Urea–Formaldehyde Crosslinked Starch and Guar Gum Matrices for Encapsulation of Natural Liquid Pesticide [Azadirachta Indica A. Juss. (Neem) Seed Oil]: Swelling and Release Kinetics

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ABSTRACT: Polymeric granules were prepared by matrix encapsulation containing 20, 35 and 50% (w/w) of natural liquid pesticide viz., Azadirachta Indica A. Juss. (neem) seed oil (NSO) per dry weight of urea formaldehyde crosslinked starch (UF-St), guar gum (UF-GG) and UF-(St + GG) matrices. Results of swelling and cumulative release kinetics are presented at 35°C for these matrices. The static dissolution experiments have been carried out at 35°C for seven days. The percentage cumulative release kinetic data have been analyzed using an empirical equation to study the release pattern of NSO through the polymeric membranes employed. Transport follows the Super Case II mechanism as tested by an empirical equation. It is found that the release of the active ingredient depends upon the type of the matrix and its swelling ability. The percentage loading of NSO with different matrices and their density exerted an influence on the release data. The FTIR results indicated the absence of chemical interactions between the matrices and the NSO. In the majority of cases, entrapment efficiency was generally more than 95% indicating the efficient encapsulation. Furthermore, the experimental results are discussed in terms of the nature and the combined effect of the individual matrices as well as the percentage loading of NSO. © 1999 John Wiley & Sons, Inc. J Appl Polym Sci 73: 2437-2446, 1999

Key words: starch; guar gum; urea formaldehyde; neem seed oil

INTRODUCTION

Application of pesticides to plants/soil to control the pests affecting plants tend to produce hazardous effects to the environment. It thus becomes necessary to apply pesticides to the soil in order to control pests like flies, nematodes, and fungus because their part of the life cycle is in the soil. Leaching of the applied pesticide will pollute the surface/groundwater, which will ultimately result in the malfunctioning of the biological system after continuous and long-term exposure. Residual toxicity of the pesticide is another important factor that needs to be considered when the pesticides are applied to vegetables and fruits. Mulberry tree leaves are being used as food for silk worm. The pesticide used for these trees should

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have less residual toxicity in order to protect the silk worms. This can be achieved by the use of a naturally occurring, less-toxic pesticide, such as neem (*Azadirachta Indica A. Juss.*) seed oil (NSO).¹ This oil is produced by extracting the seeds of the neem tree.

Commercially, NSO is being used as a liquid spray to control pests like aphids, jassids, thrips, mites, helopelties, caterpillars, whiteflies, leaf beetles, hornworms, and gypsy moth, but it cannot be used in soil as it is in the form of a liquid. However, the cake coming after extraction of oil from the neem seeds is used by the farmer for soil applications. The main drawback of this cake application is frequency, and the quantum of application is more as it contains less amount of NSO. To circumvent this problem, we have undertaken the present research on the successful encapsulation of NSO to produce the solid formulation with a longer duration of action. In this direction, in our earlier article,² we have used many polymeric systems as membrane materials for the controlled release (CR) of NSO. In continuation of this program of research, we have now selected the compatible polymeric systems on the basis of cost, method of preparation, percentage of loading of NSO, release characteristics, and stability of the final product. A starch urea-formaldehyde matrix system obeys almost all the factors mentioned above except its CR characteristics. This problem has been overcome by adding a third substance like guar gum (GG) to produce a blend matrix system.

Starch (St) is a naturally occurring biodegradable polymer that has been widely used as an encapsulating matrix for agrochemicals after derivatization and crosslinking.³⁻⁸ A process for encapsulation of pesticides in a starch matrix using urea-formaldehyde (UF) prepolymer as the crosslinking agent has been exploited widely.³⁻⁵ Research on starch and modified starch as replacements for the petroleum-based polymers has resulted in several new technologies that are now being practiced commercially. These include a matrix-forming starch for the encapsulation of pesticides, graft copolymers as absorbents for seed coatings, and as root treatments for seedling transplants, starch-synthetic polymer composites as blown films for agricultural mulch, and starch-polyacrylate grafts as soil amendments.⁹

