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Aluminosilicate formation in various mixtures of tetra ethyl orthosilicate (TEOS) and aluminum nitrate (ANN)

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Abstract

Reaction between different mixtures of tetra ethyl orthosilicate and aluminum nitrate nonahydrate at varying conditions have been studied by DTA technique. The result reveals that the two components react with the exhibition of an exotherm at ∼110 °C. During isothermal heating at ∼80 °C mixtures show rise in temperature with vigorous evolution of NO_2 fumes and form gels those on heating to ∼500 °C form aluminosilicate precursor phase. The composition of it may be close to the approximate composition of mullite.

Keywords: Aluminosilicate phase; Exothermic reaction

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1. Introduction

In the binary Al_2O_3–SiO_2 phase diagram, mullite of the composition of Al_2O_3:SiO_2 = 3:2 exists as a stable crystalline phase. It has been prepared out of different sources of silica and alumina component materials. Roy [1] first used tetra ethyl orthosilicate (TEOS) and aluminum nitrate nonahydrate (ANN) and thereafter these have been widely used till date as an inorganic cum organic source materials for synthesis of mullite precursor via sol–gel route. The essential factors namely (i) amount of hydrolysis water or alcohol used as solvent, (ii) pH of the reaction medium, (iii) gelation time, and (iv) temperature of the reaction condition influenced the overall gelation process [2–14]. It is shown that all those four reaction parameters affected the hydrolysis cum condensation processes of TEOS and hydrolysis of ANN. These parameters predict the ultimate path of mullite formation of the precursor e.g. whether it forms directly or via an intermediate Al–Si spinel formation route [3–5,8,15–21]. Accordingly, phase transformation sequences of the gels or precursors were found to be different and these are briefly shown by the following equations.

In acidic medium some precursor forms tetragonal mullite (t-mullite) directly and exhibits only 980 °C exotherm and thereafter transforms to orthorhombic mullite (o-mullite) on further heating.

1st type:

Mullite precursor

$$\xrightarrow[\text{Exo}]{980\,^{\circ}\mathrm{C}} \underset{\text{t-mullite}}{2Al_2O_3\cdot SiO_2} + \text{Aluminosilicate (A)} \xrightarrow[\text{heating}]{\text{Continued}} \underset{\text{o-mullite}}{3Al_2O_3\cdot 2SiO_2} \quad (1)$$

While in most practical cases, precursors form both t-mullite and Al–Si spinel of varying ratio's at the 980 °C and thereafter complete o-mullite formation at the second stage of transformations.

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2nd type:

Mullite precursor

$$\begin{aligned}&\xrightarrow[980\,^{\circ}\mathrm{C}]{\text{Exo}} \text{t-mullite (major)} \xrightarrow[\text{heating}]{\text{Continued}} \underset{\text{o-mullite}}{\mathrm{3Al_2O_3\cdot 2SiO_2}}\\ &+ \text{Al–Si } \underset{\text{(minor)}}{\text{spinal phase}} + \text{Aluminosilicate (A)}\\ &\xrightarrow[1250\,^{\circ}\mathrm{C}]{\text{Exo}} \underset{\text{o-mullite}}{\mathrm{3Al_2O_3\cdot 2SiO_2}}\end{aligned} \tag{2}$$

3rd type:

Mullite precursor

$$\begin{aligned}&\xrightarrow[980\,^{\circ}\mathrm{C}]{\text{Exo}} \text{Al–Si spinel (major)}\\ &+ \text{t-mullite (minor)} + \text{Aluminosilicate (A) (minor)}\\ &\xrightarrow[1250\,^{\circ}\mathrm{C}]{\text{Exo}} \underset{\text{o-mullite}}{\mathrm{3Al_2O_3\cdot 2SiO_2}}\end{aligned} \tag{3}$$

Some other precursor mostly forms Al–Si spinel at 980 °C at the first stage and thereafter forms o-mullite at two exothermic reactions, respectively.

4th type:

$$\begin{aligned}&\text{Mullite precursor} \xrightarrow[\text{1st Exo}]{980\,^{\circ}\mathrm{C}} \text{Al–Si Spinel} \underset{\text{Path I}}{\xrightarrow[\text{2nd Exo}]{1245\,^{\circ}\mathrm{C}}} \underset{\text{o-mullite}}{\mathrm{3Al_2O_3\cdot 2SiO_2}}\\ &\qquad\qquad\qquad\qquad \text{Aluminosilicate phase (A)} \underset{\text{Path II}}{\xrightarrow[\text{3rd Exo}]{1150\,^{\circ}\mathrm{C}}} \underset{\text{o-mullite}}{\mathrm{3Al_2O_3\cdot 2SiO_2}}\end{aligned} \tag{4}$$

In case of spray pyrolyzed precursor, complete mullite formation occurs at the 980 °C exotherm [21].

