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Effect of Calcination Temperature of SiO_2-TiO_2 Composite XRD Characterization of Crystal Size Structure and Phase

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Abstract: Calcination is often used to activate beneficial metal oxidation in catalysts and improve material stability. In the metal oxidation research used is SiO_2-TiO_2 nanocomposite because it has advantages over photocatalysts and thermal stability. One important factor that affects is the particle size and crystal phase formed during the calcination process at various temperatures. The purpose of the study was to determine the effect of calcination on changes in phase and crystal structure in SiO_2-TiO_2 nanocomposites. The material used for the manufacture of nanocomposites is natural silica source from coconut fiber which is burned into white ash. The ash was extracted for the preparation of silica sol by adding Tetraethyl Orthosilicate (TEOS) as a precursor synthesized by the sol gel method then, calcination was carried out with temperature variations of 700℃, 800℃, and 900℃. After calcination, XRD characterization was carried out to obtain crystal sizes of 63.48 nm, 66.87 nm, and 73.83 nm. The phase formed is dominant in TiO_2 is rutile and SiO_2 is quartz and cristobalite.

Keywords: Calcination; Crystal Size; SiO_2-TiO_2 Composite; Temperature.

1. Introduction

Calcination is the process of heating materials at high temperatures to change physical and chemical properties. The purpose of calcination is to strengthen the structure of the material surface, form oxidation, remove water content, increase or change crystallinity properties and to remove impurities such as organic compounds. Calcination is needed as a preparation of powders for further processing and also to obtain the optimum particle size and decompose compounds in the form of salts or dihydrates into oxides, forming a crystalline phase. Events that occur during the calcination process include: (1) The release of free (H_2O) and bound (OH) water takes place around 100℃ to 300℃, (2) The release of gases takes place around 600℃ and at this stage is

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accompanied by a significant weight reduction, and (3) At higher temperatures, around 800℃, the crystal structure is formed and changes the phase structure of the material [1].

The purpose of calcination is to remove unwanted materials and change the chemical structure of a material. Calcination is often used to activate the oxidation of useful metals in catalysts and increase the stability of the material. Some metal oxides used in the manufacture of composites are titanium oxide (TiO_2) and silica (SiO_2). Titanium dioxide (TiO_2) has several advantages, namely non-toxic, inert, has good photocatalyst activity, has a large surface area, photosensitive, thermal stability and high chemical stability [2]. The structure of silica can be either amorphous or polymorphic namely quartz, tridymite, and cristobalite [3]. Silica (SiO2) has the advantages of high surface area, high mechanical stability and thermal stability and is abundant. In composite synthesis, tetraethyl orthosilicate (TEOS) or tetramethyl orthosilicate (TMOS) is generally used as a source of silica or as a precursor. Silica derived from coconut coir ash synthesis can be obtained in an easier way because it is more environmentally friendly, relatively cheaper and raw materials are available in large quantities. [4].

Figure 1. Coconut Husks

In Figure 1 coconut husks, coconut plants are classified as follows [5]:

Kingdom : Plantae
Divisi : Spermatophyta
Sub Divisi : Angiospermae
Ordo : Palms
Familia : Palmae
Genus : Cocos
Species : *Cocos nucifera* L

The isolated coconut fiber silica content was analyzed quantitatively using XRF to determine the silica content of coconut fiber and other oxides. In the study of coconut fiber silica with a purity of 51.5% and the remaining 38.5% in the form of oxide compounds that cannot be removed. So, one of the ways used to obtain SiO_2 with high purity can be obtained by synthesizing SiO_2 using natrium hydroxide (NaOH) [6].

Metal nanoparticles have different physical properties from metals in bulk form, for example, they have lower melting points, larger specific surface areas, and special optical properties. Although both bulk and nanoparticle materials are made of the same atoms, they will exhibit different colloidal colors at the nanoscale. In general, the size of nanoparticles is between 1 - 100 nm [7]. How can nanoscale particles be measured? One way to measure particles on the order of nanometers is with the X-ray diffraction technique. The width of the diffraction peaks is also affected by micro tension (lattice strains), which are the effects of the displacement of a unit cell around its normal position. This is often produced by several factors including: (1) Non-uniform lattice distortion, which can result from nanocrystal surface tension, crystal shape morphology, and interstitial impurities. (2) Dislocations, (3) Inter-phase domain boundaries, which are formed during the preparation of material structures that undergo preparation transformation disorders. This technique is often used in research, especially to determine various physical parameters of materials such as crystal structure, strain, phase composition, unit cell structure, crystal defects and crystal size, even the arrangement of atoms in amorphous materials such as polymers. X-ray diffraction techniques often use samples in powder form, especially in characterizing crystallographic structure, crystal size (grain size) and crystal orientation [8].

