

Preparation of water soluble silicon compound and its application for synthesis of $(Y,Ce,Gd)_2SiO_5$ blue emission phosphor

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A novel water-soluble silicon compound was synthesized by the reaction between tetraethoxysilane (TEOS) and propylene glycol. This compound is stable in aqueous solutions and miscible with water in any ratio. The potential of this new silicon reagent for synthesis of multicomponent silicate by solution based techniques was evaluated by preparation of the $(Y,Ce,Gd)_2SiO_5$ blue emission phosphor using a polymerizable complex method. Phase compositions and fluorescence properties of the prepared material was compared against reference samples prepared by either conventional solid state reaction or by a polymerizable complex method employing the other silicon reagents. It was revealed the application of new water-soluble silicon compound as a reagent for synthesis resulted in the formation of a single phase $(Y,Ce,Gd)_2SiO_5$ material at reduced temperatures and its fluorescence intensity considerably outperformed those of the phosphors prepared by the other methods in this work.

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1. Introduction

Historically, ceramics, which contain silicon, have been almost solely utilized as high temperature or refractory materials.^{1),2)} The developments of novel silicon-containing functional materials are progressing quite rapidly, and recent achievements can be represented by the exciting discovery of strong yellow luminescence in α -SiALON activated with appropriate rare earth elements,^{3),4)} that can be excited by blue light, making it possible to use such a phosphor as a constituent of a white LED.

At the same time, in general silicon-containing ceramics are known to be difficult to synthesize in the pure form or with the desired phase composition and/or morphology. Usually conventional solid-state reaction (SSR) methods are used for synthesis, which rely upon slow diffusion of the components and require extremely high temperatures, typically higher than 1600°C.^{5),6)} This creates a strong limitation for preparation of a variety of silicon-containing ceramics in the systematic manner and to discover their possible new functionalities. In order to overcome this problem encountered in the SSR method, the attempts to synthesize silicon-containing ceramics by solution methods have been seriously considered.

Among various solution-based technologies,^{7),8)} quite significant progress was achieved in low-temperature synthesis of the functional silica glass by the so-called sol-gel method using tetraethoxysilane (TEOS) as a raw material.⁹⁾ However, use of TEOS as a Si source makes the synthesis of multi-component silicon-containing ceramics with precisely controlled chemical composition by the described sol-gel method very difficult due to possible evaporation of TEOS during the solvent elimination process. The current difficult situation in the synthesis of silicon-containing ceramics by known solution methods has stimulated

the present authors to develop the new unique water-soluble silicon compound (WSS), which may enable convenient synthesis of multi-component silicon-containing materials by employing an appropriate aqueous solution based technique.

In this paper, $(Y,Ce,Gd)_2SiO_5$ (YCGS) multicomponent oxide, which is expected to be used as a blue phosphor¹⁰⁾ for a field emission display (FED),¹¹⁾ was chosen as a model compound. This choice was also motivated by the fact that YCGS is known as a fluorescent material which is difficult to be synthesized in its pure form,^{12)–15)} and in addition it is believed that further enhancement of the fluorescence properties can be achieved by simultaneously improving the phase purity of Y_2SiO_5 as the host phase and distributing co-activators (Ce and Gd) in Y_2SiO_5 uniformly. Thus, the objective of this work was to clarify if WSS is suitable as a raw material for the preparation of silicon containing multi-component ceramics by a solution method by examining the potential of this precursor in synthesis of YCGS with high phase purity. In addition, reference YCGS specimens were prepared by the alternative solution technique such as polymerizable complex (PC) method¹⁶⁾ in which TEOS or fumed silica were used as silicon reagents. Fluorescent properties and phase composition of thus prepared materials were compared.