5th type:

Spray dried or spray pyrolyzed precursor

$$\xrightarrow[980\,^{\circ}\mathrm{C}]{\text{1st Exo}} \text{t-mullite} \xrightarrow[\text{heating}]{\text{Continued}} \underset{\text{o-mullite}}{\mathrm{3Al_2O_3\cdot 2SiO_2}} \tag{5}$$

Diphasic gel obtained in basic condition of gelation of ANN/TEOS or by using boehmite sol in acidic condition with TEOS shows that phase transformation process is entirely different from above cases of monophasic gels. It does not exhibit 980 °C exotherm but forms Al–Si spinel in the temperature range 1100–1300 °C. It finally transforms to o-mullite at ∼1300 °C exotherm:

6th type:

$$\begin{aligned}&\underset{\text{Diphasic gel}}{\text{Mullite precursor}} \xrightarrow[1100\text{–}1300\,^{\circ}\mathrm{C}]{\text{1st Exo}} \text{Al–Si spinel phase}\\ &+ \text{Aluminosilicate (A)} \xrightarrow[1270\text{–}1320\,^{\circ}\mathrm{C}]{\text{2nd Exo}} \underset{\text{o-mullite}}{\mathrm{3Al_2O_3\cdot 2SiO_2}}\end{aligned} \tag{6}$$

During the gelation process, TEOS is first hydrolyzed and developed silicic acid gel which physically adsorbed $\mathrm{Al^{3+}}$ ion from aluminum nitrate solution in its network. $\mathrm{Al^{3+}}$ also substituted $\mathrm{Si^{4+}}$ in the gel structure and formed Si–O···Al linkages as shown by IR studies [22,23]. Generally, Si–O stretching vibration for pure silica gel occurred at ∼1120 $\mathrm{cm^{-1}}$ and this decreased to 1080 $\mathrm{cm^{-1}}$ in coprecipitated gel heated to 900 °C which indicated a considerable effect possibly due to substitution. Formation of aluminosilicate compound during heating those gels was also shown by MAS NMR studies [10,11,14,24,25]. An asymmetrical shape of ^{29}Si resonance peak was noted in mullite precursor at ∼−92 ppm range. According to Irwin et al. [26,27] and Yasumori et al. [28], ^{29}Si resonances are assigned to four aluminum atoms via oxygens {Q_0 (4Al) sites} and to Q_4 (0Al) sites. While with heating of mullite precursor, ^{29}Si resonance shifted to more negative value ∼−100 ppm. This phenomenon was explained with the increase in number of –Si–O–(Si, Al) bridges i.e. Q_4 (1Al) environment which corresponds to an increasing degree of condensation of the –Si–O–(Si, Al) network. Still some researchers thought that mullite precursors is an amorphous mixture of silica and alumina phases [16]. Firstly, the detail reaction course regarding the possibility of any chemical reaction between two components at low temperature range in the aqueous or alcoholic stage is not fully explored. In the present study, DTA run of the two pure components and their mixtures prepared at different reaction conditions have been made sequentially. Secondly, the changes in temperature of these mixtures have been noted at definite time interval during an isothermal heating process. Results indicate that these components react with the exhibition of an exotherm and form an amorphous aluminosilicate phase.

2. Experimental

2.1. DTA study

To detect the temperature of occurrence of the reaction of the two components, a series of DTA run were performed and these are chronologically stated below:

(i) Components e.g., pure TEOS, ANN, concentrated aluminum nitrate solution.
(ii) Mixtures e.g., (a) ANN–TEOS paste. It was prepared as follows. Pure aluminum nitrate of 10 g and 2 ml TEOS were mixed and ground in an agate mortar for 10 min when a paste like mass was obtained. (b) Alcoholic mixture of ANN–TEOS. This was prepared by mixing 10 ml alcohol to the above paste.
(iii) Isothermal heating of TEOS–$\mathrm{Al(NO_3)_3\cdot 9H_2O}$ mixture.