Guar gum is another useful natural polymer that has been used widely in the pharmaceutical literature as an excipient.^{10–12} It is a natural water-soluble polysaccharide, absorbing many

times its weight of water, and this property is responsible for its use as an excipient in pharmaceutical, cosmetics, and food product industries. However, to the best of our knowledge, UFcrosslinked starch and GG matrices in different combinations have not been attempted in the earlier literature for encapsulating NSO for CR application. The purpose of combining these polymers is to take advantage of the higher swelling capacity of GG than that of starch in water to optimize the release of the active agent through a judicially selected ratio of St and GG. The NSO is particularly chosen as a liquid pesticide because of its free availability from the extracts of the neem seeds and its lesser toxicity compared to other synthetic pesticides.¹³ In the market, several NSO liquid formulations are available, but no solid NSO encapsulated granules have been developed for the longer persistence.

In this research, we report the preparation and characterization of granules of St, GG, and St + GG physical mixtures at a weight ratio of 1:3, 1:1, and 3:1, respectively, crosslinked with the UF prepolymer containing 20, 35, and 50% (w/w) of NSO. The crosslinked granules were prepared at a ratio of 50% of urea per dry weight of the polymer in the previously taken urea-to-formaldehyde ratio of 1:2, respectively. Swelling data of the crosslinked individual polymers, as well as their physical mixtures, have been obtained at 35°C by a newly developed technique by measuring the water uptake. The static release data have also been obtained at 35°C by measuring the NSO concentrations at different time intervals using ultraviolet (UV) spectrophotometry. The percentage of cumulative results have been analyzed using an empirical equation for understanding the release mechanism of NSO through the barrier polymers. Furthermore, the results are discussed in terms of the nature of the matrices and their composition ratios.

EXPERIMENTAL

Materials

Neem seed oil was extracted from the neem seeds collected from neem trees in the southern zones of India, and this was of technical grade sample. Urea, guar gum (approximate molecular weight 220,000), maize starch (approximate molecular weight 81,000), formic acid, chloroform, Tween-80, and formaldehyde (37-41% w/v) solution were



Figure 1 Plot of absorbance versus concentration of NSO (in μ g/mL) in 0.1% Tween-80 solution.

all purchased from s.d. Fine Chemicals, Mumbai, India.

Methods

Calibration Curve of NSO Using UV Spectrophotometer

Calibration curve for the present systems is not available in the earlier literature; and, thus, we have attempted to estimate the amount of NSO present in the granules to determine the percentage of loading (entrapment efficiency) and the release rate in the eluting solvent medium. The quantitative estimation was done by using an UV spectrophotometer (Secomam, Anthelie, France). Known concentrations of NSO in distilled water containing 0.1% Tween-80 (0.1% of Tween-80 is sufficient to dissolve 1 g of NSO in 100 mL of water) were scanned in the range of 200-400 nm. For NSO concentrations ranging from 10 to 90 μ g/mL, a sharp peak was produced at 208 nm. The absorbance values obtained with the respective concentrations are plotted in Figure 1. From the straight line plot, slope (0.0094), and intercept (0.0634) were calculated from the least-squares method with the correlation coefficient, r equal 0.9980 within the limit of the Beer-Lambert law. The slope value was used to calculate the unknown concentration of NSO as equal to absorbance at 208 nm/(slope, that is, 0.0094). The absorbance data at 208 nm were highly reproducible.

