

Improvement of Thermal Properties for Binary Systems PVF-Ph, PVF-B by Addition of Silicon Carbide

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Received on: 4/12/2005

Accepted on: 10/6/2007

Abstract

Two binary systems of (PVF-Ph and PVF-B) were prepared at specific conditions, and then thermal properties were improved by addition of a ceramic material (SiC).

A different mixing ratio of ceramic material (SiC) powder was added to a prepared binaries (PVF-Ph, PVF-B) at (polyvinyl formal-phenol, and polyvinyl formal-bitumen) with a range (0.1-1)gm varied with (1-0.1)gm addition from both phenol (Ph) and bitumen (B).

The thermal properties (thermal conductivity, stability) were measured for both improved binary systems (PVF-Ph, and PVF-B), and the results proved that:

All thermal properties of two binaries (PVF-Ph, PVF-B) are modified with excellent properties of bitumen that will be raised from (50°C to 250°C) and other system (PVF-Ph) improved from (120°C to 200°C). Also the thermal conductivity is modified at a wide range from (0.0999 w/m°C to 0.000477 w/m°C.).

A home made thermogravimetric system was designed to give TG-analysis curve and check the stability of composite prepared system, where the results proved that the optimum mixing ratio of composite system gives high stability from (100°C to 315°C) with excess of (215°C) than base one (PVF-B).

تحسين الخصائص الحرارية للأنظمة الثنائية (PVF- Ph, PVF-B) بإضافة كاربيد السيليكون.
الخلاصة

تم تحضير نظامين بوليميريين من (بولي فانييل فورمال - فينول) و (بولي فانييل فورمال - بتيومين) عند ظروف التحضير المحددة ثم تم إجراء تحسين الخصائص الحرارية وذلك بإضافة مسحوق من مادة كاربيد السيليكون (مادة سيراميكية)، ونسب خلط مختلفة لتحضير النظام المتراب بنسب (١-٠,١) غم وذلك بتغيير نسب الفينول والبتيومين المضافة بحدود (١-٠,١) غم، بعدها يتم تهيئتها للفحوصات الحرارية تم في الخطوة التالية قياس الخصائص الحرارية (التوصيلية الحرارية والاستقرارية الحرارية) لكلا النظامين المحسنين (بولي فانييل فورمال - كاربيد السيليكون - فينول و بولي فانييل فورمال - كاربيد السيليكون - بتيومين) وتم الحصول على نتائج ممتازة لنظام (بولي فانييل فورمال - كاربيد السيليكون - بتيومين) والتي ارتفعت درجة استقراريتها الحرارية من (١٠٠ إلى ٣١٥ °م) أما نظام المتراب (بولي فانييل فورمال - كاربيد السيليكون - فينول) فتحسنت استقراريتها الحرارية من (١٢٠ إلى ٢٠٠ °م)، أما بالنسبة للتوصيلية الحرارية لكلا النظامين فقد تحسنت من (٠,٩٩٩ إلى ٠,٠٠٠٤٧٧ واط / م.م). وكذلك تم تصميم منظومه يدويه خاصه لقياس التوازن الحراري (تي جي) للأنظمة المترابيه المحضره (بولي فانييل فورمال - كاربيد السيليكون - فينول و بولي فانييل فورمال - كاربيد السيليكون - بتيومين) وقد أثبتت النتائج التوازن

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الحراري للنظام المتراكب (بولي فاينيل فورمال - كاربيد السليكون - بتيومين) عند (٣١٥ °م أكبر من ٢٠٠ °م) أعلى من النظام الاساسي (بولي فاينيل فورمال - بتيومين).

1- Introduction:

In order to fulfill the requirements of polymer industry many developers usually blend polymers together in order to reach an optimum balance of properties. This approach allows high flexibility in property adjustment and avoids development of new macromolecules which is generally long and expensive compared to polymer alloying [1] compatibilization is also help the dispersion of new media to give regular and stable morphologies [2, 3].

A polyvinyl formal (PVF) has high compatibility, partially or completely with other types of polymers or resins that permit improving the structure and properties of final products such as cellulos. Bitumen, urea, melamine and phenol by enhance mechanical, thermal and chemical properties for each other [4].

And silicone carbide (SiC) is one of the important refractory applied to widely for insulating the internal wall of furnaces due to more high thermal resistance at 1500°C rather than graphite with high compatibility to polymer type such as thermoplastic PVF or emulsion bitumen black [5, 6].