Table 1
Batch composition of ANN and TEOS

Batch mark	Approximate wt.% ratio of Al_2O_3 to SiO_2 in the powder	Amount of TEOS taken (ml)	Amount of ANN taken (g)
ISG24	76/24	4	2.5
ISG40	60/40	4	5
ISG56	56.25/43.75	4	10
ISG66	66.66/33.33	4	15
ISG72	72/28	4	20
ISG76	76.27/23.72	4	25
ISG80	79.41/20.58	4	30

A constant temperature water bath was taken in this study. Its temperature was maintained constant at 80 ± 2 °C. TEOS of 4 ml was taken in a round-bottomed flask and it was placed into the water bath until it attained the temperature of the bath. To the fixed amount of TEOS, different amounts of ANN were added to make different batches as shown in Table 1. Weighed amount of aluminum nitrate crystals were taken in a separate beaker and its temperature was raised gradually up to ~80 °C when solid melted first into a solution and then it concentrated on continued heating. After attainment of constant temperature, the concentrated aluminum nitrate solution was added into the flask containing TEOS at the same temperature of ~80 °C. The temperature of the mixed solution was found to rise slowly at the beginning and then it rose rapidly just after swirling the flask with simultaneous effervescence of brown fumes of NO_2. During this heat evolution process, the temperature of the solution was noted at regular interval of time using a thermometer from the beginning of the experiment to the end of it. In case of the batch marked ISG72, the temperature of the reaction mixture was finally increased to maximum of 101 °C. Thereafter, it decreased slowly with time until temperature subsided to ~80 °C. Gelation of the reaction mixtures started in this period. On continued heating, the gel gradually dehydrated and deprepitation took place in side the flask. The product was collected and stored for X-ray analysis. The experiment was repeated for successive batches. The evolved exothermic energy as a measure of rise in the temperature of reaction batches versus time periods are plotted. All these dried gels were studied for DTA to note the occurrence of endothermic–exothermic heat effects during their transformations on heating. These gels were also heat-treated to different temperatures and analyzed for XRD to note the changes occurring during the heating process.

3. Results

3.1. DTA study of pure components

(i) Pure TEOS exhibits an exothermic peak at 275 °C (Fig. 1A) due to the decomposition of ethoxy groups.
(ii) Pure $Al(NO_3)_3{\cdot}9H_2O$ exhibits two endotherms (Fig. 1B). First endotherm of small magnitude occurs at ~95 °C may be due to elimination of physical moisture; the other at ~170 °C relatively broad and large in magnitude and is due to its water of crystallization and decomposition of nitrate.
(iii) It is observed that during heating aluminum nitrate nonahydrate crystals on boiling water bath, it first melts with its own water of crystallization, and then it is gradually concentrated. But the concentrated solution neither evaporated to dryness nor even decomposed to liberate

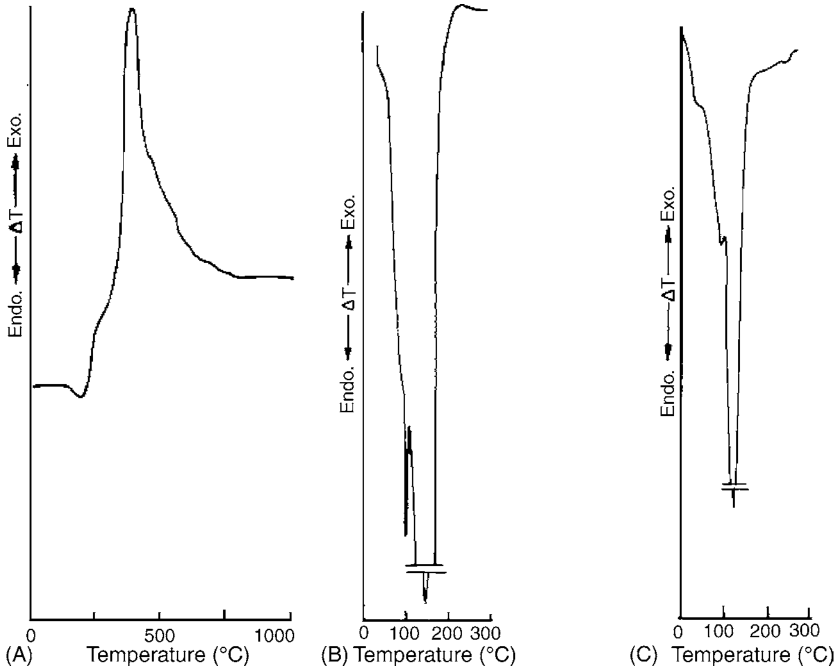


Fig. 1. DTA pattern of (A) TEOS, (B) ANN and (C) concentrated aluminum nitrate.