Preparation of Crosslinked Granules

The crosslinked granules containing NSO were prepared in the following two steps: (1) urea-

formaldehyde (UF) prepolymer was prepared by mixing urea and formaldehyde (previously made alkaline with 10% NaOH solution) in the 1:2 molar ratio in a 500 mL beaker and refluxing it for 30 min in a water bath maintained at 70°C and (2) gelatinized St, GG, or St + GG matrices containing NSO were prepared by boiling the insoluble St, GG, or St + GG combinations with distilled water until the formation of a transparent muscilage. Soon after cooling, NSO (in three concentrations, that is, 20, 35, and 50% w/w of the polymer) was added and finely dispersed by mechanical mixing. This muscilage containing NSO was crosslinked with the previously prepared UF prepolymer. The pH of the mixture was reduced to 3 (i.e., acidic) by the addition of a few drops of formic acid so as to complete the crosslinking reaction. The mass thus formed was sieved through a 10-mesh ASTM sieve, and the granules were dried in a vacuum oven at 30°C under a pressure of 600-mm Hg overnight. The dried granules were sieved through a 44-mesh superimposed on 22-mesh (ASTM) sieve to obtain the uniform size granules with their sizes ranging between 0.5 and 2 mm.

Determination of Amount of NSO in Granules and Density of the Matrices

The amount of NSO present in the granules was determined by refluxing the granules with chloroform followed by extraction with 0.1%Tween-80 solution. This method is particularly adopted to enhance the release of NSO from the granules at higher temperature and in a good solvent, such as chloroform. About 10 mg of the granules were taken in a 250-mL, round-bottom flask and refluxed with chloroform for 30 min (below 70°C). After cooling, the granules were separated and used to determine density. The chloroform layer was extracted with a 0.1% Tween-80 solution and the UV absorbance was taken at 208 nm.

In order to determine the density of the waterfilled granules, the granules were removed from the refluxed medium incubated at 25°C for 24 h with distilled water taken in specific gravity bottles. The amount of water repelled by the granules was calculated as follows: weight of specific gravity bottle with water – (weight of the specific gravity bottle with water + granules – weight of the granules taken). The amount of water repelled was considered to be the volume of the water-filled granules. The density of the granules were then calculated as

St : GG Ratio	NSO (%)	Entrapment (%)	Water Content in Granules (%)	Density of Water Filled Granules (g/cm ³)
1:0	50	99.59	9.12	1.1750
1:0	35	98.76	10.18	1.1447
1:0	20	99.61	11.80	1.1230
0:1	50	98.31	6.32	1.6733
0:1	35	97.81	7.03	1.6464
0:1	20	99.10	7.32	1.5857
1:3	50	93.88	7.03	1.5967
1:3	35	97.61	7.72	1.5882
1:3	20	99.00	7.90	1.5140
1:1	50	98.01	7.81	1.4304
1:1	35	97.38	8.53	1.4139
1:1	20	98.91	9.38	1.3842
3:1	50	97.13	8.10	1.2656
3:1	35	98.82	8.91	1.1872
3:1	20	96.31	9.93	1.2275

Table IPercentage of NSO Loadings, Percentage of Water Content inGranules, Percentage of Entrapment, and Density of St/GG Matrices

follows: weight of the granules/volume of the granules. The results of density and NSO content are presented in Table I.

Swelling and Static Release Studies

Since swelling is directly related to the amount of water uptake by the samples, we have designed a new method to measure the amount of water uptake and thereby measure the extent of swelling of the granules. To do this, stainless steel mesh bags (mesh size 100) were fabricated with a hook at the top (see Fig. 2) to lift the bags from dissolution media at definite intervals of time. A



Figure 2 Schematics of the stainless steel seive bag used for static dissolution experiments.

weighed amount (about 100 mg) of the granules were taken in these bags kept in 500-mL conical flasks containing the dissolution media, that is, 0.1% Tween-80 solution in distilled water. A 10 mL of aliquot was taken at definite intervals of time from the dissolution media to analyze the release of NSO from the matrix spectrophotometrically. At the same time, the bags were removed from the conical flasks with the help of a thin steel rod and then weighed to measure the water uptake by the granules.

