Development of an in-vitro mouth model to quantify salt release from gels

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Abstract

In the present study, an in-vitro mouth model to quantify salt release from food structures has been developed. In this instance biopolymer gels were used as model food systems. The model aimed to reproduce key phenomena occurring during oral processing, such as diffusion through the sample and compression. Salt release profiles from different gels (gelatin, gellan and alginate), under quiescent conditions and compression, were determined at temperatures of 25 and 37°C. In-vitro results indicated that salt release is affected both by the type of gelling agent and by temperature. When melting took place, release occurred within seconds. However, when diffusion through the biopolymer matrix was the controlling parameter, the time scale was in the order of hours. It has been shown that compression of the gel only affects release when fractures occur. This is believed to be a consequence of increased surface area. Finally, a mathematical model has been compiled to predict release profiles when diffusion is the controlling mechanism.

Keywords: Biopolymer, Gel, Salt Release

1. Introduction

Intake of salt is a nutritional requirement, but excessive consumption has been linked to heart disease and stroke (Cook, et al., 2007; Law, Frost, & Wald, 1991). Historically, salt has been added to foods both to enhance flavour and as a preservative (Sleator & Hill, 2007). In many cases, the removal of salt in excess of around 30% is not possible without adversely affecting the sensory characteristics of the product (Desmond, 2006). Foods such as soups, sauces, cereals and processed meats can account for around three quarters of a consumer's daily salt intake. Thus, these foods lead to many people exceeding their recommended allowance, often without their knowledge. Therefore, achieving the consumption target set by the Food Standards Agency (6g/day by 2010) is not possible without the food industry making significant efforts.

In order to reduce salt levels in processed foods, the behaviour of salt in food structures needs to be well understood (Durack, Alonso-Gomez, & Wilkinson, 2008).The development of an in-vitro mouth system for this purpose is a relatively novel idea. The models that have been developed thus far tend to involve mechanical chewing of solid foods. Work by Xu, Bronlund, & Kieser, (2005) investigated the development of a mechanical jaw using actuators. The authors attempted to mimic the chewing cycles of a person, focusing on the motion of the jaw during processing. Work by Prinz, Janssen, & de Wijk, (2007) focused more on the use of equipment to mimic conditions produced by tongue movement, and allow changes to the material to be observed as a result of this mixing. For liquid products the focus has been on flavour release. Researchers Boland, Delahunty, & van Ruth, (2006) and Juteau, Cayot, Chabanet, Doublier, & Guichard, (2004) have studied release of volatile components from gels. These studies indicated that the measurement of volatile release in a static environment does not compare to actual perception, suggesting that more dynamic in-vitro systems or in-vivo methods are needed. Similarly, Elmore & Langley, (1996) have attempted to develop a dynamic measurement using a sealed vessel, where volatiles released from material can be analysed over time. Koliandris, Lee, Ferry, Hill, & Mitchell,

(2008) investigated release rates of salt and ethyl burate (used as an artificial flavour) from gels into water. Liquid gels of bovine gelatin and locust bean gum were made at varying concentrations. The release from the material injected into a beaker of water was recorded after twenty seconds using a sodium ion probe. Researchers also investigated salt release from solid gellan and kappa-carrageenan/locust bean gum mixes. Water was added following two large strain compressions and sodium concentrations after twenty seconds of the samples was recorded. Differences in these levels were related to strain at rupture and to the extent of fracture in the compression experiments.

The perception of flavour in a product is a combination of factors, including the experience of gustatory and olfactory sensations (Verhagen & Engelen, 2006). The complexity of the system combined with the diversity of an individual's experience makes it challenging to critically assess products in-vivo. This problem can be mostly overcome using trained panels and repeated testing, but this route requires training and ethical approval, which is time and resource intensive (Carpenter, Lyon, & Hasdell, 2000). Trying to mimic the conditions experienced during the eating process would prove invaluable when attempting to reformulate products, as their behaviour could be tested in-vitro. The conditions of release of salt in food products can have a large impact on its perception by the consumer. An example of this is seen where high viscosity fluids containing the same salt as a low viscosity version are perceived as less salty. This is due to differences in mixing, a higher viscosity fluid takes longer to mix with saliva and as a result less material comes into contact with the perceiving surfaces (Ferry, et al., 2006). Many studies have been undertaken trying to link sensory perception with product properties and formulation. Malone, Appelqvist, & Norton, (2003b) studied the link between lubrication properties of hydrocolloids and emulsions and perceived smoothness and fattiness. van Vliet, (2002) also investigated a similar problem, commenting on links between texture perception and mechanical properties of products. These studies suggested that links can be made, but that the relationship between perception and material properties is complex.

In order to produce a working model, which can relate to the oral processing of products, the conditions that are experienced in the mouth need to be identified. The process can be approximated by partitioning it into three main sections (Heath, 1991; Malone, Appelqvist, & Norton, 2003a). Firstly, the oral processing stage. For liquid products this is a relatively short stage lasting approximately two seconds, and involves taking in the product and mixing it with saliva (Ertekin & Aydogdu, 2003). The product will experience low shears and a limited amount of mixing in this time. Initial processing for a solid product takes longer. The product is broken down into smaller pieces by chewing, while mixing with saliva to allow the formation of a soft bolus. The second stage is oral propulsion. This is thought to be the level which has the most influence on the breakup of complex liquid structures such as emulsions (Malone, Appelqvist, et al., 2003a). This stage involves the movement of the tongue towards the top palate, squeezing and inducing flow towards the back of the mouth, ready for swallowing. Here the fluid will experience high shears and forces, along with further mixing, before finally being swallowed (van Vliet, 2002). The final stage is swallowing, a small amount of material is retained by the oral surfaces which can then go on to play a further role in perception as salt is released by diffusion (Prinz, Huntjens, & de Wijk, 2006).