Both Stephaniel and Nesbitt in 1996 have studied the possibility of improving the thermal properties of Bitumen by the addition of polymers and resins[7]. Also Thomas Swan has studied the improvement of adhesion techniques and chemical properties of polymer by

additive of black emulsion Bitumen in [8].

Watford and Herts have studied the application of binary systems such as bitumen-polyethylene, bitumen – polystyrene to modify the chemical and thermal insulated properties [9].

And Hewietin has applied flexiphatte baryprene membranes as a composite membrane to concrete in order to enhance insulated thermal properties as chemical and thermal resistance to environmental severe conditions [10].

Recently case study has studied the application of a binary systems of film in metal laminate structure of (PVF- Ph and PVF – B) [11].

The aim of this work focuses upon modifying a thermal properties of both binary system of (PVF- Ph, PVF-B) by addition of ceramic powder (SiC) to enhance its thermal properties such as thermal conductivity, stability, also TG – analysis was designed in order to check the stability mechanism of composite system (PVF-SiC, and PVF-SiC-B).

2- Experimental:

2-1 Materials:

- a) polyvinyl formal commercially available by Monsanto company methods.
- b) Phenol crystal white laboratory material of high purity >99% coupling agent. supplied by BDH London.

- c) Emulsion bituminous resin (black waste coupling agent) of high penetration point at 120°C and 30 CST viscosity at 60°C supplied by Al-Durra Petroleum Refinery company for middle spot.
- d) Silicon carbide commercially available at-high purity >99% supplied by BDH.

Other reagents of chemicals were of general purpose grade from different sources

2-2 Preparation of Composite Systems:

The preparation of composite system were achieved by three steps, one step was preparing the first base binary system polyvinyl formal- phenol (PVF-Ph) where constant amount of base resin polyvinyl formal (2g) was added in glass container (250ml), and then coupling agent solution (10% H₃PO₃) was added with a continuous stirring by using of mechanical stirrer for 15min at 95rev/min to give homogenous mixture, afterward different mixing ratios of active site resin phenol were added (1,0.5,0.75) g/2g PVF with continuous stirring for 30min until all particles of both resins were dissolved to give high homogeneity binary system mixture (PVF-Ph).

Second step was achieved by adding fixed amount of base resin polyvinyl formal in glass container (250ml), then adding the coupling agent

solution (35% of organic solvent chloroform) with continuous stirring for 15min, afterward different mixing ratios of active site resin bitumen (1,1.5,1)g B/1g PVF were added with continuous stirring for 30min until reach high homogeneity emulsion mixture of (PVF-B).

The third step was concluded the addition of different mixing ratio of modified ceramics powders (SiC) for both prepared binary systems (PVF-Ph, and PVF-B) with continuous mixing for 15min to give homogeneity solid- liquid composite mixtures (PVF-SiC-Ph, and PVF- SiC-B) and both tables (1 and 2) shows the mixing ratios of both composite systems.

Then spread this prepared systems by brushing it equally in a specified dimension stainless steel molds of 3cm dia. and 1/2 cm thickness after oiling it by a Vaseline to simplify the exiting of compositing system products, afterward curing these molds product in an oven (0-250) °C at 120 °C and 100 °C for 1.5 hrs in order to give homogenous cured products then, it cools gradually to 70°C for first system (PVF-SiC-Ph).

And cools to 30°C for second composite systems of (PVF- SiC- B) afterward these molds were less dropped to give a homogenous composite products of softness faces [3].
cvaient where, both phenol crystal were milled to a fine particle size < 50µm products of softness faces [3].

Table (1) mixing ratio for PVF- SiC- Ph Composite system

Sample no.	PVF (gm)	Phenol (gm)	Sic (gm)
1	2	1	0
2	2	0.5	0.5
3	2	0.75	0.25

Table (2) mixing ratios for PVF-SiC- B composite system

Sample no.	PVF (gm)	Bitumen (gm)	Sic (gm)
1	1	1	0
2	1	1.5	0.25
3	1	1	0.5

2-3 Testing of composite system

This stage was achieved by three steps firstly, by thermal conductivity test, stability test and finally by the TG-analysis curve in order to check their thermal insulation and stability properties.