2. Experimental

2.1 Synthesis of water-soluble Si-compounds (WSS)

The WSS stock solution was prepared according to the procedure shown in Fig. 1. 22.4 mL of Tetraethoxysilane (TEOS Kanto Chemical Co., Inc., 95%) and 22.4 mL of propylene glycol (PG Kanto Chemical Co., Inc., 99.9%) were transferred by a digital pipette into a 50 mL Erlenmeyer flask with a stopper. The two-phase liquid was magnetically stirred at 80°C for 48 h. Due to the vigorous stirring the initially transparent system consisting of an upper layer of TEOS and bottom layer of PG transformed into a white emulsion. At this stage 0.1 mL of hydrochloric acid

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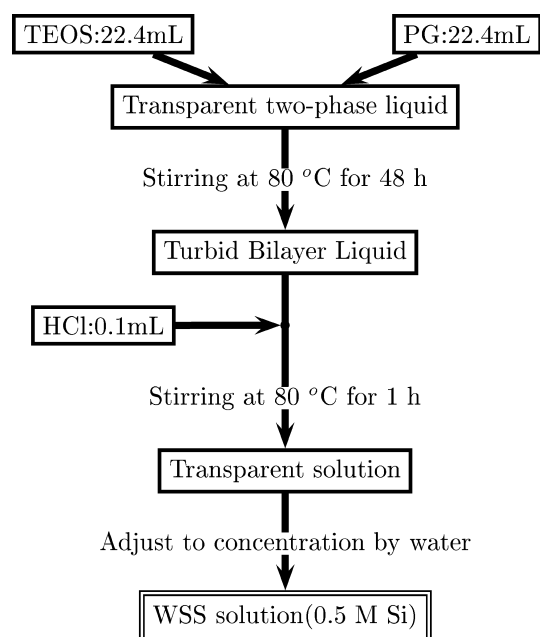


Fig. 1. Flowchart of experimental procedures for preparation of water soluble silicon compound aqueous solution.

(Kanto Chemical Co., Inc., 35 wt%) was introduced by a digital pipette and after stirring for additional 1 h at 80°C the emulsion turned into a transparent uniform solution. This solution can be mixed with water in any ratio without hydrolysis of the silicon compound. This water-soluble silicon compound is referred to as WSS hereinafter. The WSS was transferred into a 200 mL volumetric flask and diluted with distilled water to obtain a 0.5 mol/dm³ of silicon-containing aqueous stock solution.

2.2 Synthesis of (Y,Ce,Gd)₂SiO₅

In this work a phosphor material of (Y_{0.79}Ce_{0.01}Gd_{0.20})₂SiO₅ composition was synthesized by two methods such as a polymerizable complex (PC) method and a conventional solid-state reaction (SSR) method. For synthesis by the PC method three Si compounds including WSS, silicon dioxide (SiO₂ Sigma, USA, fumed silica 7 nm) and TEOS were used as Si sources. The final phosphors were obtained by heat-treating the precursors prepared by each technique under the same conditions.

2.2.1 Preparation of precursors for (Y,Ce,Gd)₂SiO₅ by PC method

The sequence of procedures for preparation of a precursor by PC method is shown in **Fig. 2**. Solutions of yttrium and gadolinium nitrates with 1 mol/dm³ concentration were prepared by dissolving Y₂O₃ (AGC Seimi Chemical Co., Ltd., 98.5%) and Gd₂O₃ (Shin-Etsu Chemical Co., Ltd., 99.9%) into aqueous solutions of nitric acid (Kanto Chemical Co., Inc., 60%). To remove the excess of nitric acid, the solvent was evaporated under a reduced pressure at 80°C using a rotary evaporator. The obtained solid nitrates were re-dissolved in distilled water. In the case of Ce, 1 mol/dm³ cerium nitrate aqueous solution was used. As a silicon source, three different reagents were compared: 0.5 mol/dm³ stock solution of WSS prepared as described above, SiO₂ (Fumed silica) and TEOS. In addition, 2 mol/dm³ aqueous solution of citric acid (CA) (Wako Pure Chemical Industries, Ltd., Japan, 98%) and PG were used as chelating agent and as esterification reagent respectively. Three samples were prepared in parallel to ensure similar synthesis conditions. Solutions of