Fourier Transform Infrared Measurements

The Fourier transform infrared (FTIR) spectral data were taken on urea, starch, gelatinized starch, and crosslinked starch with urea formaldehyde containing NSO using FTIR (Nicolet, Model Impact 410, USA). About 2 mg of the samples were ground with KBr, and the pellets were made under a hydraulic pressure of 6 tonnes. The spectra were obtained by eliminating the background noise, and scanning was done from 4000- 500 cm^{-1} . The instrument uses a He–Ne laser (632.8 nm) as a equipment carrier with deuterated triglycine sulfate (DTGS) detector. The laser is used as an internal calibrator because it emits light at precise calibrated points that generate wave number with an accuracy better than ± 0.01 cm^{-1} . The laser controls the moving mirror's position and triggers the capture of data.

RESULTS AND DISCUSSION

The mean size ($\sim 2 \text{ mm}$) with a narrow distribution of particles was found for St, GG, and (St + GG)-UF matrices. The reason for this type of narrow distribution is attributed to 44-mesh superimposed on a 22-mesh size sieve. This is extremely useful in designing the dissolution experiments because the rate of dissolution depends upon the surface area of the particles. In the narrow range distribution of particles, the dissolution data are better reproducible than the wide range distributed particles. This is because dissolution is dependent upon the surface area of the particles exposed to the surrounding environment; and, therefore, the batches of the samples taken for dissolution study or even for field application should possess uniform sizes.

The percentage of entrapment data presented in Table I show that NSO is being encapsulated efficiently in all the matrices, irrespective of the type of the matrix material. In all the cases, the value of the percentage of entrapment efficiency exceeded more than 96, except in the case of 1:3 matrix, for which the efficiency was about 94%. Such higher values of entrapment efficiency might be the result of efficient entrapment of NSO by the crosslinked polymeric barriers employed. Among the two matrices studied, GG has higher density values than the St matrices. However, with increasing amounts of NSO in all the matrices, a decrease in density is observed. This might be due the fact that when NSO loading is high, this will subsequently hinder the crosslinking process, thereby decreasing the density of the matrices. With increasing amounts of GG in the binary polymer mixture (St + GG), density also increases because the density of the pure GG matrix is higher than that of St.

In this study, in order to maintain the optimum value of viscosity of the polymer solutions for a uniform distribution of NSO to get a higher percentage of entrapment efficiently after crosslinking, we have measured the dynamic viscosity as well as torque, shear stress, and shear rate for St, GG, and their binary dispersions before and after crosslinking using a Brookfield Rheometer, Model DV-III (made in the USA). The selection of the polymer dispersion concentration was made based on the accessible range of viscosities for the particular spindle used (SC 21). The measured values of viscosity, shear stress, shear rate, and torque for 5% St, 0.5% GG, and their physical mixtures (2.5% St + 0.25% GG) before and after

Table II Viscosity, Torque, and Shear Stress of
St, GG, and St + GG Mixtures Before and After
Crosslinking with UF (50%) at 100 RPM Spindle
Speed (at Shear Rate 93/s) at 303.15 K

Torque (%)	Viscosity (mPa s)	Shear Stress (N/m ²)
23.8	119.0	11.1
30.7	153.5	14.3
12.2	61.0	5.7
74.0	370.0	34.4
36.4	182.0	16.9
24.9	125.0	11.6
	Torque (%) 23.8 30.7 12.2 74.0 36.4 24.9	Torque (%) Viscosity (mPa s) 23.8 119.0 30.7 153.5 12.2 61.0 74.0 370.0 36.4 182.0 24.9 125.0

crosslinking at 303.15 K are presented in Table II. The results indicated that viscosity, shear stress, and torque measured for St, GG, and St + GG were increased after crosslinking with UF. NSO is a hydrophobic molecule; thus, it exhibits a poor solubility in water. However, with the addition of Tween-80, its solubility can be increased. Furthermore, its solubility was studied at various concentrations ranging from 0.05 to 0.25% of Tween-80 solution in water. With the addition of 0.05 wt % of Tween-80, the solubility of NSO increases nearly fivefold. However, the optimum concentration of Tween-80 selected in the present study was 0.1%, and this is sufficient to dissolve 1 g of NSO in 100 mL of 0.1% Tween-80 solution.