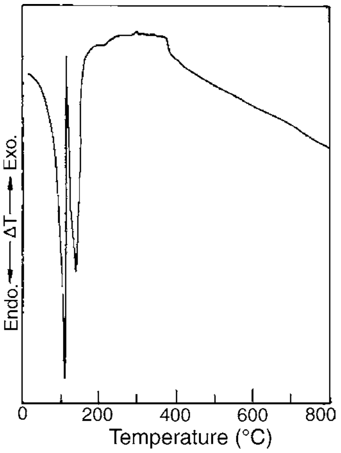


Fig. 2. DTA pattern of TEOS–ANN paste.

NO_2 fumes even on continued heating for long duration of time. pH of this solution is shown to be acidic. DTA run of such concentrated mass is shown in Fig. 1C. It shows a small endotherm at 48 °C, and a large endotherm at 170 °C, respectively. The second endotherm is found to be less intense in comparison to the same effect noted in Fig. 1B.

3.2. *DTA study of TEOS–Al(NO$_3$)·9H$_2$O mixture*

(i) DTA analysis of ANN–TEOS paste shows a completely different picture (Fig. 2) other than that noted in Fig. 1A. Contrary to the exhibition of a sharp exotherm at 275 °C vide Fig. 1A, it exhibits a broad exotherm between 260 and 435 °C. Secondly, it also shows two endotherms, one at 85 °C and the other at 137 °C, respectively. Significantly, the large endotherm which was noted at ~170 °C in case of DTA run of pure ANN now becomes reduce. Moreover, a sharp exotherm newly appears at ~110 °C. The results indicate that although both the components decompose independently, a partial interaction between them may occur and it leads to the exhibition of an exotherm at ~110 °C.

(ii) Alcoholic mixture of ANN–TEOS when analyzed for DTA shows two endotherms as usual. First one occurs at 78 °C and the other at 160 °C, respectively. The second endotherm increases to a large extent which may be related to the addition of alcohol (Fig. 3). Exotherm for decomposition of TEOS still persists, however, the nature of the peak is broad. Exotherm at 110 °C within the two endotherms becomes more predominant.

3.3. *Isothermal heating of TEOS–Al(NO$_3$)$_3$·9H$_2$O mixture*

During heating TEOS and ANN at constant temperature at ~80 °C in water bath, it is shown that the temperature of the reaction medium increases as well as fumes of NO_2 emerges out. The two phenomenon e.g., sudden rise of temperature and vigorous evolution of fumes as observed in isothermal heating studies is equivalent to the exhibition of an exothermic reaction at ~110 °C during heating of the two reactants (Figs. 2 and 3). This study also shows that the temperature of the reaction bath increases first with the increase of Al_2O_3/SiO_2 ratio of the batch mixture of the components up to ISG72 and then decreases (Fig. 4).

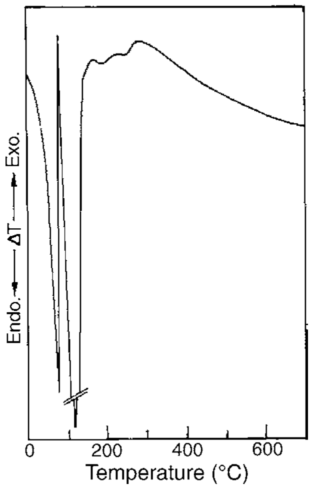


Fig. 3. DTA pattern of alcoholic solution of TEOS–ANN paste.

3.4. *DTA studies of isothermal heat-treated gels*

DTA study of two representative samples marked ISG40 and ISG72 are shown in (Fig. 5). DTA run of both the gels exhibit the 1st exotherm at ~976 °C. ISG72 gel further exhibits 2nd exotherm at ~1250 °C. The other gel marked ISG80 shows a small 1st exotherm peak only (not shown).

3.5. *Phase identification of heated gels*

The phase transformation of these gels when studied by XRD shows the following results (Fig. 6A–C).