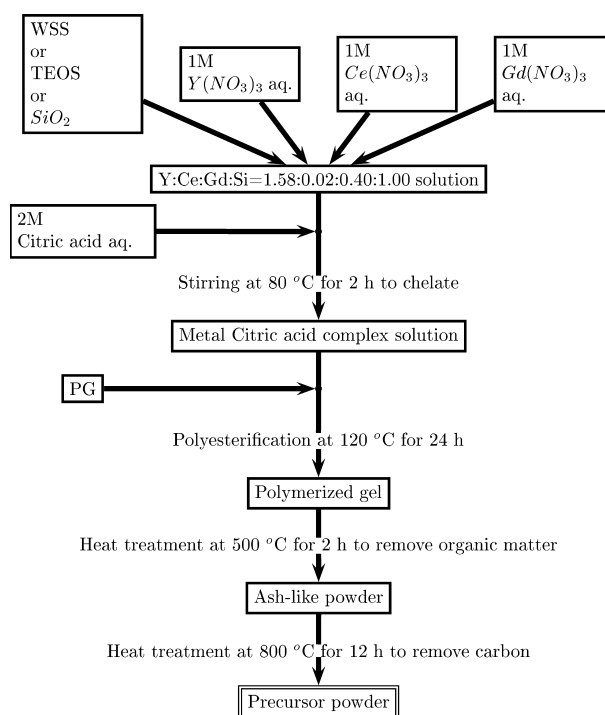


Fig. 2. Flowchart of precursors preparation by polymerizable complex method for (Y,Ce,Gd)₂SiO₅ phosphor material.

yttrium, cerium and gadolinium nitrates were mixed with silicon containing precursors in the stoichiometric ratio to obtain (Y_{0.79}Ce_{0.01}Gd_{0.20})₂SiO₅ target composition. When WSS was used, a clear and uniform solution was obtained. In the case of fumed silica the obtained solution was turbid due to light scattering by SiO₂ colloid particles. When TEOS was utilized, the liquid sample consisted of two immiscible liquid phases.

Then aqueous solution of CA was added to these three solutions to establish final [All cations]:[CA] molar ratio equal to 1:4 followed by stirring at 80°C for 2 h using a hot stirrer. Finally, PG was introduced to obtain mole ratio of [CA]:[PG] = 1:1. The solutions were heated up and kept at 120°C for 24 h with continuous stirring to promote polyesterification reaction between CA and PG. It should be mentioned that when WSS or SiO₂ were used as silicon sources, polyesterification process yielded a yellow transparent and homogeneous resin, while in the case of TEOS as a precursor the system contained white cluster species separated from the yellow transparent resin.

To decompose organic compounds involved in the prepared polymerized resin samples, these products were heat-treated at 500°C for 2 h in a sand bath placed into a mantle heater. The obtained ash-like powders were transferred to alumina crucibles and heat treated at 800°C for 12 h in an electric furnace to remove residual carbon components completely. The samples after oxidation of carbon were used as precursors for the target phosphor.

2.2.2 Preparation of precursors for (Y,Ce,Gd)₂SiO₅ by SSR method

Y₂O₃, Ce(CH₃COO)₃ (Kanto Chemical Co., Inc., 99.99%), Gd₂O₃ and SiO₂ (Wako Pure Chemical Industries, Ltd., 70 nm) were mixed manually for 1 h in the required stoichiometry using an agate mortar. The powder was transferred into an alumina crucible and heat treated at 800°C for 12 h in the electric furnace to obtain a precursor for the phosphor.