Calcination of SiO_2-TiO_2 nanocomposite is one of the important effects of changing the structure of phase formation. At high calcination temperatures can cause an increase in crystal size and a change in crystal phase. In the research [9] the resulting data shows that increasing the calcination temperature results in higher crystallinity, larger crystallite size, and phase transformation in TiO_2 nanoparticles. SiO_2 affects the phase transition of TiO_2 such as from anatase to rutile form, where the addition can change the transition rate and modify the crystal properties of TiO_2 during the high temperature calcination process [10].

In this research also [11] observed for TiO_2 samples calcined at 700-750℃, where most of the TiO_2 is almost in pure rutile phase while the surface is in a mixed phase of anatase and rutile. The anatase phase in the surface region of the TiO_2 sample will change completely to the rutile phase when the calcination temperature is increased to 800℃. This study can be defined that by increasing the calcination temperature or calcination time, anatase TiO_2 gradually changes to the rutile phase. The new thing in this research is adding TiO2 with SiO2 using higher calcination, where the addition of materials will be able to change the shape of the phase and particle size so that it has good thermal stability.

This research will focus on the phase and particle size of the effect of calcination temperature on SiO2-TiO2 nanocomposites and is expected to provide insight into the structure of nanocomposites to achieve thermal stability.

2. Materials and Method

This research is a type of experimental research that discusses the Effect of Calcination Temperature of SiO_2-TiO_2 Composite XRD Characterization on Crystal Size and Phase Structure. The research was conducted in August 2023 at the Physics Department Laboratory and the Chemistry Department Research Laboratory, Faculty of Mathematics and Natural Sciences, Padang State University. Calcination temperature variations are 700℃, 800℃, and 900℃. In this study, the control variables are the furnace temperature of 600℃ for coconut fiber.

The materials used in the synthesis and characterization of SiO_2-TiO_2 composites include, *Tetraethyl Orthosilicate* (TEOS, Sigma Aldrich, 98%), Titanium dioxide, distilled water, ethanol (Merck, 99.0%), hydrochloric acid (0.1 M HCl) and 0.1 M NaOH. The tools used in the synthesis and characterization of SiO_2-TiO_2 composites include beakers, measuring cups, test tubes, magnetic stirrers, spatulas, drop pipettes, HEM, and furnaces.

Preparation of Coconut Coir Ash. The sample used in this study was old coconut fiber obtained from Hole village. In this study using the sol-gel method to synthesize silica from coconut fiber ash processing and applied to the calcination of SiO_2-TiO_2 nanocomposites. XRD Characterization. Characterization of the calcination composite was carried out using an X-ray diffraction to determine the phase and crystal size. The data from this study consisted of SiO_2-TiO_2 nanocomposites calcined with temperature variations, morphology identification test data with XRD characterization of SiO_2-TiO_2 nanocomposite layers.

Figure 2. Sample Preparation

The coconut fiber sample was cut according to Figure 2. Next, the ashing process aims to perfect combustion and remove organic components that are still present in the sample. coconut fiber in the furnace with a temperature of 600℃. Coconut fiber that has become ash weighed 10 grams, carried out wet milling with a HEM milling tool for 5 hours. In accordance with Figure 3

Figure 3. Coconut Coir Ash

Preparation of Sodium Silicate (Na_2SiO_2). First, coconut coir ash that had been HEM was dissolved in NaOH with a silica to NaOH ratio of 0.1M 50ml at a temperature of 250℃ for 2 hours and stirred using a magnetic stirrer. Next, 200 ml of distilled water was added and stirred using a magnetic stirrer for 1 hour. Stirring was done with the aim that the coconut fiber ash could be mixed evenly with the NaOH solution. Then the solution that has been made is filtered using filter paper, the solution that is filtered through is a solution of Sodium Silicate (Na_2SiO_3). The Sodium Silicate solution was then stirred using a magnetic stirrer and titrated using 0.1M HCl. The solution was then precipitated for 24 hours, then washed with distilled water to remove the salt (NaCl) formed during the titration process. The precipitate was then filtered and dried, then ground with a mortar.