Hydrocolloids are widely used in food formulations, mainly as thickeners and gelling agents. However they are of increasing interest for use in designing complex food structures. An example exists in the use of gel particles in the place of fat droplets for low fat foods (Malone, Appelqvist, et al., 2003b). Research has been carried out in the area of flavour release from hydrocolloids, showing that gel structure can affect the release of volatiles (Boland, Buhr, Giannouli, & van Ruth, 2004; Koliandris, et al., 2008). However, molecular sizes of the flavour compounds are likely to be greater than those of sodium ions (approximately 0.1nm). Currently, the main application for controlled release from gels seems to be in drug release. Mangione, et al., (2007) uses an in-vitro approach to look at carrageenan structure effects on drug release in a franz cell. This work, and others

(Dortunç, 2001), (Murata, Miyashita, Kofuji, Miyamoto, & Kawashima, 2004), show that gels can be used to control release, and that manipulating them can alter release profiles.

Overall, the aim of this work was to develop an in vitro system for following release of salt from structures, which can be related to oral processing. Initial experiments provide data on NaCl release from three common food grade gels into water. The phenomena which govern the release were investigated and related to polymer type and concentration, salt concentration and temperature.

2. Experimental

2.1. Materials

Experiments were carried out using three commercially available food grade biopolymers: 250 bloom gelatin from porcine skin (Sigma); low acyl gellan (Kelcogel); and sodium alginate (Sigma). All were prepared as per manufacturer instructions, outlined below. The different formulations made for comparison are detailed in Table 1. All sample concentrations are percentage weight concentrations.

Table 1.

Polymer Type	Polymer Concentration (%)	NaCl Concentration (%)
Gelatin	6	3,1,0
	8	3,1,0
	10	3,1,0
Gellan	0.5	3,1,0
	1	3,1,0
	1.5	3,1,0
Alginate	3	3,0
	4	3,0

2.1.1. Gel Preparation

Gelatin samples were made by adding dry gelatin and NaCl to a beaker, then adding distilled water to make the total sample weight of 100g. The beaker was then covered, stirred and heated to approximately 60^{o}C and left to dissolve for 30 minutes. The samples were then

poured into plastic cylinders (76mm height, 22mm diameter), covered with Parafilm and chilled at 6°C for 24 hours. Gellan samples were prepared in a similar way at 90 °C, however the NaCl was added at the end of the 30 minutes stirring time, allowing the gellan powder to dissolve more efficiently. Samples were then set and stored as with gelatin. Alginate samples must be set chemically with the addition of Ca ions. Sodium alginate solutions with the desired alginate concentration were made up by heating a total sample weight of 100g to 80°C, and stirring until the alginate was fully dissolved. The liquid solution was then poured into a length of 12mm diameter dialysis tube, which was then sealed and immersed in a water bath containing 100ml of distilled water and 1% calcium chloride. Due to the dimension restrictions of the dialysis tube alginate samples had a reduced diameter and increased height compared with the other samples.

2.2. Methods

Experiments were carried out using two experimental setups. Firstly, a baffled jacketed vessel (75mm in diameter, 112mm in height) was used to study release of salt from a system under application of low shear. Secondly, a square jacketed vessel (200mm by 100mm and 50mm in height), beneath a 40mm diameter texture analyser (Stable Micro Systems, TA XT2) probe to investigate the effect of cyclic compression during release. Both systems were fitted with a conductivity probe (Mettler Toledo, inlab 710 platinum 4-cell conductivity probe) and overhead stirrer (25mm diameter propeller type). Vessel specifications are shown in Figure 1.

2.2.1. DSC Experiments

Phase transitions of the prepared gelatin were investigated using DSC (Perkin-Elmer DSC7). Gelatin samples were prepared in the same way as for release experiments and were heated at 10˚C /min from 5 to 45˚C. All experiments were performed in triplicate.

2.2.2. Salt Release Experiments

Each of the formulations was tested to follow the release of NaCl from the gel structure to a surrounding volume of water. Samples of each gel were cut into 3g (±0.1g) cylindrical segments (22mm diameter, approximately 7.5mm height) from the preparation cylinders. These were then covered and replaced in the fridge. The vessel was set up and filled with 200ml of distilled water and allowed to equilibrate at 25˚C while stirring at 100rpm to ensure a completely mixed environment. The conductivity probe was then placed into the vessel and set to record every 2 seconds. A single gel sample kept in place by a wire mesh was then added to the vessel 10 seconds after data logging was started.

Experiments were carried out for 4 hours at both 25˚C and 37˚C, and performed in triplicate. Gelatin experiments at 37˚C reached a maximum significantly before this time and so were performed for 20 minute runs.

NaCl release from the structures was followed using a conductivity probe in the main body of water. Maximum expected conductivity was calculated from calibration curves that had been previously obtained. Actual maximums tended to vary from the expected value due to drifting probe calibration and slight sample variability. Consequently, results have been normalised and presented as a fraction of total release.

2.2.3. Mathematical Modelling

Effective diffusivities of each system can be obtained using the diffusion mode in COMSOL multiphysics (COMSOL Inc. Burlington, MA, USA), with comparison to the experimental data. The system was drawn in 3D using the transient diffusion model showing the gel sample within a volume of water. The diffusion equation (1) was used. Physical properties (density and heat capacity) for both gel and water were set using COMSOL default values for water.