The static dissolution results at 35°C of the granules are expressed as cumulative release for different loadings of NSO and different combinations of the matrices. In all the cases, dissolution time for the experiments was kept up to 7 days. Figure 3 presents the cumulative release data of the crosslinked pure St and GG matrix with 20, 35, and 50% loadings of NSO. It is observed that the matrices containing 20% NSO show lower release rates than those containing 35%, which, in turn, exhibits the slower rates than the 50% NSO-containing matrices. In the case of St matrices containing 20 and 35% of NSO, the cumulative release curves vary almost identically, while the curve for 50% NSO loading deviated drastically from those of the other two curves. Also, the release data at 50% NSO increase very fast compared to those at lower loadings of NSO. In both the matrices, the sigmoidal shapes of the release



Figure 3 Plot of cumulative release versus time (in hours) for (A) St and (B) GG matrices with (\Box) 20% NSO, (\blacktriangle) 35% NSO, and (\bigcirc) 50% NSO in static dissolution media of 0.1% Tween-80 solution at 35°C.

curves are observed, indicating that the polymer matrices attain more of a rigid structure after crosslinking. This is further supported by the swelling data presented in Figure 4; that is, the swelling of the St, and GG matrices show a decrease with an increasing amount of NSO. The swelling curves of the St matrices vary linearly in the beginning and attain equilibrium within 2–3 days. However, in the case of GG matrices, the swelling curves are not so systematic and also these behave more sigmoidally than those observed for the St matrices. This is further indicative of the fact that the GG matrix becomes more rigid when compared to the St matrix after crosslinking. This also suggests that the GG polymeric chains tend to relax much slower than the movement (diffusion rates) of the water molecules.

The cumulative release data of the St and GG matrices in different combinations are presented in Figure 5. First of all, for the 1 : 1 combination of the matrices, the release rates are very different for all the loadings (20, 35, and 50%). A systematic increase in the release rates with increasing loadings of NSO was observed. On the other hand, for the matrices of St : GG in the ratio of



Figure 4 Plot of the percentage of water uptake (swelling) by the granules versus time (in hours) for (A) St and (B) GG matrices with (\Box) 20% NSO, (\blacktriangle) 35% NSO, and (\bigcirc) 50% NSO in static dissolution media of 0.1% Tween-80 solution at 35°C.



Figure 5 Plot of cumulative release versus time (in hours) for St : GG (A) 1 : 3, (B) 1 : 1, and (C) 3 : 1 matrices with (\Box) 20% NSO, (\blacktriangle) 35% NSO, and (\bigcirc) 50% NSO in static dissolution media of 0.1% Tween-80 solution at 35°C.

1:3, the release rates of 35 and 50% loaded NSO are almost identical; and, at the same time, these values are higher than those observed for the 20%

NSO-containing matrix. The same dependence is also observed in the case of St : GG matrix in the ratio of 3 : 1, but the release curves for the latter system vary within a narrow range when compared to the 1:1 system, for which a more regular and equidistant pattern for all the three loadings of NSO are observed. A close observation of the swelling results displayed in Figure 6 for different combination systems, that is, 1:1, 1:3, and 3:1, respectively, of the St : GG matrices reveals no systematic dependence for the 1:1 and 3:1matrices for different loadings of NSO. However, for the 1:3 matrix, we find that a distinct swelling behavior is observed; that is, an increase in the percentage of swelling occurred with a decrease in NSO loading.

The transport of NSO from the present granules could be explained on the basis of their matrix structure and their composition. Different matrices release to different extents, depending upon the nature and type of the matrix material. In any case, the mechanism of pesticide release from the matrices involves three steps, that is, water absorption, matrix swelling, and diffusion of the active agent from the swollen membrane polymers. Thus, the transport phenomenon in these systems involves the relative contribution of the diffusion process governed by a concentration gradient, as well as a relaxation process controlled by a time-dependent polymer response to a swelling stress. In all the cases, the in vitro release data show an increase with increasing pesticide loading in the matrices and that nearly 90% of the release occurred. However, with pure St matrices, the release is higher than that of pure GG. The release also shows an increase with increasing content of starch in the matrices. The addition of GG to St leads to a decrease in the release of NSO, probably due to the rigid structure of the crosslinked GG when compared to St matrix. Figure 7 displays the results from duplicate experiments to illustrate the reproducibility of the release data from 35% NSO-loaded St matrix.