2.2.3 Synthesis of (Y,Ce,Gd)₂SiO₅ from the precursors by a double crucible method

Final heat treatment was performed under a reducing atmosphere in the presence of graphite as a reducing agent at high temperatures using a double crucible setup. 0.5 g of a precursor powder was put into the inner alumina crucible of 30 mm diameter, and 4 g of graphite was placed into the alumina crucible of larger diameter. Subsequently, the inner crucible with the sample was put into the outside crucible with graphite and the system was covered by a lid to setup a “double crucible” as it is shown in **Fig. 3**. This double crucible system was heat-treated at 1600°C for 2 h in an electric furnace to obtain final phosphor materials.

2.3 Characterizations

The obtained samples were analyzed by a powder X-ray diffractometer (Rigaku Co., RINT-2200) and using a fluorescence spectrometer (Hitachi, Ltd., F-4500). Lattice parameters of the target compound X₂-Y₂SiO₅ (ICSD #27007) for each individual samples were evaluated based upon 37 independent reflections observed between 20 and 50° using a software PowderX. The measured excitation and emission intensities were rescaled and plotted as relative values compared against one of the best commercially available (Y, Ce)₃Al₅O₁₂ (YAG: P46, Kasei Optnix Ltd., Tokyo) excited under its optimal conditions (the excitation wavelength: 460 nm, the emission wavelength: 573 nm).

3. Results and discussion

3.1 Characterization of samples phase purity by X-ray diffraction

X-ray diffraction patterns of the synthesized samples are shown in **Fig. 4**. The maximum diffraction intensities in each pattern were normalized to the same value to make data comparison more convenient. In the case when WSS and fumed silica were used as starting reagents (“WSS” and “SiO₂” in the picture) the diffraction patterns of the final materials matched well to the reference pattern of the high temperature phase X₂-Y₂SiO₅ (ICSD #27007), indicating a single-phase nature of the sample. On the other hand, Y₂O₃ was detected as the main phase when TEOS was used as a source of silicon (“TEOS” in Fig. 4). This result indicates that silicon was lost from the system due to volatile nature of TEOS (*T*_{bp} = 169°C), and as a result Y₂O₃

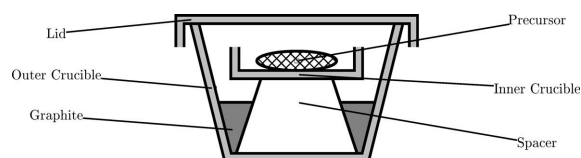


Fig. 3. Schematic view of a double crucible system used for synthesis of (Y,Ce,Gd)₂SiO₅ phosphor in the reducing atmosphere.

appeared as an yttrium-rich phase in the silicon-deficient sample. Although the material prepared by conventional solid state reaction (“SSR” in Fig. 4) was not single phase either, presence of Y₂SiO₅ and Y₂Si₂O₇ reflections in the XRD patterns indicate that such a phase composition is not related to the deficiency of one of the components, rather it is connected with the incomplete reaction resulting from the inhomogeneous nature of the mechanical mixture of the starting powder materials.

The calculated values of the lattice parameters of the samples ((Y_{0.79}Ce_{0.01}Gd_{0.20})₂SiO₅) and those for the non-doped X₂-Y₂SiO₅ are shown in **Table 2**. The lattice parameters of the doped sample prepared by use of WSS are considerably bigger than those of the non-doped sample, which may indicate substantial substitutions of both Ce and Gd with higher ionic radii for Y with a smaller ionic radius. On the other hand, the corresponding lattice parameters of the remaining samples prepared by use of SiO₂ or TEOS as well as by the solid-state reaction method are practically identical to those for the non-doped X₂-Y₂SiO₅, the reason for which remains unclear. Insufficient substitution of Gd for Y in the X₂-Y₂SiO₅ samples could partially explain the unchanged behavior of their lattice parameters, although we have no experimental evidence for that.