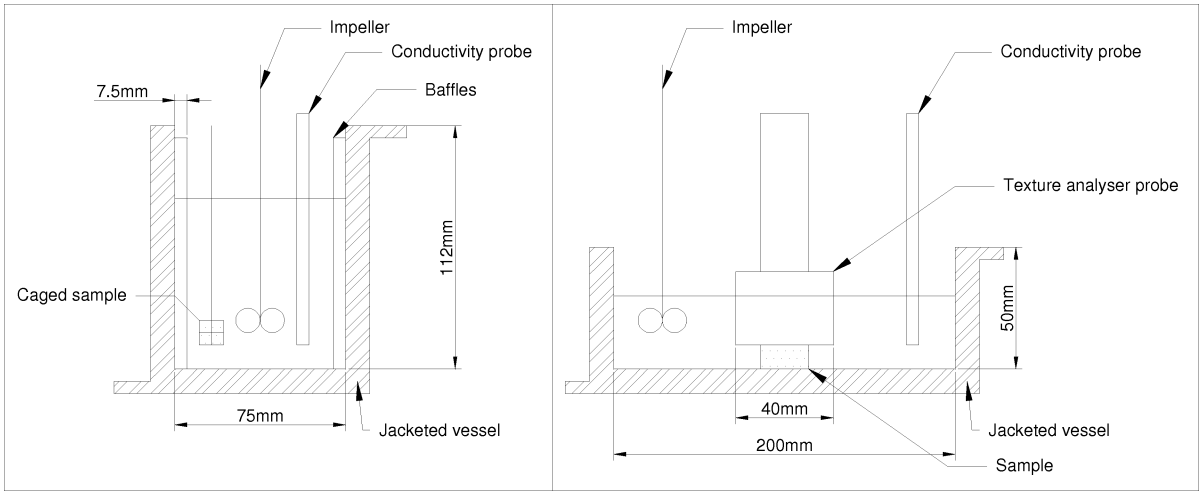
$$\partial c/\partial t + \nabla(-D\nabla c) = 0 \quad (1)$$

Where c is concentration of salt, t is time and D is diffusivity. The diffusivity of the water was set to a high value ($10m^2/s$) to represent a completely mixed vessel. External boundaries of the water were set to insulated, and temperature was fixed at the desired test level. The program was linked to MATLAB and run for a number of diffusivities within the sample and compared to average experimental data for each experiment. Values for diffusivity were estimated by minimising the sum of squares between experimental and theoretically predicted data.

2.2.4. Compression testing

For the second stage of experiments, the effect on release of repeated compression of the gel samples was studied to mimic some oral processing. The tank geometry was changed to allow a flat bottom surface to be used with a texture analyser. Samples were fixed within the vessel by lowering the compression arm to contact point. 200ml of water was then added and compressions were carried out every 3 minutes over a 20 minute period. Conductivity was recorded as per the initial experiments. Control tests with no compression were carried out as a baseline. Following this, tests using a low strain (30%) and high strain (75%) were conducted. Alginate samples were not tested in this section since the sample dimensions made compression difficult to carry out and compare.

Figure 1



3. Results and Discussion

3.1. Release experiments

DSC curves for gelatin samples were obtained. Samples of approximately 20mg were heated at 10˚/min. The peak melting point for all three concentrations of gelatin was approximately 32˚C, with an increase in energy required to melt the samples being proportional to increasing gelatin concentration.

Figure 2 shows the release of ions from gel samples as a function of time at 25˚C. The results were similar for all samples. Initially, there was a fast release which slowed as equilibrium was reached. Experiments at 25˚C resulted in relatively slow release rates, with 90% release of salt taking approximately 2 hours; this would be expected for a diffusion-controlled process (Crank, 1975). At 37˚C the results for gellan and sodium alginate are similar to the release profiles at 25˚C. However, a slightly faster release rate was observed, as expected for diffusion at the higher temperature (Figure 3). Figure 4 shows the results for gelatin. Release does not occur on the same time scale at 37˚C as for gellan and alginate. When added to the water, the gelatin sample begins to melt releasing salt at a much faster rate, with the final salt level being reached after around 10 minutes instead of the 3 hours required at 25˚C. The release follows a sigmoidal pattern, with a lag phase initially followed by an increase in concentration which slows as the final equilibrium concentration was achieved.

Results showed good repeatability proving the method useful for gathering data on salt release from a variety of structures. Between each polymer concentration some difference was observed suggesting that increasing polymer concentration slows release from the structures, due to greater physical interference from polymer chains within the structure. However, for the concentrations used here the results were not significantly different.