2-3-1 Thermal conductivity

This type of test was achieved by using Lee disk (Takasago power supply) method [12] due to its designer through the use of instrument found in the polymer lab of material engineering department

A specified specimens dimensions were prepared of 3cm dia. and ½ cm thickness then put this specimen between two disk manufactured from copper then passing a specified current and voltage then check its temperatures of specimen was located between two copper disk by thermometers until reach failure then , record these temperatures T_1, T_2 after ward apply the two equations below in order to calculate the coefficient of thermal conductivity k.

$$IV = r^2 e (T_1 + T_2) + 2re [d_1 T_1 + ds/8(T_1 + T_2) + d_2 T_2 + d_3 T_3] \dots (1)$$

$$-k (T_2 - T_1 / ds) = e [T_1 + 2(d_1 + 1/4 d_2) T_1 + 1/2r d_2 T_2] \dots (2)$$

where:

k= thermal conductivity coefficient w/m.°C.

T_3, T_2 = thermometer temperature at equilibrium °C.

T_1 = the first disk temperature °C.

ds= sample thickness cm.

e= energy loss.

r= radius of sample.

d_1 = thickness of first disk.

d_3, d_2 = thickness of second disk.

2-3-2 Thermal stability

This test is achieved by the application of the same specific dimension samples for two composite systems PVF-SiC-Ph and PVF-SiC-B by using an oven (0-250°C) of heating cycles where, the temperature allows raising to 50°C each 30min for each cycle until the failure of sample

occurred by deflection, temperature which is recorded in °C.

2-3-3 TG-analysis

This test was achieved by the use of a home made system to suit our study of weight loss for bulky samples of composite material systems.

This thermogravimetric system was applied under a nitrogen flow at $0.35 \text{ m}^3/\text{hr}$, where the sample was placed in a stainless steel basket of (5 cm dia. And 20 mm length). This basket was hung to the bottom of semi micro balance (0.0000) gm by 100cm stainless steel wire as shown in figure (1).

The temperature was measured at a specified point along the reactor of test (center wall), during constant time interval each (10) min, both temperature and weight loss were recorded in order to investigate the TG-analysis curve and stability for each systems (PVF-SiC-Ph, PVF-SiC-B).

4-Results and Discussion:

4-1 Composite systems preparation

During the preparation of composite systems (PVF-SiC-Ph, PVF-SiC-B), it can be seen that the second system of PVF-B is more compatible than other PVF-Ph at different weight ratios of ceramic additive SiC, due to chemical structure straight series of PVF-B, than cyclic chemical structure of PVF-Ph, the other case fine particle size and oily appearances of bituminous Ph that required limited condition of temperature and pressure as shown in equation below.

4-2 Testing thermal properties

4-2-1 Thermal conductivity

Figure (2) shows the effect of silicon carbide powder on the values of thermal conductivity of base binary system (PVF-Ph, and PVF-B) compared with composite improvement systems (PVF-SiC-Ph, PVF-SiC-B), where an increasing amount of silicon carbide added (0-0.5) g caused a decreasing of thermal conductivity values for both binary systems (PVF-Ph, PVF-B) with preference of (PVF-SiC-B) composite system due to more compatibility between bitumen coupling agent and silicon carbide.

Also high percent of oily carbon black appearance (>55 % wt.) and fine particle size (<50 μm), gave high compatibility and insulated properties for composite system (PVF-SiC-B), which reached (0.000477 w/m.°C.) [6, 13, 14].

4-2-2 Thermal stability

Figure (3) shows the effect of silicon carbide on stability of prepared composite systems (PVF-SiC-Ph, and PVF-SiC-B), where an increasing the amount of silicon carbide caused an increased the stability resistance of composite systems preference for (PVF-SiC-B) system, due to high compatibility between silicon carbide and coupling agent (organic solvent) [6, 13, 14].

An optimum mixing ratio of both systems are sample 2, where PVF-SiC-Ph with (0.5/0.5) (SiC/Ph), reached optimum temperature at 335°C.

And PVF-SiC-B system of (0.25 /1.5) (SiC/B) ratio reached optimum

temperature at 200°C respectively as shown in figure (4).

4-2-3 TG-analysis

Figure (5) shows the stability between two binary systems (PVF-Ph and PVF-B) at its optimum mixing ratios (2/0.5, and 1/1.5)g PVF/gPh or B.

The result shows high stability of (PVF-B) system at temperature range (300-315)°C than other system (PVF-Ph) that gave thermal stability at 228°C only due to the high content of carbon black content in bitumen system (>55 % wt.).

Figure (6) shows the effect of silicon carbide addition on the thermal stability of PVF-Ph system, where the (0.5/0.5) (SiC/Ph) of silicon carbide additive was result as optimum mixing ratio.

And the results indicated that the composite system gave high stability than base binary system of PVF-Ph at 300°C and 228°C respectively due to high thermal resistance of SiC at 1500°C[5, 6].