The phase purity of a ceramics strongly depends on how homogeneously metallic constituents are distributed in a given precursor for the final ceramic material and usually one circle of grinding and calcinations is not sufficient to achieve excellent homogeneity for the silicate materials. On the other hand, when

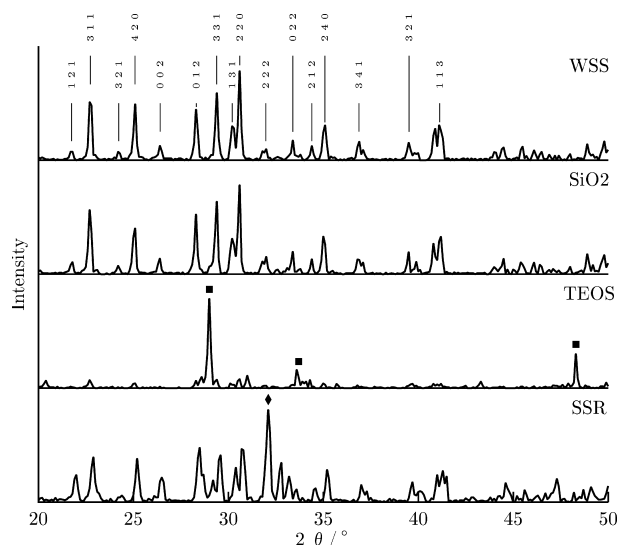


Fig. 4. XRD patterns of (Y,Ce,Gd)₂SiO₅ materials synthesized by PC method using water soluble silicon reagents. “SSR” denotes the sample prepared by solid state reaction. ■: Y₂O₃, ◆: Y₂Si₂O₇.

Table 2. Lattice Constant of Samples

Sample	a/pm	b/pm	c/pm	α/°	β/°	γ/°
Ideal Y ₂ SiO ₅	1459.000	1052.000	682.000	90.000	90.000	122.250
WSS	1497.928 ± 0.059	1067.200 ± 0.042	696.377 ± 0.032	90.707 ± 0.002	88.675 ± 0.002	122.257 ± 0.003
TEOS	1458.362 ± 0.062	1050.084 ± 0.045	680.119 ± 0.034	90.017 ± 0.002	89.968 ± 0.002	122.250 ± 0.003
SiO ₂	1456.204 ± 0.068	1050.592 ± 0.049	680.635 ± 0.038	89.999 ± 0.003	90.001 ± 0.003	122.250 ± 0.003
SSR	1458.383 ± 0.062	1052.536 ± 0.045	680.936 ± 0.034	89.994 ± 0.002	90.011 ± 0.002	122.250 ± 0.003

PC method together with WSS was applied for synthesis of $(Y_{0.79}Ce_{0.01}Gd_{0.20})_2SiO_5$ all four components, such as Y, Ce, Gd and Si as water-soluble compound, were homogeneously mixed on the atomic level in the aqueous solution leading to the most uniform precursor among different systems investigated in this work. For the other systems, the uniformity may be inferior since TEOS forms a bilayer liquid, fumed silica is suspended in the homogeneous aqueous solution of Y, Ce and Gd, while in the case of SSR relatively large particles are mechanically mixed together. The results of phase composition analysis by XRD measurements support these speculations.

3.2 Fluorescence properties of the prepared materials

The excitation and emission spectra of the prepared samples are shown in Fig. 5. The solid lines correspond to the emission spectra excited with 356 nm wavelength, while the corresponding excitation spectra with an emission wavelength of 422 nm are plotted as dotted lines. The emission peaks with maximum at 420 nm were observed for all samples when they were excited by 356 nm light of and this emission is characteristic for Ce^{3+} activator in Y_2SiO_5 host.¹³⁾ The shape of the emission spectrum of the material prepared by the PC method using WSS appears to differ to some extent from that of the XRD-single phase sample prepared by the PC method using SiO_2 as a silicon source, which might be due to the difference of crystal structures (the size of lattices) between the two samples revealed by their lattice parameters.