Figure 2

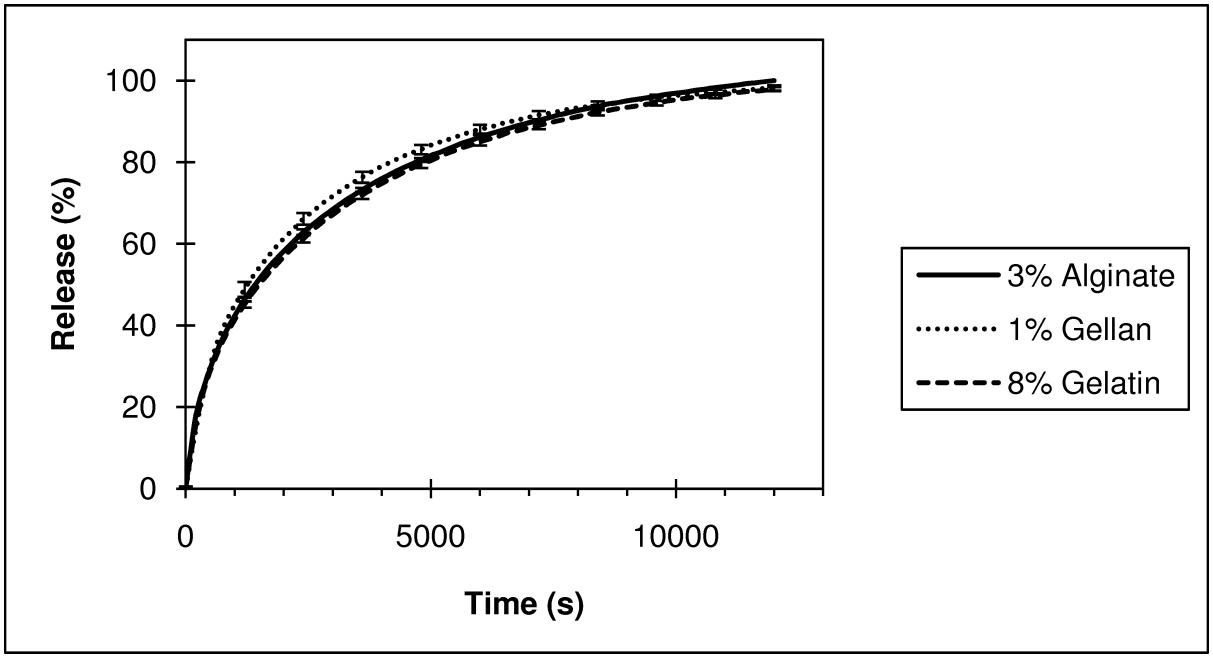


Figure 3

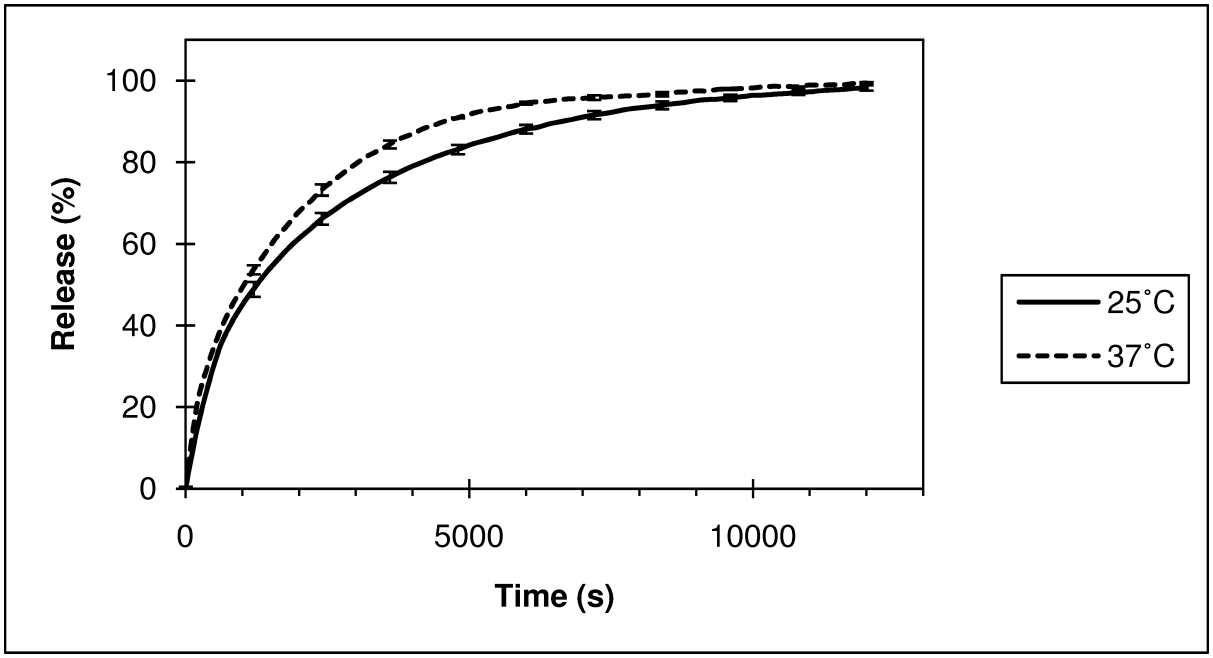
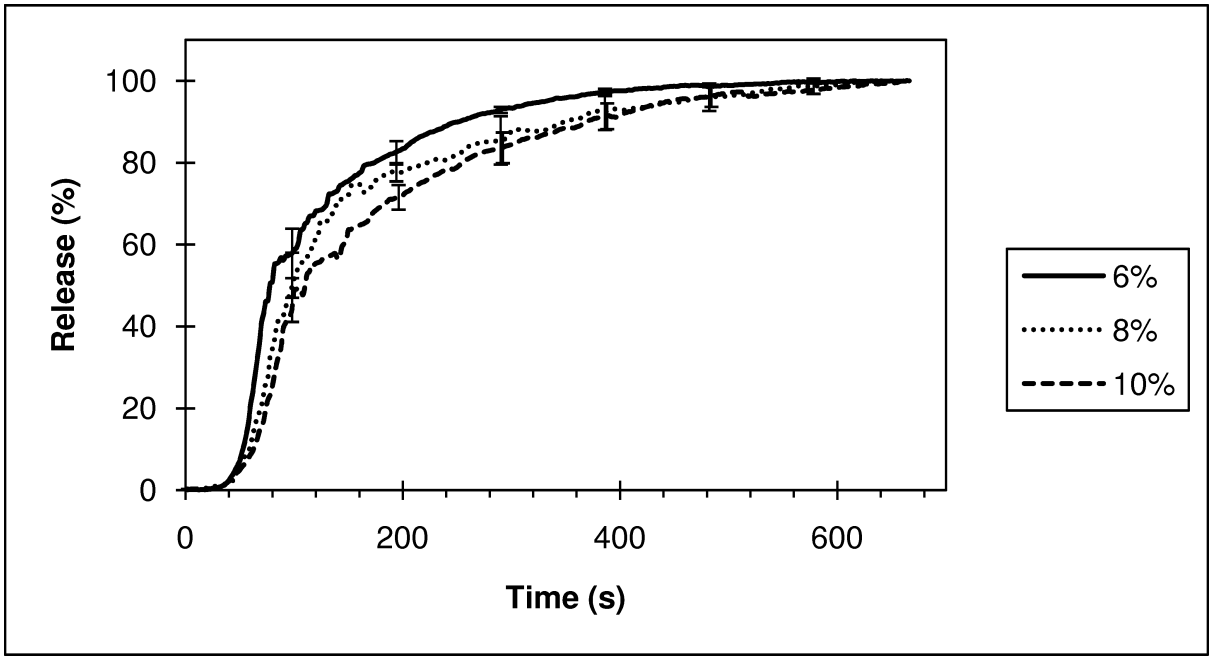