Figure (7) shows the effect of silicon carbide addition on the stability of binary system (PVF-B) at its optimum ratio of (0.25/1.5) (SiC/B).

And the results stated than an optimum mixing ratio of composite system (PVF-SiC-B) gave high stability than base system (PVF-B) at 315°C with excess of 215°C, due to high compatibility between Sic and black bituminous binder.

Also high content of CB (> 55% wt.) that characterized it was improved the insulated properties of produced

composite system (PVF SiC –B) [6, 13, 14].

Figure (8) shows the stability of composite systems indicates a comparison between two composite systems of (PVF-SiC-PH and PVF-SiC-B) as TG-curve at its optimum ratios (2/0.5/0.5, and 1/1.5/0.25).

The results gave high stability at (300-315)°C for (PVF-SiC-B) than other system (PVF-SiC-PH) that gave a thermal resistance at 228°C due to high content of CB (> 55%wt) in bitumen than others system of cyclic chemical structure[13, 14].

Also indicated that high content of CB percent (> 55%wt) in bitumen black waste support the compatibility with silicon carbide additive than other system (PVF-PH) as shown in equation 1 and 2 in section 4-1[11, 12].

5- Conclusions

1- All composite samples gave high thermal resistance in addition to high insulated properties at high temperatures by the additive of Sic ceramic material.

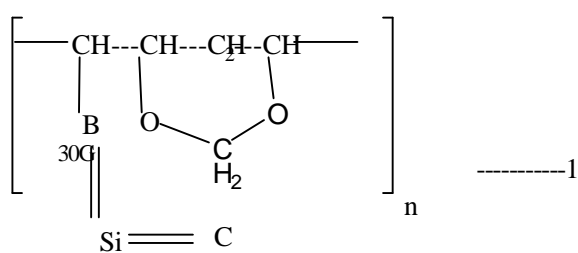
2- Optimum mixing ratio of high stability are 2 of (0.25/1.5) (Sic/B) and (0.5/0.5) (Sic/Ph) ratios respectively to give high stability at 300°C and 315°C.

3- Composite system of PVF-Sic-B gave high stability and high thermal resistance than PVF-Sic-Ph with excess 15 degrees.

4- Bituminous black waste binder gave high compatibility to Sic than other binder of white crystal phenol due to high carbon black content fine particle size, and oily natural which simplify this additive homegionty

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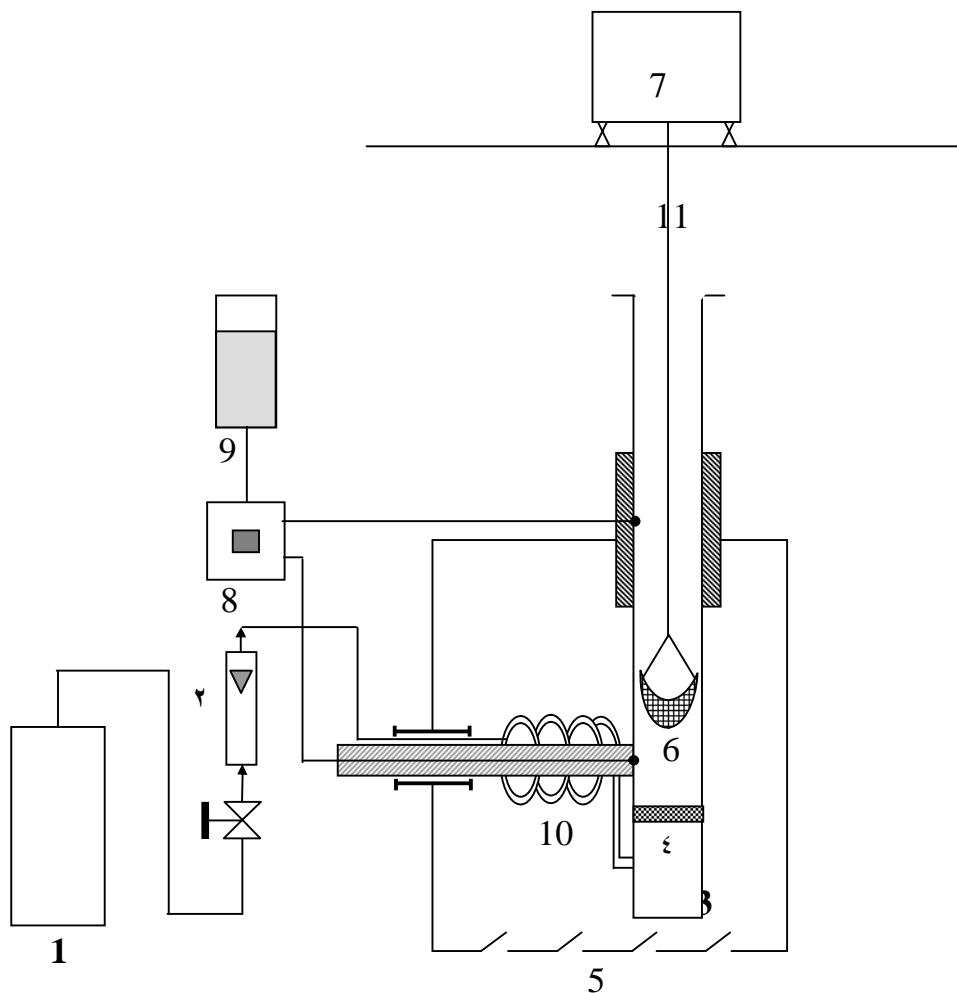