The results of fractional amount of NSO released; that is, M_t/M_{∞} have been fitted to an empirical equation proposed by Ritger and Peppas.¹⁴

$$\frac{M_t}{M_{\infty}} = kt^n \tag{1}$$

This equation characterizes the diffusion mechanism by the parameter n. The constant k is a



Figure 6 Plot of the percentage of water uptake (swelling) by the granules versus time (in hours) for St : GG (A) 1 : 3, (B) 1 : 1, and (C) 3 : 1 matrices with (\Box) 20% NSO, (\blacktriangle) 35% NSO, and (\bigcirc) 50% NSO in static dissolution media of 0.1% Tween-80 solution at 35°C.

characteristic of the polymer-pesticide interaction. A typical plot of $\ln M_t/M_{\infty}$ versus $\ln t$ for St matrix is shown in Figure 8, which exhibits a straight line behavior, indicating the satisfactory fitting of our release data to eq. (1). Using the



Figure 7 Plot of cumulative release data obtained in duplicate versus time (in hours) for St matrix with 35% NSO in static dissolution media of 0.1% Tween-80 solution at 35°C.

least-squares procedure, we have estimated the values of n and k for all the systems, and these results, along with the values of the correlation coefficients r, are presented in Table III. The magnitude of n characterizes the release mechanism, and it is a function of the polymer type. In the case of solute release from swellable polymer matrices, if the fractional release data give a value of 0.5, then the transport follows the Fickian trend. If n = 1.0, the transport follows Case-II, and the intermediary values are generally suggestive of the non-Fickian (anomalous) transport.^{15–17} However, if the values of n exceed unity, then the transport follows Super Case II. Diffusion anomalies of this type have been ad-



Figure 8 Plot of $\ln (M_t/M_{\infty})$ versus $\ln t$ for St matrix with 50% NSO.

 Table III
 Statistical Analysis of Equation (1)

St:GG			
Ratio	n	$k (\min^{-n}) 10^{-3}$	r
1:0	1.19	1.79	0.992
1:0	1.42	0.52	0.988
1:0	1.12	0.52	0.988
0:1	1.40	0.67	0.910
0:1	1.59	0.17	0.992
0:1	1.54	0.19	0.978
1:3	1.23	0.96	0.993
1:3	1.22	1.10	0.997
1:3	1.18	1.04	0.987
1:1	1.00	0.62	0.994
1:1	1.24	0.96	0.989
1:1	1.17	0.84	0.975
3:1	1.40	0.67	0.961
3:1	1.35	1.24	0.994
3:1	1.19	1.31	0.995

dressed earlier in the literature.^{18–20} In the majority of present systems, the values of n range between 1.0 and 1.6, suggesting that the diffusional transport follows Super Case-II.

From a close observation of the results of kgiven in the Table III, though it is not possible to ascribe any clear physical meaning to the experimental k values, our explanations may be regarded qualitative at best. It is found that for the St matrix, the k values are higher than the GG matrix. This is indicative of the fact that St matrix interacts more efficiently with the NSO because it is more of a flexible polymer when compared to GG. The 1:1 (St:GG) matrix shows the intermediary values of k between the 1 : 3 (St:GG) and 3:1 (St:GG) matrices. However, when we compared the values of k for 1:3 (St : GG) and 3:1 (St : GG) matrices, we find that for the 3:1 matrix, the values of k are higher than 1 : 3 matrices. This is clearly indicative of the greater contribution of the more flexible St matrix towards the binary polymer mixture than the GG matrix alone. In all the cases, the k values show no dependence on the percentage loading of NSO. Additionally, the FTIR results of the individual and physical polymer mixtures, as well as the crosslinked products, are not supportive of any chemical interactions between NSO and the membrane polymer.