Figure 4



3.2. Mathematical Modelling

In order to calculate the effective diffusivities a model was developed using COMSOL. The system was set up to assume pure Fickian diffusion within the gel samples. This model can therefore be used for the results exhibiting diffusion behaviour. The code was run as a script coupled with MATLAB, altering the effective diffusivity to minimise the sum of squares between actual and calculated values. Figure 5 shows a sample comparison of calculated and experimental data. Initially the predicted salt released was faster than the measured salt release, but overall there was close agreement between methods. The initial difference could be attributed to a lag in recording in the physical system.

Figure 5

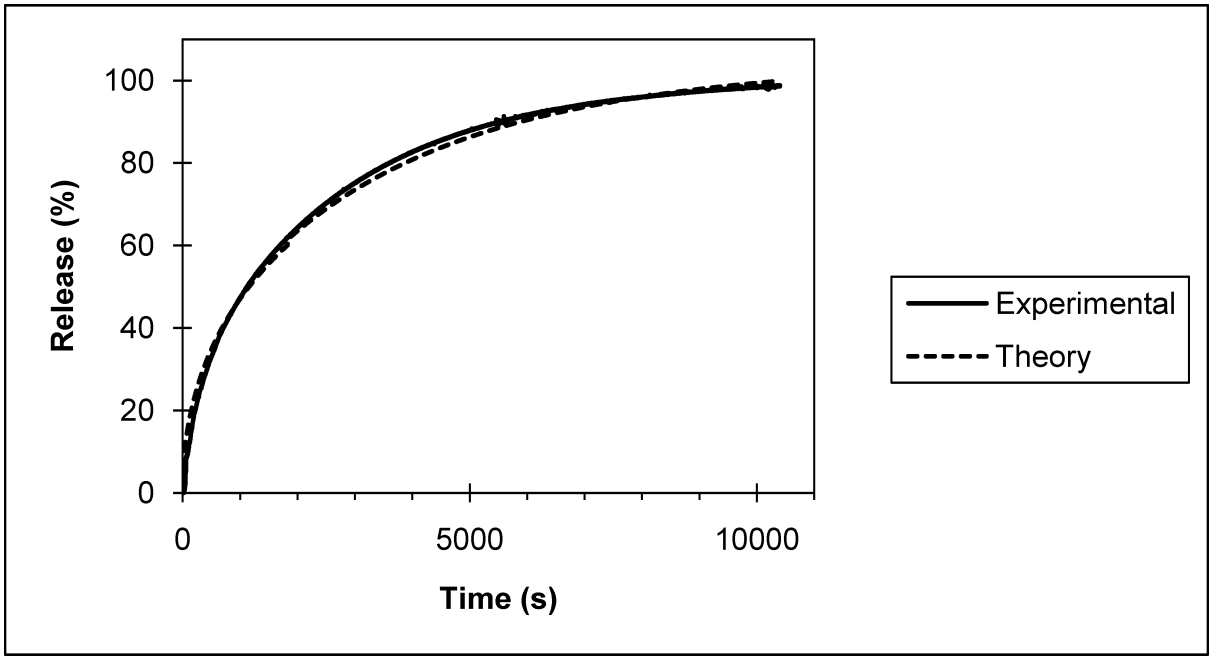


Table 2 and Table 3 show a list of estimated effective diffusivities gathered from the model at 25˚C and 37˚C respectively. Values varied slightly with concentration across samples. The results were very close to the value of free diffusion of sodium ions in water given in the literature (1.334×10^{-9}) (Lide, 2004). This indicates that the sodium ions move through the gel structure somewhat slower than in water, although not significantly so. A similar finding was reported by Floury, Rouaud, Le Poullennec, & Famelart, (2009), showing that the effective diffusivities of sodium ions through cheese into the saliva was influenced by the structure, with the observed rates varying with age and composition of the cheese. For gelatin similar experiments were previously carried out using silver ions, which have a similar size to sodium. The nature of the gelatin's open network structure means that diffusion of silver ions was unaffected, at least in the range of concentrations used (Yabuki, 1927). A similar situation must exist for gellan, whereby the structure generated during gelation was not sufficiently dense to prevent small ions diffusing through. Details from a recent review, (Miyoshi, 2009) show that most of the water within the gellan structure remains as clusters of free water which sodium ions would diffuse through at a normal rate. There are some studies in which diffusion through gel structures was shown to be slower than water (Stiles,

1920). However the methods were set up specifically to calculate the values on a small scale. Furthermore, the magnitude of differences between water and gels used was very small, the same order as the differences seen in this work.