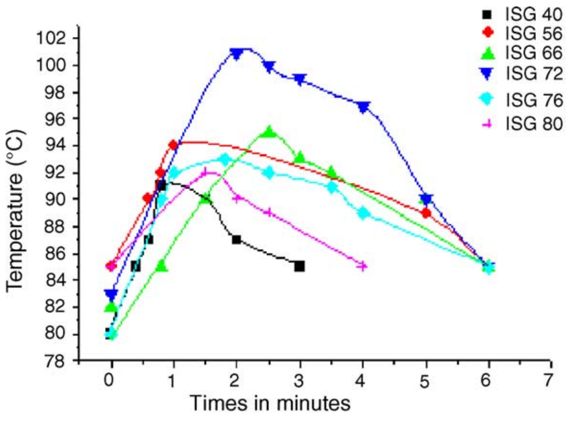


Fig. 4. Time–temperature plot of various mixtures of TEOS–ANN at ~80 °C.

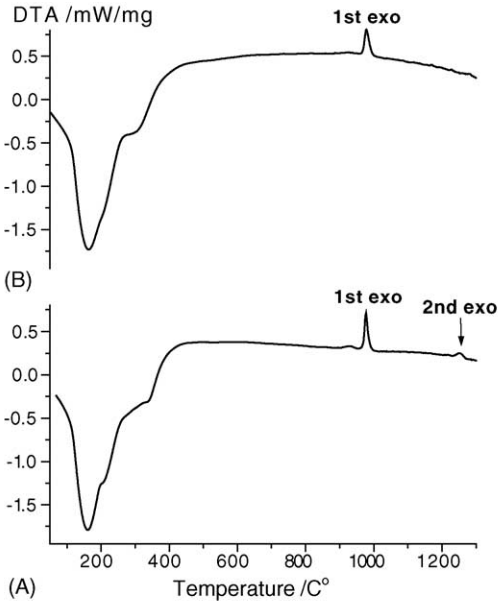


Fig. 5. DTA traces of batches mark ISG40 and ISG72.

Raw ISG40 and ISG72	Amorphous in nature
At ∼800 °C	Both are amorphous
At ∼1000 °C	Both form a mixture of Al–Si spinel and t-mullite (tetragonal mullite). These phases are found to coexist at 1100 °C of heat treatment
At ∼1200 °C	Both the gel form a mixture of major quantity of orthorhombic mullite and minor amount of Al–Si spinel phase. The intensity peaks of mullite increase to more extent at this temperature of heating
At ∼1250 °C	Residual Al–Si spinel phase transforms and complete mullite formation occurs, splitting of peaks begin to take place at this temperature

Contrary to the above phase changes, ISG80 forms spinel phase as early as at ∼800 °C. It starts mullite formation at ∼1000 °C and contains a large proportion of spinel phase. At ∼1100 °C, significant amount of mullite forms along with some residual amount of spinel phase. At ∼1200 °C, o-mullite forms usually in addition to formation of small quantity of corundum.

4. Discussions

4.1. Heat evolution behavior of gels

Isothermal heating of the components shows the evolution of heat/rise of temperature in all cases of reaction mixture under study (Fig. 4). Question is: what happens during heating of two components? To understand it, DTA run of gels leading to mullitization is first of all presented.

4.2. Phase transformation of gels

XRD analysis of ISG40 and ISG72 heated around 1000 °C show the formation of both Al–Si spinel phase and t-mullite. Peak intensity of residual Al–Si spinel decreases on further heating with consequent increase of mullite phase. At ∼1250 °C, complete transformation of it occurs to orthorhombic mullite. By corroborating DTA and XRD results the phase transformation of gel ISG72 (equivalent to the composition of mullite) is given below:

