Preparation of Gypsum/ poly (Butyl Acrylate) Composites Using Gamma Irradiation

ZAKI AJJI*
Polymer Technology Division, Department of Radiation Technology, Atomic Energy Commission of Syria, P. O. Box 6091, Damascus, Syria

Gypsum/ poly(butyl acrylate) composites have been prepared based on natural gypsum and the monomer butyl acrylate by means of gamma irradiation. The conversion of polymerization was followed up with respect to the irradiation dose using thermogravimetric analyzer (TGA). The data show that yield of polymerization increases with increasing the irradiation dose. A yield of about 98% was achieved by exposure of the samples to a dose of 5 kGy. A thermomechanical analyzer (TMA) was used to locate the range of the glass transition temperatures (Tg) using the mode with alternative variable force; the mode with constant force was used to determine the Tg of the pure polymer and the composite prepared at the same irradiation dose. Also differential scanning calorimeter (DSC) was used to ensure the location of the glass transition temperatures.

Keywords: Gypsum; polymer composites; butyl acrylate; polymerization; radiation

In the past few decades, gypsum-based renders and plasters have become the material of choice for indoor finishing in many countries. Excellent performance, attractive appearance, easy application, and its healthful contribution to living conditions have made gypsum a most popular finishing material for centuries [1–3]. This building material is abundant in Syria, and also in Turkey [4]. The natural gypsum rock reserves of Turkey are estimated to be about 1.2 billion tons [4]. Availability, the relatively low level of start-up investments, and a favorable market situation, all provide conditions for growth and the profitable industrial production of gypsum-based materials [5]. The majority of gypsum-based composite materials can be specified within the following groups: plasters and renders, adhesives, jointing/filling compounds.

By incorporation of special fillers into a polymer matrix, desired composite properties could be achieved to suit various applications [6]. In the rubber industry, finely divided fillers have already been utilized to modify the polymer properties. Carbon black has been used for a long time [7-8]. Silicas are also used to modify polymeric materials [9-11]. Gypsum and alumina were also used as filler for preparing polymer composites [12-14].

The present work reports the preparation of composites consisting of butyl acrylate and commercially available gypsum using gamma radiation.

Experimental part

Natural gypsum powder was used, which is produced domestically from natural rocks in a mine near the city of Latakia in Syria. The sieved gypsum powder (0.3 mesh) was added to the liquid monomer step by step during stirring at room temperature so that a weight ratio of 1:1 was achieved.

The monomer/ gypsum/ mixtures were filled up in polyethylene ampoules, and then subjected to a ^{60}Co γ irradiator (Russian Gamma Cell, Type: Issledovatel); the irradiation was carried out at ambient temperature and a dose rate of around 3.5 kGy/h to different doses.

Thermogravimetric Measurements (TGA)

The dynamic weight loss tests were conducted using a Mettler instrument (TG50). The tests were carried out in a nitrogen or oxygen atmosphere, purged (30 mL/ min) using sample weights of 10-15 mg at a heating rate of 10°C/ min. The resolution of the balance is given, as 1 microgram for weights less than 100 milligram, and the temperature precision of the instrument is ± 2°C. The total experimental errors in the determination of the decomposition temperatures were estimated to be not more than ± 3°C.

Thermomechanical Analysis (TMA)

TMA was used to locate the glass transition temperatures of the prepared samples. A Mettler equipment (TMA 40) was utilized in order to record the TMA spectra. The used instrument has a precision of ±2°C, and the total experimental errors in the measurements were estimated to be about ± 3°C.

The PE container, used for the preparation of the samples, was removed and then sheets of ≈ 1mm were cut, polished, and cleaned. TMA spectra were recorded in two modes. The first mode was using an alternated force in order to ensure that there is a real glass transition; alternated force between 0.1 and 0.3 N was applied in most cases. The second mode was using a constant force of 0.1 N, and the T_g was determined from these TMA thermograms using the on-set method.

Differential Scanning Calorimetry (DSC)

DSC spectra were recorded using a Setaram instrument (type DSC131) in order to determine the glass transition temperatures of the samples. All samples were tested in aluminum pans at a heating rate of 10°C/ min over a suitable temperature range. The precision of the used instrument is ± 0.2°C, and the experimental errors in the measurements were estimated to be about ± 0.5°C.

Results and discussions

Thermogravimetry

Natural gypsum powder was used without drying because it has been previously showed that there is no significant influence of drying on the polymerization conversion by preparing polymer/ gypsum/ composites. Furthermore, the polymerization yield using chemically pure calcium sulfate powder and natural gypsum powder seems to be very similar [13].