Preparation of SiO_2 Precursor. In a glass beaker Tetraethyl orthosilicate (TEOS) as much as 5 ml was added to 5 ml ethanol and then stirred for 10 minutes at room temperature at 300 rpm. TEOS solution was formed. Next, in a new beaker glass, 10 ml of distilled water is added plus 0.1 M HCl as much as 3-4 drops and then stirred. After that, add sodium silicate solution and then stirred for 5 minutes at room temperature at 300 rpm. Then transfer the solution to a hotplate at 60°C for 2 hours and stirred at 500 rpm. Then the solution was cooled until it became a gel.

Silica gel is modified with TiO_2, which functions as a catalyst. The results of mixing Titanium Dioxide (TiO_2) powder and silica gel powder were composited by sol-gel method with TiO_2 that had been purchased weighed as much as 4 grams, then mixed into SiO_2 sol and added 1% acetic acid as much as 60 ml using a magnetic stirrer for 1 hour at 800 rpm. Then added 0.1 M NaOH as much as 50 ml and stirred at 60℃ for 2 hours. Then dried using an oven at 70℃ for 5 hours and formed a composite powder.

The SiO_2 -TiO_2 composite was dried and calcined using a furnace with a temperature variation of 700℃, 800℃, and 900℃ with a time of 1 hour, after which it was allowed to stand. Furthermore, grinding the composite results that have been in the furnace until smooth. The smooth composite was put into each sample bottle and labeled according to the calcination temperature variation.

XRD data interpretation is carried out. Determination of the particle size of the SiO_2 -TiO_2 composite can be calculated by using the equation

$$D = \frac{\lambda.\kappa}{\beta cos\theta} \quad (1)$$

Where:

D = size of the crystal (nm)

λ = Wavelength of X-ray radiation (nm)

β = FWHM value in radians (°)

θ = Bragg diffraction angle

Actual K values vary from 0.62 to 2.08. Commonly used values for K are: 0.94 if theta is FWHM and 0.89 for Integral Breadth [7]. From equation 1 it can be observed that the peak width varies with the angle 2theta in the form $\cos\theta$.

3. Results and Discussion

Characterization of SiO_2-TiO_2 nanocomposites using XRD testing of SiO_2-TiO_2 composites aims to characterize the crystal structure and determine the phase formed after calcination of each sample due to the influence of variations in calcination temperature used. Analysis of the diffraction patterns obtained is compared with the diffraction pattern database (such as JCPDS or ICSD) to determine the phase formed. Figure 4, Figure 5, and Figure 6, are the results of XRD testing of SiO_2-TiO_2 composites due to the influence of calcination temperature variations, where there is a rutile phase for TiO_2 and α and γ phases for SiO_2.