The intensity of the emission peak was the highest for the material prepared using WSS precursor ("WSS" in Fig. 5) and it decreased in the following order "WSS">" SiO_2 ">"SSR">"TEOS". Relative to the peak intensity of one of the best commercial YAG: Ce^{3+} phosphor P46, emission intensities of the prepared samples were 0.78, 0.59, 0.48, and 0.24 for "WSS", " SiO_2 ", "SSR" and "TEOS" respectively. This trend in emission intensities corresponded well to the order of materials with respect to their phase purity as it was observed by X-ray diffraction (Fig. 4). Fluorescence intensity of a solid state material is the convenient structure-sensitive property, which reflects not only phase composition, but also homogeneity of activator ions distribution in the host lattice. Thus, although for both "WSS" and " SiO_2 " samples single-phase Y_2SiO_5 was obtained, the emis-

sion from "WSS" sample was significantly stronger than from the single phase " SiO_2 " sample having the same bulk composition. This fact indicates that Ce emitting centers in "WSS" material were distributed in the host compound more uniformly. In the case of solid state reaction process, which employs metals oxides and SiO_2 as starting reagents, diffusion is the main kinetic process, and one may expect that due to the different atomic weight and ionic radius Y, Ce and Gd ions will possess different diffusion coefficients, so it might be very difficult to achieve high homogeneity in the $(Y,Ce,Gd)_2SiO_5$ solid after one heat treatment step. Therefore, when a solution process is combined with a solid silicon precursor, it can be expected that yttrium ions will have higher mobility due to smaller radius and lower weight than Gd and Ce. Thus, with respect to Ce^{3+} homogeneity " SiO_2 " sample was inferior to "WSS", and as a result its emission was weaker. Another factor influencing the emission intensity of a phosphor includes its particle size. SEM observations of all the samples indicate almost identical morphology, which is characterized by particles with diameters of approximately 1 μm agglomerated each other. We therefore conclude that the variation of the observed emission intensities from sample to sample was not attributed to the particle size but to the degree of the homogeneity of the activator (Ce^{3+}) distributed in the host material as well as to the phase purity of the host.

$(Y,Ce,Gd)_2SiO_5$ is an important phosphor material for the devices which use the electronic excitation. In this case the mechanism of the emission involves excitation of Ce ions by an ultraviolet light emitted by Gd ions, which were excited directly by the electron beam.¹⁷⁾ Thus, from the practical viewpoint the degree of uniformity of not only Ce but also Gd ions will affect strongly the emission brightness. In such a case one should expect even larger difference between the luminescence intensities of "WSS" and " SiO_2 " materials.

3.3 Recovery factor and silicon deficiency

Except for the sluggish kinetics inherent to the solid state reaction, which leads to the inhomogeneous materials or poor phase composition, this work revealed one more aspect important for synthesis of silicon containing multicomponent materials by solution methods. Namely silicon deficiency might be quite significant if volatile compounds are used for synthesis. To obtain a quantitative estimation of silicon loss during the synthesis of silicon-containing ceramics by a solution method we defined a *Recovery Factor*. It can be presented by the Eq. (1) involving the theoretical mass (M_I) of the oxide material calculated from the starting composition and mass of the experimentally obtained product (M_P) from the precursor:

$$R(\%) = (M_P/M_I) \times 100\% \quad (1)$$

One may notice that definition of the recovery factor is very similar to the definition of the yield of chemical reaction since it combines in one equation theoretical and actual amounts of products. Nevertheless, there is one important difference between definition (1) and yield since M_P may include different crystal phases and therefore in the strict sense it can not be referred to as yield. Furthermore, if we assume that in our particular case the deviation of the recovery factor $R(\%)$ from 100% is explained by solely volatilization of silicon component, the recovery factor $R_{Si}(\%)$ can be defined for Si in the similar manner. The detailed explanations for $(Y_{0.79}Ce_{0.01}Gd_{0.20})_2SiO_5$ composition are provided below¹⁸⁾ and here we will present only the final formula (2).