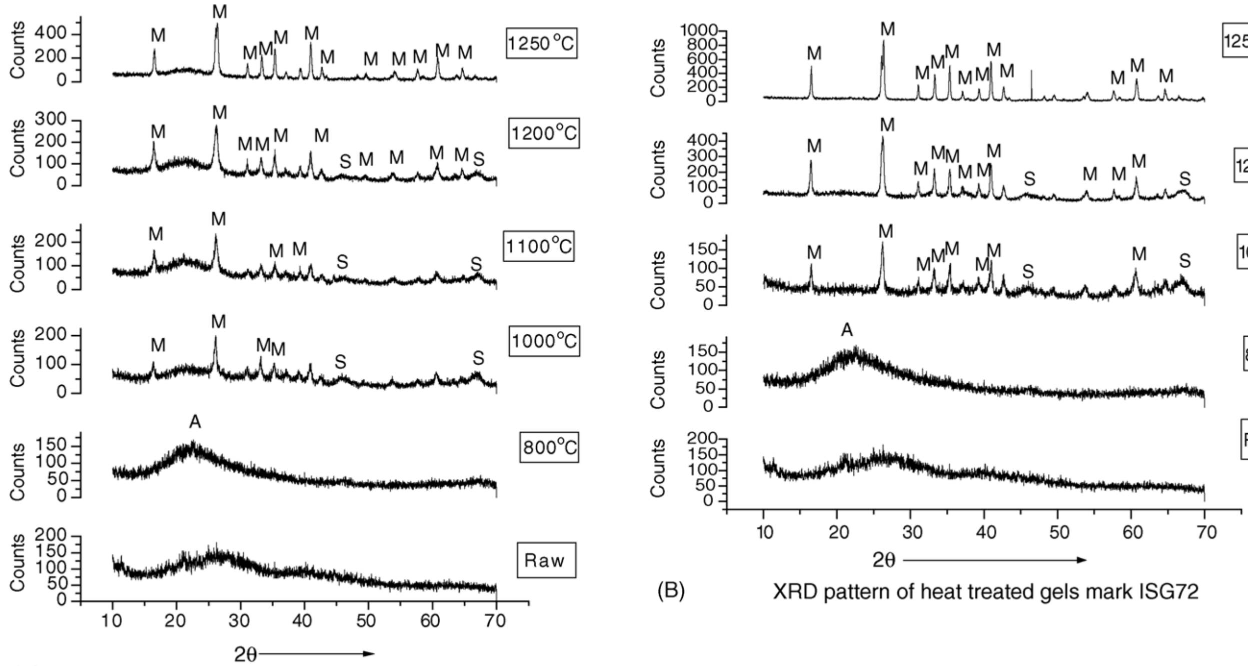
$$\text{Mullite gel} \xrightarrow[\sim 977\,^{\circ}\text{C}]{\text{1st Exo}} \text{Al–Si spinel} + \text{t-mullite} \xrightarrow[\sim 1250\,^{\circ}\text{C}]{\text{2nd Exo}} \underset{\text{o-mulite}}{3Al_2O_3 \cdot 2SiO_2} \tag{7}$$

Thus transformation of ISG72 is analogous to that in case of second case of mullite gel as stated in the review section.

This phase transformation behavior is more or less alike to that of other batches except formation of two individual component oxides excess to that of 3:2 mullite. For example, when the alumina content in gel is in excess to that of 3:2 mullite, the gel ISG80 partially crystallizes to α-Al_2O_3 in addition to usual mullitization process (Fig. 6C). Further, when the silica content in the gel is higher to that of 3:2 mullite, the gel ISG40 shows an amorphous band at $\sim 22° 2\theta$ due to free glassy silica in addition to usual mullite (Fig. 6A). Mullite is the absolute phase when there is no free alumina or free silica in gels other than 3:2 composition of mullite.

4.3. Explanation of ANN–TEOS reaction

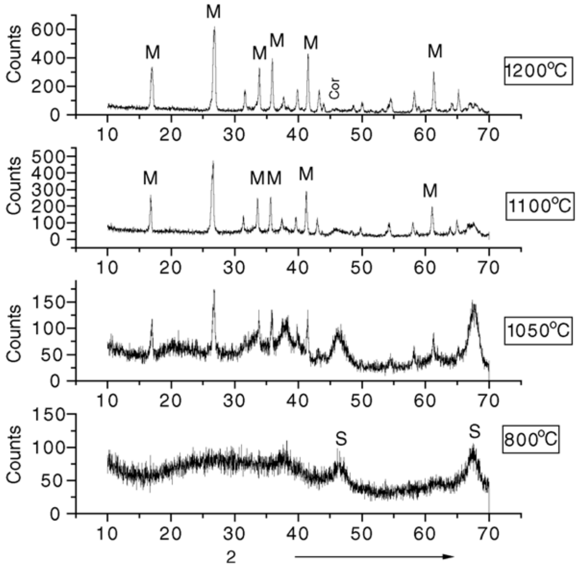
DTA studies of different runs of TEOS–ANN mixtures prepared at various conditions exhibit an exotherm at ∼110 °C (Figs. 2 and 3) in comparison to the traces of individual components (Fig. 1A–C). The exhibition of this exotherm could be explained as due to the reaction between to components during dynamic heating process. The result further shows that the reaction is accelerated in the presence of alcohol used as a medium of hydrolysis of TEOS, and it is even more favorable when ANN is concentrated expecting few moles of its water of crystallization is removed. In isothermal heating condition, the reaction shows evolution of heat