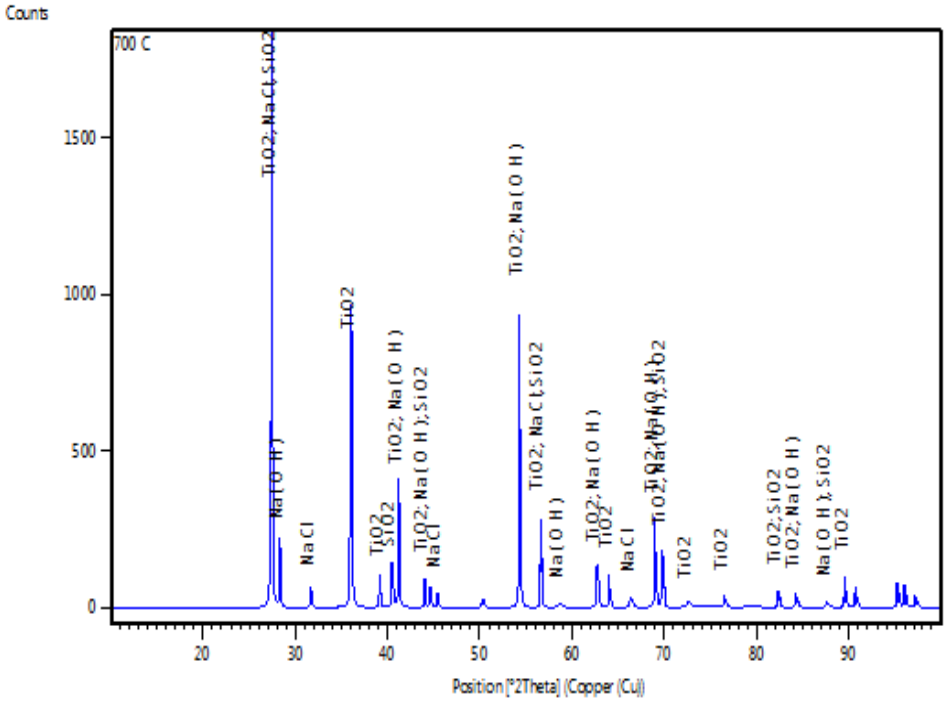

Figure 4. XRD Results of Calcination Temperature 700°C

The addition of SiO_2 to the TiO_2 composite causes the peaks to widen slightly towards the left so that the SiO2-TiO2 composite has an amorphous crystal form. The resulting peak in the synthesized composite shows the phase formed, namely: anatase and rutile. On SiO_2 formed quartz and cristobalite [12]. As referred from the Joint Committee on Powder Diffraction Standards (JCPDS card number: anatase, 21-1272 and rutile, 21-1276). At low calcination temperatures (<500°C), broad XRD peaks are observed due to the amorphous structure of TiO_2 nanoparticles. With increasing calcination temperature (>500°C), the XRD peaks continue to narrow and sharpen. This may be due to the elimination of grain boundary defects during calcination at high temperatures, which therefore increases the crystallinity of the TiO_2

nanoparticles [13]. This indicates that the composite contains both oxides that form it. TiO_2 nanoparticles exist in three polymorphic phases, namely rutile, anatase, and brookite. The low-density anatase phase is less stable and undergoes a transition to rutile, accelerated by calcination in the temperature range of 650°C to 1200°C [14]. It is concluded that the higher the calcination temperature, the change in the crystalline phase of the SiO_2-TiO_2 composite and shows peaks associated with the crystalline phase.

Analysis of the phase structure of the photocatalytic is by using XRD. XRD diffractogram can also show the presence of TiO_2 that has been buffered on SiO_2. Based on the XRD test results, silica at 700°C has a low quartz phase. This can be seen based on the peaks formed which indicate that the sample has a quartz low phase with a trigonal crystal system. The amorphous phase in silica that is not very visible is caused by the presence of a more dominant NaCl halite impurity phase, so that many peaks belonging to NaCl impurities are visible. Where NaCl has a crystalline phase so that the amorphous phase of the silica itself shifts and is not too visible low. This is because when washing the sodium silicate gel with distilled water, not all NaCl salts are wasted [15]. The results of XRD characterization can be viewed in Figure 4. ICDD data with reference code 01-085-0459 silica crystals are in the quart low phase with a hexagonal structure formed at 700°C with $\alpha = \beta = 90^o$ and $\gamma = 120°$, while on TiO_2 calcined at 700°C there began to be a phase transformation, where the anatase phase began to change into a rutile phase. In this process, the deposition results formed with diffraction angles of 27.4093°, 36.0296°, 44.6249°, 54.2768°, 68.9603°, 76.4835°, 89.4994°, and 95.1858°.