$$R_{Si}(\%) = 5.23 \times R(\%) - 423.05 \quad (2)$$

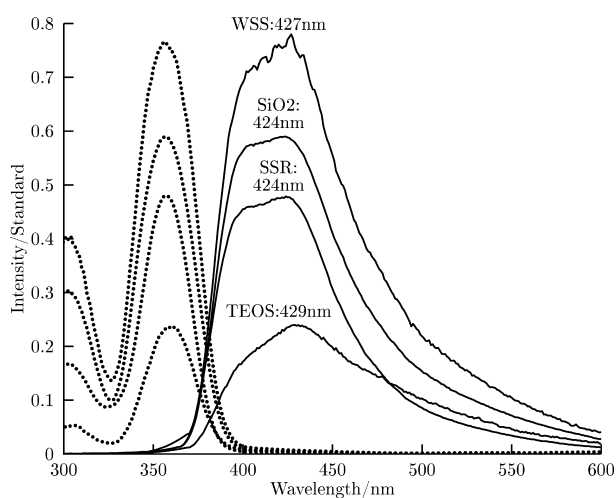


Fig. 5. Excitation (dotted line) and emission (solid line) spectra of $(Y,Ce,Gd)_2SiO_5$ materials prepared by different techniques. ($\lambda_{em} = 422$ nm, $\lambda_{ex} = 356$ nm).

Table 1. Overall Recovery Factor $R(\%)$ and Silicon Recovery Factor $R_{Si}(\%)$ Estimated for Synthesis of (Y,Ce,Gd)₂SiO₅ Phosphor by Polymerizable Complex Method Employing Different Silicon Sources and by Solid State Reaction

Recovery/%	WSS	TEOS	SiO ₂	SSR
R	101.1	89.6	99.2	99.3
R_{Si}	105.3	45.1	95.3	95.8

The values of recovery factor of $R(\%)$ and the silicon recovery factors $R_{Si}(\%)$ are shown in **Table 1**. The symbols “WSS”, “TEOS” and “SiO₂” in this table correspond to the silicon precursors used in the polymerizable complex method. The result for synthesis by solid state reaction is denoted as “SSR”. One may notice that $R(\%)$ values for “WSS” and “SSR” were close to 100%. Since both of these samples were single phase, the recovery factor for solid state reaction gives a good idea of typical yield in the usual sense of chemical reaction, while slightly higher than 100% value for WSS may allow to estimate typical uncertainty of these calculations. Thus, the recovery rate of 90% in the case when TEOS was used is an evidence of loss in terms of the final product. Now if we will assume that all 10% loss of weight is due to escape of silicon in the form of TEOS or organic compounds, which may form with PG or other products in the course of polymerizable complex method, and such a deficiency corresponds to SiO₂, it means that 55% of Si was lost during the synthesis (Table 1). Therefore, TEOS should be regarded as inappropriate Si source for synthesis by this solution process. In sharp contrast to TEOS, volatility of Si component is negligible in the case of WSS, and a material with precisely controlled stoichiometry can be obtained.

4. Conclusion

The water soluble silicon compound (WSS) was synthesized by reaction between tetraethoxysilane (TEOS) and propylene glycol. A single-phase (Y,Ce,Gd)₂SiO₅ phosphor exhibiting a strong emission was successfully prepared by a solution based method utilizing this aqueous solution of WSS. The synthesis of high quality phosphor material was possible due to the combination of two important factors: high degree of components mixing on the atomic level achievable by employing an aqueous solution of WSS and low volatility of silicon components during the solvent removal process if WSS is used. Based on the obtained results, it can be concluded that WSS is an effective Si source for synthesis of silicon containing ceramics by use of aqueous

solution based techniques.

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