Figure (1) Experimental apparatus for TG-analysis studies.

- 1- gas source with regulator. 2-gas rotameter . 3- stainless steel reactor .
- 4- gas distributor . 5- electrical furnace. 6- stainless steel basket. 7- sensitive balance. 8- selector switch . 9- digital thermometer. 10- stainless steel coil .
- 11- stainless steel wire.

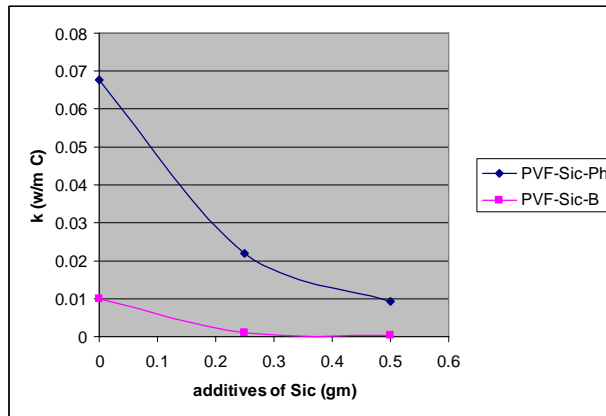


Figure (2): effect of silicon carbide additives on thermal conductivity coefficient of composite systems.

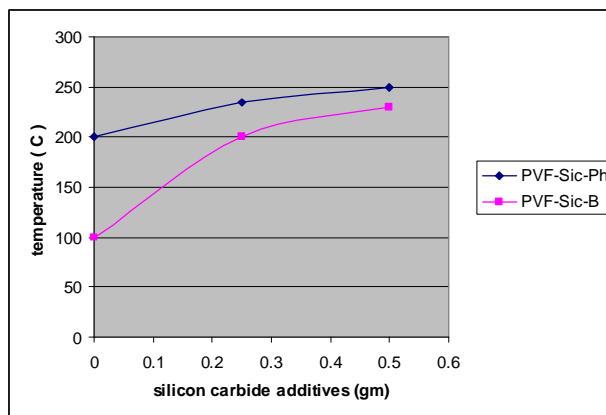


Figure (3): the effect of silicone carbide on thermal stability of composite systems (PVF-SiC-Ph, PVF-SiC-B).

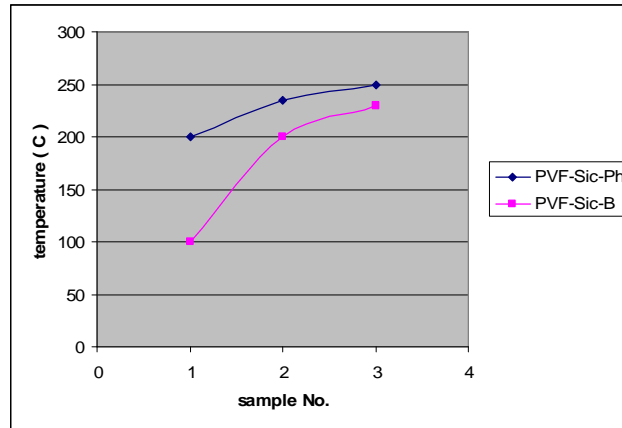


Figure (4): shows the comparison between two composite systems (PVF-SiC-Ph, PVF-SiC-B) for thermal stability test.