Table 2.

Polymer Type	Polymer Concentration (%)	Effective Diffusivity ($x10^{-9}$ m^2/s)
Gelatin	6	0.99
	8	0.93
	10	0.93
Gellan	0.5	1.15
	1	1.12
	1.5	1.01
Alginate	3	0.96
	4	0.93

Table 3.

Polymer Type	Polymer Concentration (%)	Effective Diffusivity ($x10^{-9}$ m^2/s)
Gellan	0.5	1.52
	1	1.32
	1.5	1.20
Alginate	3	0.93
	4	0.99

3.3. Compression effects

Figure 6 shows three repeat stress/strain curves for a single gelatin sample. As can be seen the structure is viscoelastic under the small strains where the curves overlap for each compression. These results can be correlated with a low force environment. In order to investigate structure breakdown effects on salt release, repeat compressions at 75% strain was carried out. This could be likened to chewing of large particles or fracturing from oral propulsion of smaller particles in the mouth. At larger compressions (75% strain, Figure 7) the curves change with each compression. Initially, there is a large peak where the gel begins to break. On subsequent compressions the gel is further broken up, and smaller sections are pushed out from under the probe.

Figure 6

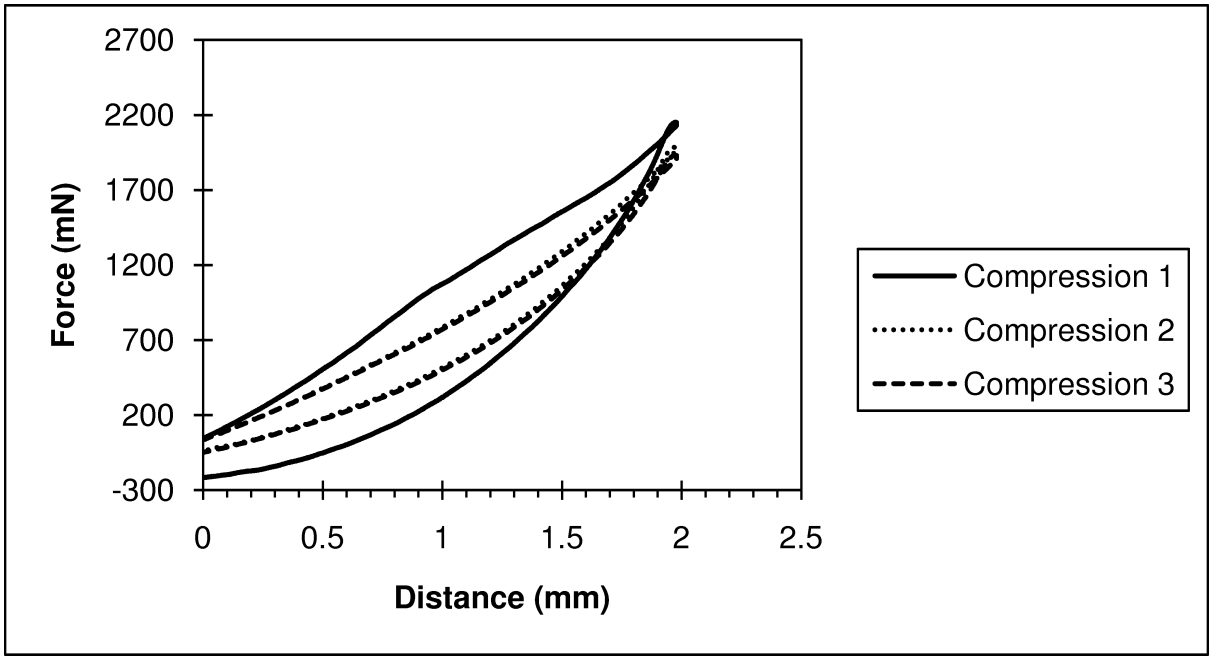


Figure 7

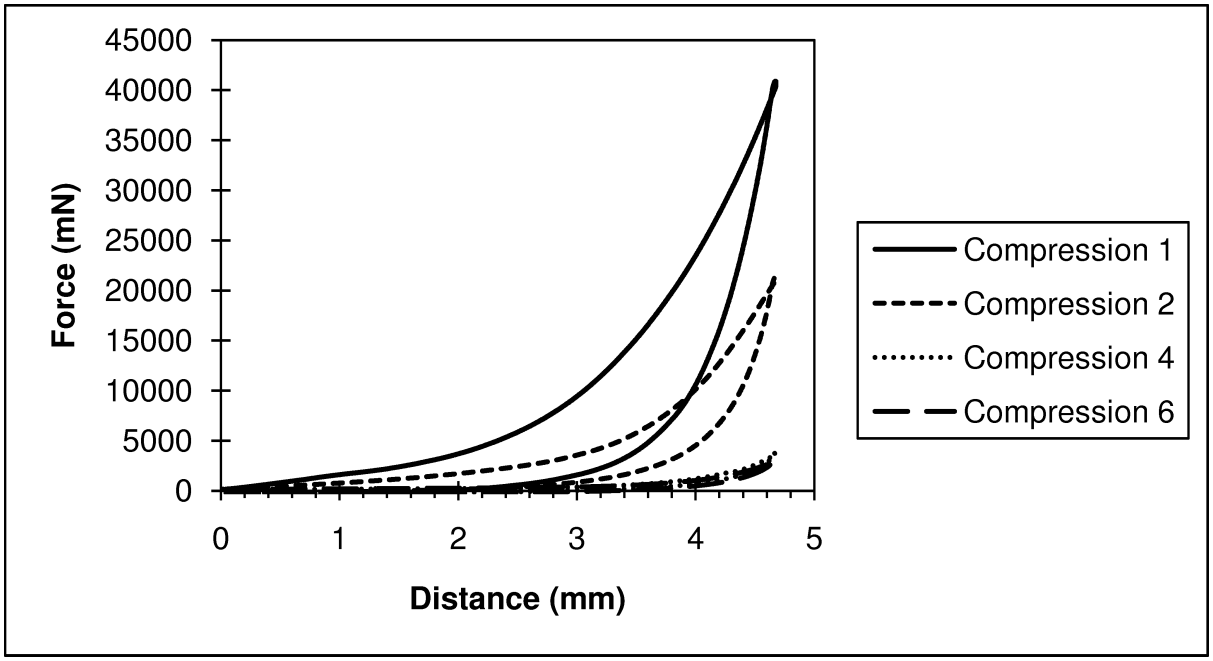
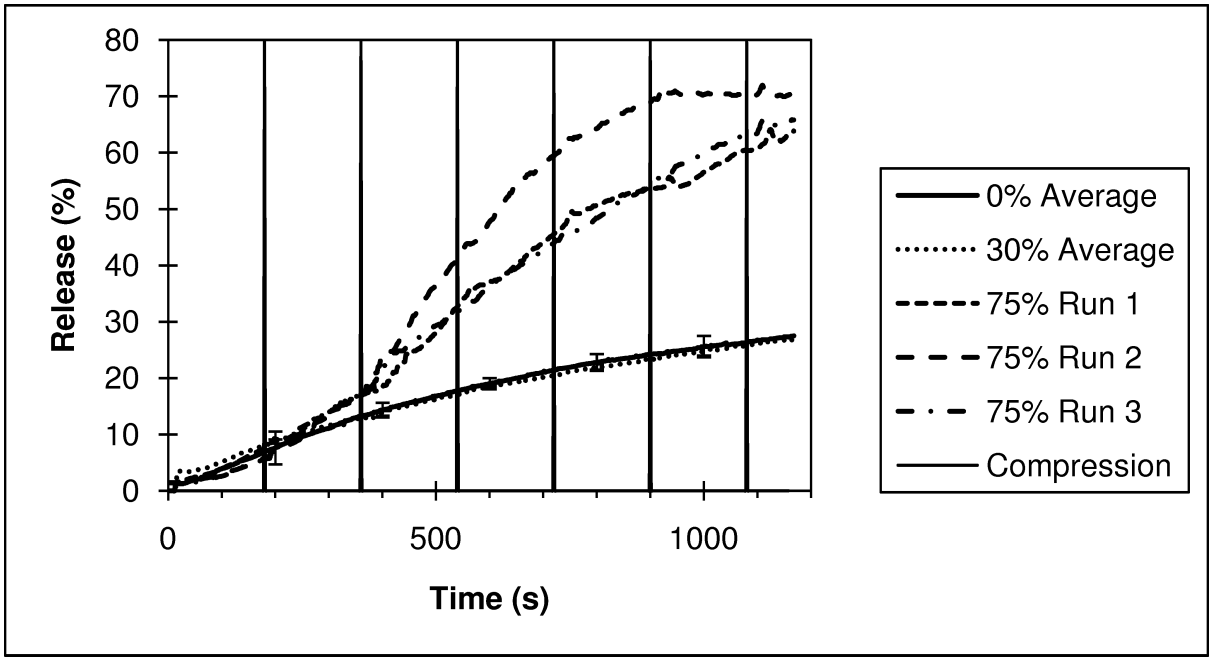


Figure 8 shows release results for gelatin. At low compressions salt release was unaffected. This is as to be expected as the gel deforms but does not break. Bot, van Amerongen, Groot, Hoekstra, & Agterof, (1996) showed that compression up to the fracture point leaves gelatin relatively unchanged, and so no increase in release rate was expected. As can be