(A) XRD pattern of heat treated gels mark ISG40

M = Mullite , S = Spinel

(B) XRD pattern of heat treated gels mark ISG72



(C) XRD pattern of heat treated gels mark ISG 80

Cor = Corundum

Fig. 6. (A–C) XRD pattern of heat-treated gels mark ISG40, ISG72 and ISG80.

over a long period of time and the reaction temperature drops to 80 °C from the same effect occurs at 110 °C in DTA (Fig. 4)

Before drawing any conclusion regarding the interaction between TEOS and ANN, we should discuss hydrolysis process of TEOS. TEOS is first hydrolyzed to develop silanol groups. These groups thereafter polymerize to develop Si–O–Si bonds. Polymerization and condensation rate is dependent on the pH and quantity of water [30–32]. When pH is fixed between 3 and 4.5 and excess water is used, a three-dimensional chain of Si–O–Si cluster may possibly develop [12]. However, in absence of use of free moisture, when water of crystallization of ANN is the only source of water, hydrolysis of TEOS is restricted and a two-dimensional chain of Si–O–Si may form [13]. When aluminum nitrate solution is added to TEOS solution as in the later case, substitution of Al^{3+} in Si–O–Si network might have occurred. The resulting gel is termed as slow hydrolysis gel (SH gel) by Okada and Otsuka [3]. Question is: to what extent Al^{3+} replaces Si^{4+} in tetrahedral position. The usual rule is that Al_2O_3 to the extent of 30–50 wt.% of SiO_2 may be substituted in the network and form aluminosilicate [32,33]. It would be presumed that residual alumina is present in free state and is mixed uniformly with aluminosilicate hydrate phase. Various researchers called this state of mixture as homogeneous mixing of two oxides components. There is a possibility that amorphous Al_2O_3 might diffuse into the hydrated aluminosilicate network during continued heating and form amorphous aluminosilicate phase.

When 9 mol of water of crystallization of ANN is mostly eliminated by repeated evaporation technique, it is expected that silanol group is not formed. The resultant mullite gel directly forms t-mullite as in first case of transformation and in case of spray pyrolysis precursor.

The reaction condition followed in the present study is somewhat different. For example, temperature of hydrolysis is increased form 60 °C in usual case to 80 °C and secondly the water used for hydrolysis is reduced by concentrating ANN solution. In consequence, TEOS is not allowed to hydrolyze fully and the possibility of hydrolysis of ANN to develop $[Al(H_2O)6]^{3+}$ is reduced when warm concentrated solution of ANN is added to TEOS at the same temperature at ∼80 °C. Under these circumstances, it is assumed that silanol groups may form to partial extent. Therefore, in absence of silica network substitution phenomenon may not occur at higher temperature of 80 °C. Considering a large heat evolution started just after mixing of the two reactants around 80 °C, it may be conceived that nitrate radicals of concentrated aluminum nitrate solution oxidizes ethoxy groups of tetra ethyl orthosilicate and itself reduces to NO_2 groups and with generation of Si and Al linkages via oxygen atoms. Heat of reaction of this TEOS–ANN interaction may claim for rise of the temperature of the reaction bath. This event is related to the exhibition of an exotherm at ∼110 °C in DTA analysis (Figs. 2 and 3).

The process of hydrolysis of TEOS is also exothermic. Jons and Fischbach [34] noted that no reaction was apparent when a mixture of water and TEOS was kept for several hours. When it was acidified with HNO_3, the reaction started, and showed an exothermic reaction. But it lasted only for about 15 s and thereafter the solution became clear. The exothermicity is depending upon the effects of water to TEOS ratio, and HNO_3 to TEOS ratio used in the reaction condition. In the present study, external water and HNO_3 are not used, moreover, the heat evolution continues for ∼10 min. The most striking observation is that during the reaction course of TEOS–ANN reaction vigorous evolution of NO_2 fumes take place.

