem2008; 181:724–9.
16. A.G. Merzhanov, Ceramic international 1995, 21, 371-391.
17. Merzhanov AG, Mukasyan AS. Combustion of solid flame. Moscow: Torus Press; 2007. 336.
18. Mukasyan AS, Martirosyan K, editors. Combustion of heterogeneous systems: fundamentals and applications for material synthesis. Kerala, India: Transworld Research Network; 2007. 234.
19. Borisov AA, De Luca L, Merzhanov AG, editors. Self-propagating high temperature synthesis of materials. New York: Taylor and Francis; 2002. 337.
20. Segadaes AM. Oxide powder synthesis by the combustion route.Eur Ceram News Lett 2006; 9:1–5.
21. [6] Sharma A, Modi O P, Gupta G K, Combustion Synthesis of Nanocrystalline Al_2O_3 Powder using Aluminium Nitrate and Urea as reactants—influence of reactant composition, Advances in Applied Science Research, 2012, 3 (6):3819-3824.
22. Sharma A, Modi O P, Gupta G K **,** Effect of fuel to oxidizer ratio on synthesis of alumina powder using Solution Combustion Technique-Aluminium Nitrate & Glycine combination, Advances in Applied Science Research, 2012, 3 (4):2151-2158.
23. Sharma A , Rani A, Singh A ,Modi O P, Gupta G K, Synthesis of alumina powder by the urea–glycine–nitrate combustion process: a mixed fuel approach to nanoscale metal oxides, Appl Nanosci, DOI 10.1007/s13204-013-0199-8.
24. Mukasyan AS, Rogachev AS. Discrete reaction waves: gasless combustion of solid powder mixtures. ProgEnerg Comb Sci 2008; 34:377–416.
25. [10] Filimonov IA, Kidin NI. High-temperature combustion synthesis: generation of electromagnetic radiation and the effect of external electromagnetic fields. Comb Explos Shock Waves 2005; 41:639–56.
26. Lou XM, Chen DH. Synthesis of CaWO4: Eu3+ phosphor powders via a combustion process and its optical properties. Mater Lett 2008; 62:1681–4.

27. Qiu Z, Zhou Y, Lu M, Zhang A, Ma Q. Combustion synthesis of long-persistent luminescent $MAl_2O_4:Eu^{2+}$, R^{3+} (M = Sr, Ba, Ca, R = Dy, Nd and La) nanoparticles and luminescence mechanism research. Acta Mater 2007; 55:2615–20.
28. Krsmanovi´c R, Morozov VA, Lebedev OI, Polizzi S, Speghini A, Bettinelli M, et al.Structural and luminescence investigation on gadolinium gallium garnet nanocrystalline powders prepared by solution combustion synthesis. Nanotech 2007; 18:325604–13.
29. Xu L, Wei B, Zhang Z, Lü Z, Gao H, Zhang Y. Synthesis and luminescence of europium doped yttria nanophosphors via a sucrose-templated combustion method. Nanotech 2006; 17:4327–31.
30. [31] Russo N, Mescia D, Fino D, Saracco G, Specchia V. N_2O decomposition over perovskite catalysts. IndEngChem Res 2007; 46:4226–31.
31. Ribeiro NFP, Souza MMVM, Schmal M. Combustion synthesis of copper catalysts for selective CO oxidation. J Power Sour 2008; 179:329–34.
32. Roy S, Hegde MS. Pd ion substituted CeO_2: a superior de-NOx catalyst to Pt or Rh metal ion doped ceria. CatalCommun 2008; 9:811–5.
33. Schuyten S, Dinka P, Mukasyan AS, Wolf E. A novel combustion synthesis preparation of $CuO/ZnO/ZrO_2/Pd$ for oxidative hydrogen production from methanol.Catal Lett 2008; 121:189–98.
34. Chen YZ, Zhou W, Shao ZP, Xu NP. Nickel catalyst prepared via glycine nitrate process for partial oxidation of methane to syngas.CatalCommun2008; 9:1418–25.
35. Naik MA, Mishra BG, Dubey A. Combustion synthesized WO_3–ZrO_2 nanocomposites as catalyst for the solvent-free synthesis of coumarins. ColloSurf A PhysicochemEng 2008; 317:234–8.
36. [37] Morales W, Cason M, Aina O, de Tacconi NR, Rajeshwar K. Combustion synthesis and characterization of nanocrystalline WO_3. J Am ChemSoc2008; 130:6318–9.
37. Aarthi T, Madras G. Photocatalytic reduction of metals in presence of combustion synthesized nano-TiO_2. CatalCommun 2008; 9:630–4.
38. Nagappa B, Chandrappa GT. Mesoporous nanocrystalline magnesium oxide for environmental remediation. MicroporMesopor Mater 2007; 106:212–8.
39. [Patil KC, Hegde MS, Tanu Rattan, Aruna ST. Chemistry of nano crystalline oxide materials: combustion synthesis, properties and applications. Singapore: World Scientific; 2008.
40. Sharma N, Sharma A, comparative study of aluminium based metal oxide Synthesized by using different fuel combinations. Irji, june 2016, vol-1,issue-x.