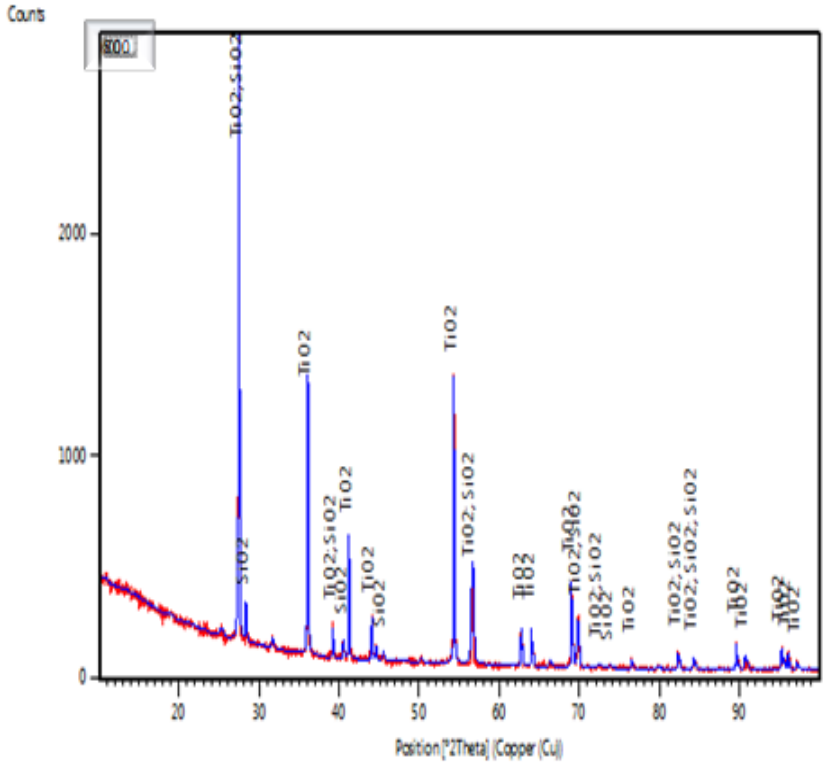

Figure 5. XRD Results of Calcination Temperature 800°C

XRD results in Figure 5 calcination temperature 800°C can be seen in Figure there are sharp diffraction peaks. Miller index [011] is produced at the highest peak with angle 2Θ = 27.324º for quart silica. According to ICDD data with reference code 01-083-0540 silica crystals are in the low quart phase with a hexagonal structure formed at 800°C with α = β = 90º and γ = 120°. The TiO2 phase formed turns into a rutile phase with JCPDS data: rutile, 21-1276 a phase that is stable at high temperatures and the SiO_2 phase is still not whole, so at this temperature γ is still formed with a value of 120º and this determines that SiO_2 has not become one phase with TiO_2. In this process, the deposition results formed with diffraction angles of 27.4193°, 37.1296°, 44.6249°, 56.3876°, 69.9653°, 82.6835°, 84.4764°, 90,2761°, and 95.1858°.