CONCLUSIONS

A number of different combination systems containing crosslinked starch and guar gum have

been developed for the successful encapsulation of liquid neem seed oil. From the data presented in this article, it is found that for the release of the shorter time with higher concentration of neem seed oil, starch matrix itself is more effective in soil applications, especially if the moisture content is very low. In the case of higher moisture containing soil applications, GG is a preferred matrix for the effective release of neem seed oil as it swells less when compared to starch. In the absence of any actual soil condition data, one can effectively use the 1:1 combination of the starch and guar gum systems. More research findings are presently in progress in our laboratory to evaluate these products in actual field applications for the control of oozy flies (in sericulture industry) during their earlier life cycle. Additional research is underway to investigate the suitability of the polymeric systems used here for the encapsulation of other highly toxic pesticides to reduce their toxicity levels.

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REFERENCES

- Aminabhavi, T. M.; Kulkarni, A. R.; Soppimath, K. S.; Balundgi, R. H.; Mehta, M. H.; Dave, A. M. Polym News 1998, 23, 246.
- Kulkarni, A. R.; Soppimath, K. S.; Aminabhavi, T. M. Proceed Intern Symp Control Rel Bioact Mater Boston USA 1999, 26, 437.
- Stout, E. I.; Shasha, B. S.; Doane, W. M. J Appl Polym Sci 1979, 24, 153.
- Shasha, B. S.; Trimnell, D.; Otey, F. H. J Polym Sci, Polym Chem Ed 1981, 19, 1891.
- Shasha, B. S.; Trimnell, D.; Otey, F. H. J Appl Polym Sci 1984, 29, 67.
- Shasha, B. S.; Doane, W. M.; Russell, C. R. J Polym Sci, Polym Lett Ed 1976, 14, 417.
- Doane, W. M.; Shasha, B. S.; Russell, C. R. ACS Symp Ser 1977, 53, 74.
- Kulkarni, N. V.; Rajgopalan, N.; Kale, R. P.; Khilar, K. C. J Appl Polym Sci 1992, 45, 915.
- Aminabhavi, T. M.; Balundgi, R. H.; Cassidy, P. E. Polym Plast Technol Eng 1990, 29, 248.
- Sakar, A. M.; Elsabbagh, H. M. Pharm Ind 1977, 39, 399.
- 11. Feinstein, W.; Bartilucci, A. J. J Pharm Sci 1966, 55, 332.

- Goldstein, A. M.; Alter, E. N.; Seaman, J. K. in Industrial Gums, 2nd ed.; Whilster, R. L., Be-Miller, J. N., Eds.; Academic Press: New York, 1973, Chap. 14.
- Pawar, A. D.; Singh, B. Prospects of Botanical and Biopesticides in Botanical and Biopesticides; SPS Publication No. 4; Parmamr, B. S., Devkumar, C., Eds.; Society of Pesticide Science, India, and Westvill Publishing House: New Delhi, India, 1993; p 188.
- 14. Ritger, P. L.; Peppas, N. A. J Controlled Release 1987, 5, 37.
- 15. Aminabhavi, T. M.; Harlapur, S. F.; Aralaguppi,

M. I. J Appl Polym Sci 1997, 66, 717.

- Aminabhavi, T. M.; Phayde, H. T. S.; Ortego, J. D. Polym Polym Comp 1996, 4, 103.
- 17. Aminabhavi, T. M.; Harlapur, S. F.; Balundgi, R. H.; Ortego, J. D. J Appl Polym Sci 1996, 59, 1857.
- Crank, J. The Mathematics of Diffusion, 2nd ed.; Clarendon: Oxford, UK, 1975.
- Petropoulous, J. H.; Papadokostaki, K. G.; Amarantos, S. G. J Polym Sci Part B: Polym Phys 1992, 30, 712.
- Korsmeyer, R. W.; Lustig, S. R.; Peppas, N. A. J Polym Sci Part B: Polym Phys 1986, 24, 395.