* email: scientific@aec.org.sy

Various mixtures of butyl acrylate and gypsum powder were exposed to different Gamma doses. Figure 1 shows two typical TGA thermograms of mixtures, which are irradiated at different doses; the first step in the thermogram corresponds to the evaporation of the monomer in the samples; the second step corresponds to the decomposition of the polymer. The yields of polymerization are represented in figure 2 versus the irradiation doses. The polymerization conversion increases with increasing the irradiation dose, and achieves a value of about 98% using an irradiation dose of 5 kGy. This can be explained that the radical concentration, which acts as initiator, increases with increasing the irradiation dose and thus the reaction conversion.

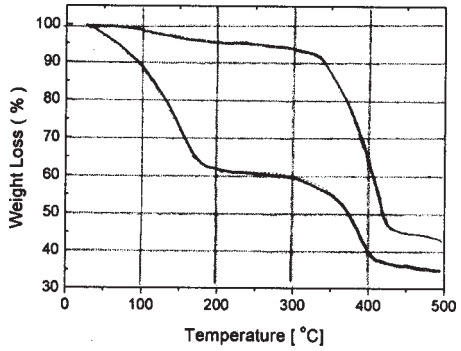

Fig. 1 Typical TGA thermograms of butyl acrylate/ gypsum/ composites for two different preparation doses

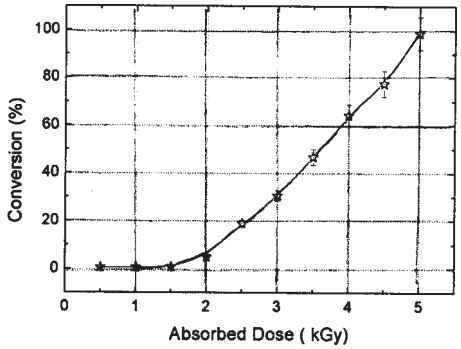

Fig. 2. The yield of polymerization of butyl acrylate versus the irradiation dose for the butyl acrylate/ gypsum composites

Glass Transition Temperature

TMA spectrum has been recorded using an alternated force of 0.2 N in order to ensure that there is a real glass transition and to show the *Tg* region as represented in figure 3; it can be seen that the elongation increases significantly at the Tg region. In order to determine the Tg, TMA spectra were recorded with constant force, and the Tg was determined from these thermograms using the on-set method. The Tg of poly(butyl acrylate) and poly(butyl acrylate)/ gypsum/ composite were -60°C and -56.1°C, respectively. DSC spectra were also recorded for pure poly(butyl acrylate) and poly(butyl acrylate)/ gypsum/ composite in order to compare and ensure their glass transition temperatures as shown in figure 4. It can also be seen that the glass transition temperature of the poly (butyl acrylate)/ gypsum/ composite is higher than the glass transition temperature of poly(butyl acrylate); $T_{on\text{-}set}$ and T_{mid} of pure poly (butyl acrylate) are -55.68 and -50.4°C, respectively. $T_{on\text{-}set}$ and T_{mid} of poly(butyl acrylate)/ gypsum/ composites are 53.74 and 48.71°C, respectively.

This difference is most probably due to the interaction between the polymer matrix and filler particulates, which reduces the segmental mobility of the chains near the filler

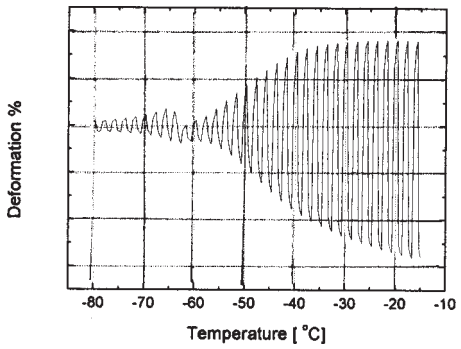

Fig. 3. TMA spectrum with alternated force of 0.2 N for poly (butyl acrylate)/ gypsum/ composite irradiated at 5 kGy

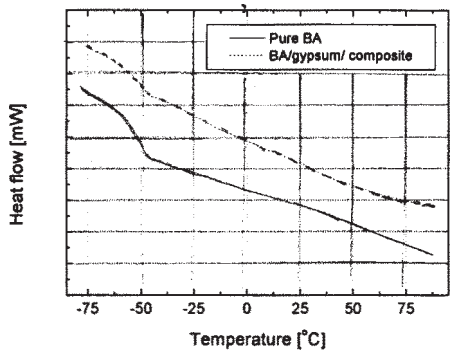

Fig. 4 . DSC spectra of poly (butyl acrylate) and poly (butyl acrylate)/ gypsum/ composite (irradiation dose = 5 kGy)

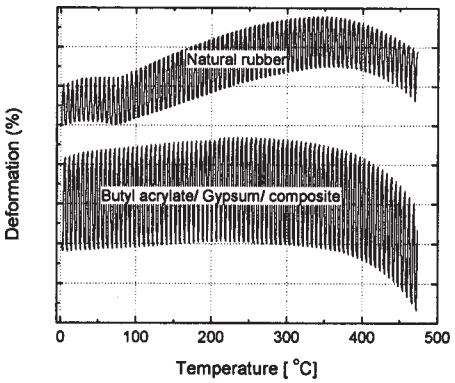

Fig. 5. TMA spectra of poly (butyl acrylate)/ gypsum/ composite and butadiene rubber with variable alternated force

particles. This behavior has also been observed and documented in other composite systems, and has been explained on the basis of reduced mobility of molecular segments in the vicinity of the filler particulates [12].