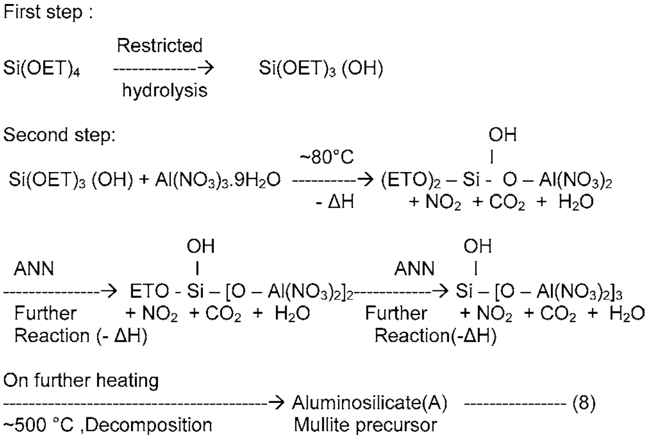
Thus, it is conjectured that interaction between two components occurs definitely during evolution of brown nitrous fumes and exothermic heat energy evolved during isothermal heating process may be considered as the heats of reaction of the batch taken under study. The dried precursor is amorphous to X-ray and it is an aluminosilicate other than free oxide mixture as assumed earlier [1,4]. Yamada and Kimura [22] believed that during gelation Al^{3+} and Si^{4+} were bonded through oxygen and formed alumina gel of silicic acid.

Thus, in the changed condition of gelation process, especially when the temperature is increased from 60 °C as maintained in synthesis of SH gel to 80 °C, interaction between components is predominant. The entire process of batch mark ISG72 is explained in the following ways. TEOS first hydrolyses to develop silanol group. The newly formed $(OR)_3Si(OH)$ may react vigorously with $Al(NO_3)_3$ or a direct reaction between the free components may take place in partially acidic medium as follows:

1st step:

$$Si(OET)_4 \xrightarrow[\text{hydrolysis}]{\text{Restricted}} Si(OET)_3(OH)$$

2nd step:



$$Si(OET)_4 \xrightarrow[\text{hydrolysis}]{\text{Restricted}} Si(OET)_3\,(OH)$$

$$Si(OET)_3\,(OH) + Al(NO_3)_3.9H_2O \xrightarrow[-\Delta H]{\sim 80°C} (ETO)_2 - \overset{\overset{OH}{|}}{Si} - O - Al(NO_3)_2 + NO_2 + CO_2 + H_2O$$

$$\xrightarrow[\text{Further Reaction } (-\Delta H)]{ANN} ETO - \overset{\overset{OH}{|}}{Si} - [O - Al(NO_3)_2]_2 + NO_2 + CO_2 + H_2O \xrightarrow[\text{Further Reaction}(-\Delta H)]{ANN} \overset{\overset{OH}{|}}{Si} - [O - Al(NO_3)_2]_3 + NO_2 + CO_2 + H_2O$$

$$\xrightarrow[\sim 500\ °C\text{ ,Decomposition}]{\text{On further heating}} \underset{\text{Mullite precursor}}{\text{Aluminosilicate(A)}} \quad (8)$$

Thus, the final reaction product of Eq. (8) which is designated as mullite precursor may be an aluminosilicate. This hypothesis concurres with previous NMR results [10,11,14, 29–33]. The DTA exotherm at ∼110 °C shown by ANN–TEOS mixture would be an indication of aluminosilicate formation. XRD shows aluminosilicate is amorphous (Fig. 6B).

Isothermal heating study shows rise of temperature during gelation process (Fig. 4). Now question is: what should be the composition of aluminosilicate? The above figure also shows that with increase of wt.% of Al_2O_3 in the batch, the apparent area under the time–temperature curve during heat evolution in the temperature range 80–90 °C first increases slowly in the batch number ISG40 to ISG56 and then attains rapidly in the batch ISG72. Even with increase of wt.% of Al_2O_3 the said area decreases slowly as in the case of batch ISG76 and then to the last batch ISG80. The batch ISG72 shows only mullite when heat-treated. Gels with higher or lower composition of silica beyond 3:2 composition of mullite form not only mullite but form additional silica or corundum phases. Thus both isothermal heating experiments of components and phase transformation studies of the resultant gels indicate that interaction between TEOS with melted aluminum nitrate crystals shows maximum in case of ANN/TEOS mixture for the composition corresponding to 3:2 mullite approximately. Therefore, the composition of aluminosilicate (A) phase which is formed out of TEOS and ANN reaction is likely to the composition close to 3:2 mullite.

5. Conclusions

Interaction between TEOS and ANN has been studied by DTA technique and by isothermal heating process. The following conclusions are arrived. TEOS reacts vigorously at ∼80 °C with evolution of heat/exothermic energy. During the said exothermic reaction nitrous fumes are rapidly emerged out. The resultant mullite precursor is an amorphous aluminosilicate. The composition of it may be near to the composition of 3:2 mullite.

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