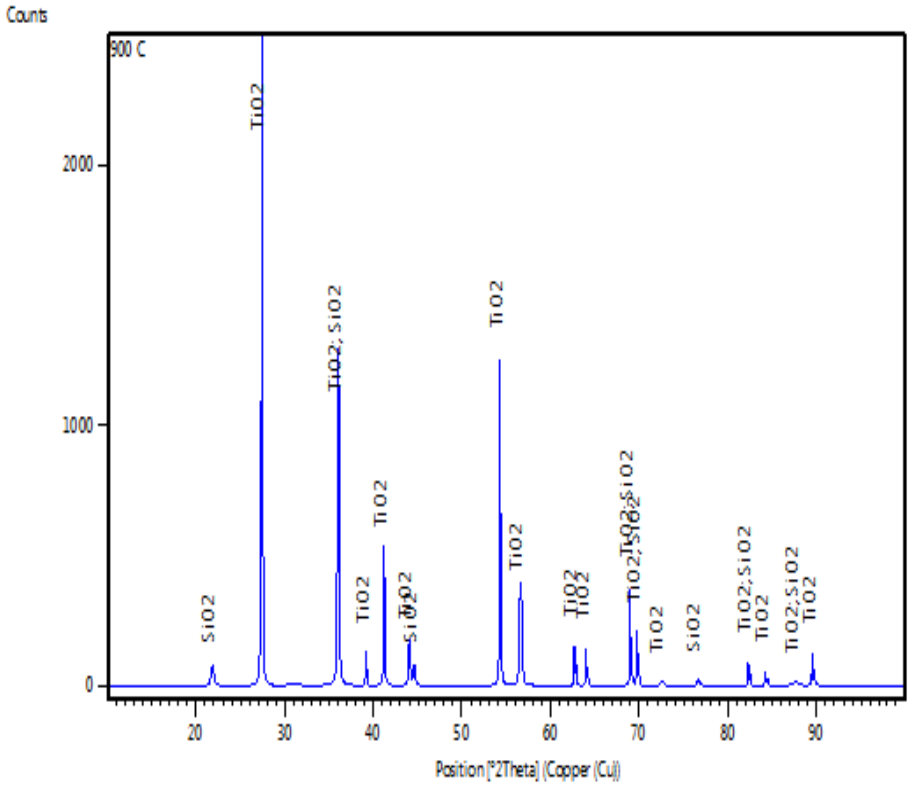

Figure 6. XRD Results of Calcination Temperature 900°C

The XRD results of silica at a calcination temperature of 900°C can be seen in Figure 6. In Figure 6, there are sharp diffraction peaks. The miller index [101] is produced at the highest peak with an angle of 2Θ = 21.840º for low cristobalite silica. According to ICDD data with reference code 01-083- silica crystals are in the cristobalite low phase with a tetragonal structure formed at 900°C with α = β = γ = 90º. The sample calcined at 9000C shows a phase transformation that occurs perfectly in SiO_2. At this temperature γ surah changes entirely into α phase with the same value of 90º. So that in samples calcined at this temperature there is only one phase for TiO_2 and SiO_2, namely the rutile phase. In this process, the deposition results formed with diffraction angles of 27.3925°, 39.0296°, 44.0191°, 69.7383°, 84.2057°, 87.5294°, and 89.1858°.

From the picture of the results of phase analysis on the effect of calcination temperature variation temperature 700℃, 800℃, and 900℃ at calcination temperature and 700℃ there are anatase and rutile and quartz phases until at calcination temperature 800℃, and 900℃ formed rutile phase and the formation of crystobalite phase. This indicates that phase transformation occurs due to the effect of additional calcination temperature. At high calcination temperatures can increase the distribution of SiO_2. SiO_2 will move into the TiO_2 crystal structure lattice and cause the rearrangement process of TiO_2. This rearrangement process will affect the change in the distance from the Ti-O-Ti and O-Ti-O bonds to be longer, resulting in a change in the anatase phase to rutile. Analysis of the combined images 2-4 of XRD characteristic data using High Score Plus Origin Lab 2023 software on the effect of calcination temperature obtained the data plot in figure 5.