TMA spectra with alternated force of 0.2 N were recorded for butadiene rubber and poly (butyl acrylate)/ gypsum/ composite in order to compare their elasticity. The spectra showed that the elongation of poly (butyl acrylate)/ gypsum/ composite is higher than that of butadiene rubber. Furthermore, this elastic behavior of both polymer systems retains up to a temperature over 400°C.

Conclusion

Polymer/ Gypsum/ composite / poly (butyl acrylate) composites have been prepared based on natural gypsum and butyl acrylate by means of gamma irradiation. The conversion of polymerization was followed up with respect to the irradiation dose using thermogravimetry. The data show that yield of polymerization increases with increasing

the irradiation dose. A yield of about 98% was achieved by exposure of the samples to a dose of 5 kGy. Also the glass transition temperatures have been determined for the pure polymer and composites prepared at the same irradiation dose.

Acknowledgement: The author would like to thank Mrs. M. K. Aljoumaa, A. Mougrabya, H. Alkassiri, and Z. Daoud for their effort during the experiments.

References

1.ARPE H..J., Ullmann's Encyclopedia of Industrial Chemistry, Calcium Sulfate, vol. A4, Wiley-VCH, Verlag, Germany, 1984, p. 555
2.DUGGAL, S. K., Building Materials, A.A. Balkema Publishers, Rotterdam, 1998, p. 368
3.RAGSDALE, L.A., RAYNHAM E.A., Building Materials Technology, Edward Arnold, London, 1972, p. 243
4.ARIKANA, M., SOBOLEV, K., The optimization of a gypsum-based composite material, Cement and Concrete Research, **32**, 2002, p. 1725
5.TOPKAYA, M., KARAYAZICI F., ORAL, I., SEZEN, S., Chemical Investigation on Gypsum Rock and Anhydrite, Kimya, **23, nr.** 2, 1980, p. 11
6. KAT,Z H., MILEWSKI, J. V. eds., Handbook of Fillers and Reinforcement for Plastics, Van Nostrand, New York, 1978
7.YANG, H., TANG H., PIAO, J., CHEN-XINFANG, The stabilization effect of radiation crosslinking on positive temperature coefficient performances of carbon black-polymer composites, Radiation Physics and Chemistry, **42. nr.** 1-3, 1993, p.135
8.SHALTOUT, N. A., ABOU-ZEID, M. M., MOUSTAFA M,. H, EL-MILIGY A,. A., Effect of Gamma Irradiation and Degree of Loading of Carbon Black on the Properties of Styrene-Butadiene Rubber, Arab-Journal of Nuclear Sciences and Applications, 2002, **35** 1, p. 25
9.ÇAYKARA, T., GUEVEN, O., The Effect of preparation methods on the thermal properties of ploy(acrylic acid)/ silica composites, Journal of Applied Polymer Science, **70, nr.** 5, 1998, p. 891
10.ÇAYKARA, T., EROGLU, M,. S., GUEVEN, O., Thermal and oxidative degradation of vinyl triethoxy silane-methyl methacrylate copolymers, Polymer Degradation and Stability, **63**, 1999**,** p. 65
11.ÇAYKARA, T., GUEVEN, O, Kinetic analysis of the radiation induced polymerization of an acrylic acid-silica system, European Polymer Journal, **35**, 1999, p. 113
12.ÇAYKARA, T., GUEVEN, O., Effect of preparation methods on the thermal properties of ploy(acrylic acid)/ alumina composites, Polymer Composites, **19,** nr. 2, 1998, p. 193
13.AJJI, Z., ALKASSIRI ,H., Preparation of gypsum/ polymer/composites using Gamma radiation, In Proceedings of the Symposium on Radiation Technology in Emerging Industrial Applications, Beijing, China (2000), 2003 p. 41-45 (IAEA, TecDoc 2003)
14.AJJI ,Z., Preparation of polyester/gypsum/composite using gamma radiation, and its radiation stability, Radiation Physics and Chemistry, **73,** nr. 3, 2005, p. 183

Manuscript received: 30.12.2008