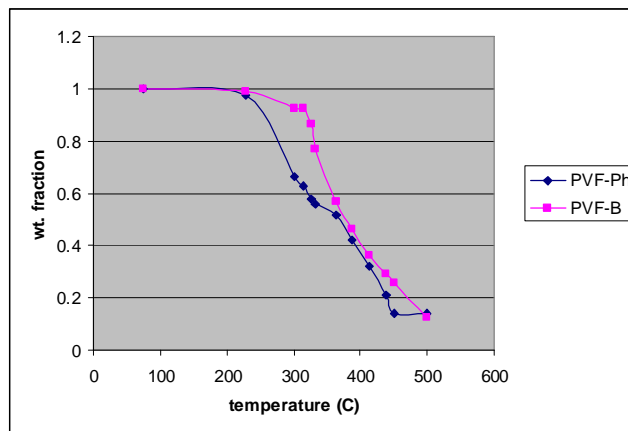


Figure (5): TG-curve analysis between two base binary systems (PVF-Ph, PVF-B).

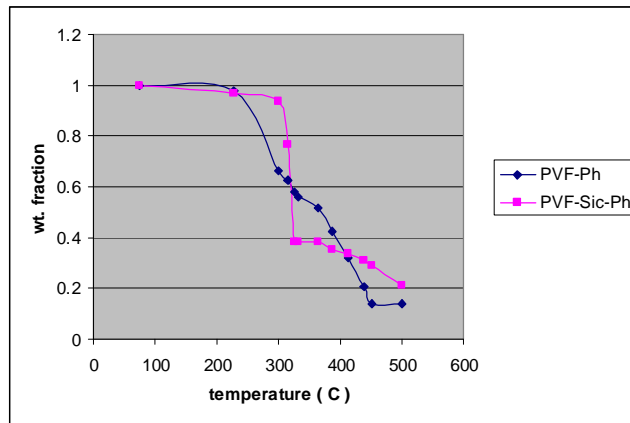


Figure (6): TG-curve analysis between base binary system (PVF-Ph) and composite system (PVF-SiC-Ph).

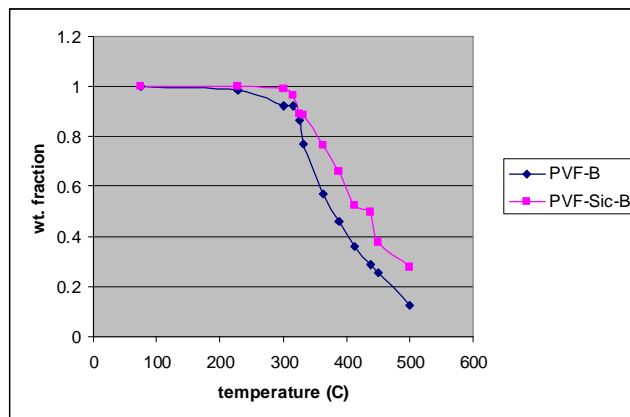


Figure (7): TG-curve analysis between base binary system (PVF-B) and composite system (PVF-SiC-B).

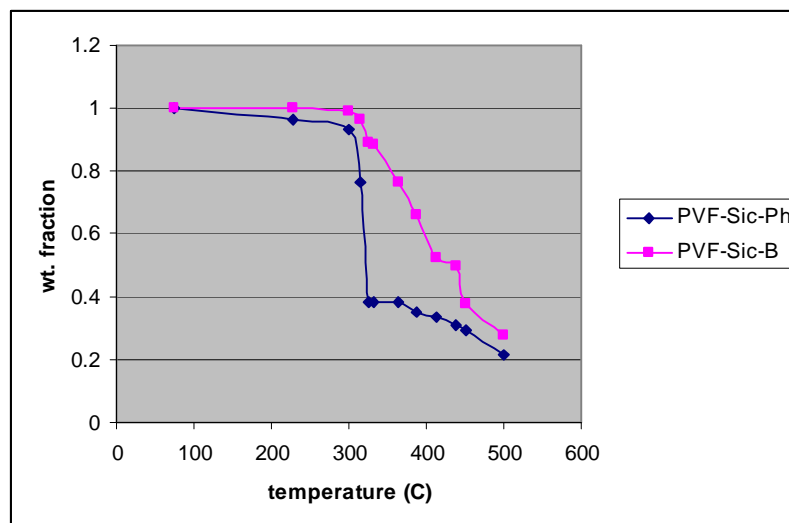


Figure (8): TG-curve analysis between two composite systems (PVF-SiC-Ph, PVF-SiC-B).