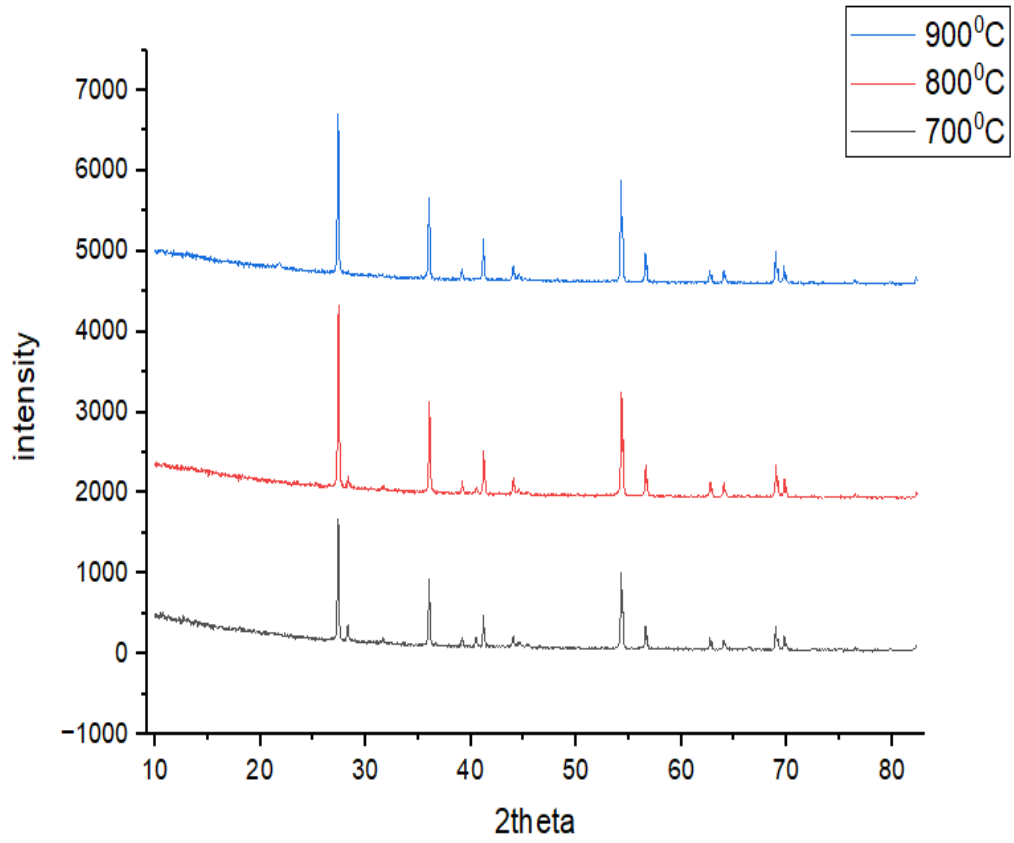

Figure 7. XRD Results of Calcination Temperature 700°C, 800°C and, 900°C

The results of the analysis using XRD obtained an array of lines or peaks with different intensities and positions that are specific to the material being analyzed. Each phase has a typical array of diffraction patterns containing intensity prices (counts per second) with a diffraction angle of 2Θ (in degrees). Determination of the crystal structure formed is done by matching each peak that appears in the diffraction pattern on the analysis results with data from JCPDS (Joint Committee Powder Diffraction Standards) so that information on the orientation of the crystal planes formed is obtained. If all crystal plane orientations are identified, it can be ascertained that the crystal structure has a match. X-ray diffraction can be used to determine the crystal size (crystallite size) with a particular phase [16]. Crystal type is one of the parameters to analyze the

composites that have been produced both qualitatively and quantitatively to identify the type of crystal formed, the sample characterization is carried out with XRD.

XRD diffraction patterns can also determine the crystal size. The crystal size is calculated using the Scherrer equation. The results of crystal size calculations of silica and titanium with calcination temperature variations of 700℃, 800℃, and 900℃ can be viewed in Table 1.

Table 1. Crystal sizes of SiO_2-TiO_2 composite

Calcination temperature (℃)	Phase TiO2	Phase SiO2	Crystal size
700℃	Rutile Anatase	Quartz low	63.48nm
800℃	Rutile	Quartz	66.87nm
900℃	Rutile	Cristobalite low	73.83nm

Based on table 1 the results of phase analysis on the effect of calcination temperature variation temperature 700℃, 800℃ and, 900℃ at calcination temperature 700°C there are anatase and rutile and quartz phases until at calcination temperature 800℃, and 900℃ formed rutile phase and the formation of quartz and crystobalite phase. This indicates that phase transformation occurs due to the effect of additional calcination temperature. At high calcination temperatures can increase the distribution of SiO_2. SiO_2 will move into the TiO_2 crystal structure lattice and cause the rearrangement process of TiO_2. This rearrangement process will affect the change in the distance from the Ti-O-Ti and O-Ti-O bonds to be longer, resulting in a change in the anatase phase to rutile. The results of crystal size analysis on the effect of calcination temperature variation temperature 700℃, 800℃ and, 900℃ respectively are 63.48nm, 66.87nm, and 73.83nm Research conducted [17] on SiO_2-TiO_2 nanocomposites in the absence of precursors found that the phase formed is the anatase phase because the calcination temperature used by researchers is 300℃ - 500℃ with a crystal size of 10-20nm. From this study it can be identified that the rutile phase can be formed at temperatures> 600℃, a stable rutile phase and a larger crystal size associated with the efficiency of the photocatalytic process and can be used in applications that require resistance to high temperatures. The use of precursors can affect the phase change as well as prevent the formation of unwanted phases.

4. Conclusion

Research that has been done by utilizing SiO_2-TiO_2 nanocomposites at various calcination temperatures obtained crystal sizes are 63.48 nm, 66.87 nm, and 73.83 nm. Crystal size can affect the stability and changes in the phase where the calcination temperature at 900℃ will transition into a more stable phase where in TiO_2 the formation of rutile phase and in SiO_2 the formation of cristobalite phase along with increasing crystal size and calcination temperature.

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