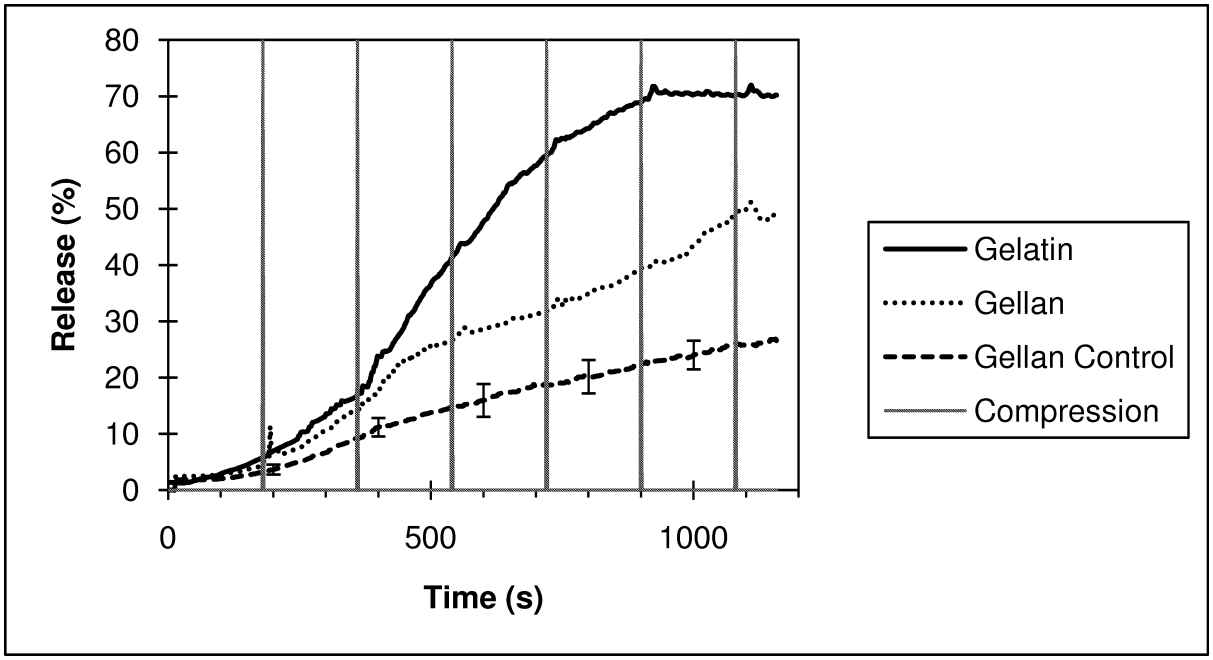
seen in Figure 8 cyclic compression of the samples does show an effect on release profiles at the higher strain of 75%. There is a distinct increase in release rate after each compression. This effect is likely to be a result of the increase in surface area generated when the gel is fractured, as discussed previously.

Figure 8



The release from different hydrocolloid gels is shown in Figure 9. This figure shows that there are initial similarities between gelatin and gellan samples in the region before compression, which corresponds to pure diffusion. However gelatin reaches a higher maximum. It appears that this is a result of the nature of the gels fracture. Gelatin tended towards multiple failures, resulting in many small segments, which mix into the bulk and have a large surface area. The gellan samples tended towards large fractures and deformation while remaining in a single block or splitting into two large sections. Similar behaviour has been reported by Harris, Smith, Campbell-Lynch, & Shelton, (2008), where gellan with different cross linkers were subjected to similar strain experiments. The gels split diagonally through the middle of the sample, but remained in large parts. These different fracture properties result in different surface areas being exposed. This leads to differences in the release profiles gained, since diffusion rates will depend upon surface area.

Figure 9



For gellan, Harris, et al., (2008) showed that in some cases water is expelled from the structure on compression, although this is not evident in our study. If water were expelled during the compressions it would be expected to release salt at a faster rate. It is possible that the high levels of salt added, needed to form cross links, affected the behaviour of the gel, giving structures and mechanical properties which are different above the critical ion concentration of the gellan (Tang, Tung, & Zeng, 1996). This would explain our observations.

The model that was developed for diffusion can be used to investigate how salt release time is dependent on gel sample size and whether this fits to the observations made during compression experiments. Firstly, by comparing the release times under compression data, an estimate of particle sizes present after compression can be calculated. Secondly, salt release times for an emulsion type product with salt containing gel droplets can be calculated. Currently, some low fat spread products use gel encapsulated oil droplets to control lipophilic volatile release and so a similar approach for salt may be relevant (Malone & Appelqvist, 2003). In order to model these systems, the cylinder geometry used in the previous models was altered to an equivalent volume sphere geometry. This made

automation of the model more practical and is a better representation of an emulsion type product with gelled droplets. However it does change the surface area of samples with the same volume.

The time taken to release 70% of the contained salt for a range of sample sizes relevant to those used in this study is shown in Figure 10. The maximum point shows time for a sphere, which is volume equivalent to cylinders used in previous experiments. The time predicted for the sphere to release 70% of the contained salt is around 30% longer than actual data for the cylinder, this is due to the reduction in surface area (by about 25%) caused by the geometry change. The time taken to release 70% of the contained salt for experiments at 75% compression in gelatin would be expected for spheres with a radius of around 3.6 mm. The time to release 50% salt in gellan again is predicted to be around 30% slower than in the actual cylindrical data, here the results taken from the 75% compression experiments would be expected for spheres with a radius of around 6 mm (Figure 10). This indicates that the increase in salt release rate seen is due to the increase in surface area caused by breakup under compression, and that gelatin breaks into smaller segments than gellan. In both cases the particle sizes would be smaller for cylinders and in reality irregular shapes are formed. Studies into particle sizes created during mastication have shown that a range of particles sizes are created. Jalabert-Malbos, Mishellany-Dutour, Woda, & Peyron, (2007) show that for a range of foods, sizes from 0.4-4mm with a median around 2mm were found when ready for swallowing. Mielle, et al., (2010) show particle sizes after 4 and 8 chews of peanuts which vary over a similar range. This places the sample sizes created from compression experiments at the very beginning of mastication. Figure 11 shows expected release times for small gelled emulsion droplets (20-60 microns). Release at this scale is very fast (less than 1 second), indicating that this alone would not form an effective method for controlling salt release.

For all instances, increasing radius does not give a linear increase in time. An analytical solution to this situation can be found in Crank, (1975) as a sphere in a stirred media. This

has been applied in the release of volatiles in emulsion droplets which are of comparable scale and time to release 50% of the contained volatiles is described by:

$t_{1/2}=(0.162r^2)/D$

Where $t_{1/2}$ is time to release 50% material, r is droplet radius and D is diffusivity. This means that time to release a set percentage of salt will increase proportionally with radius squared.

Figure 10

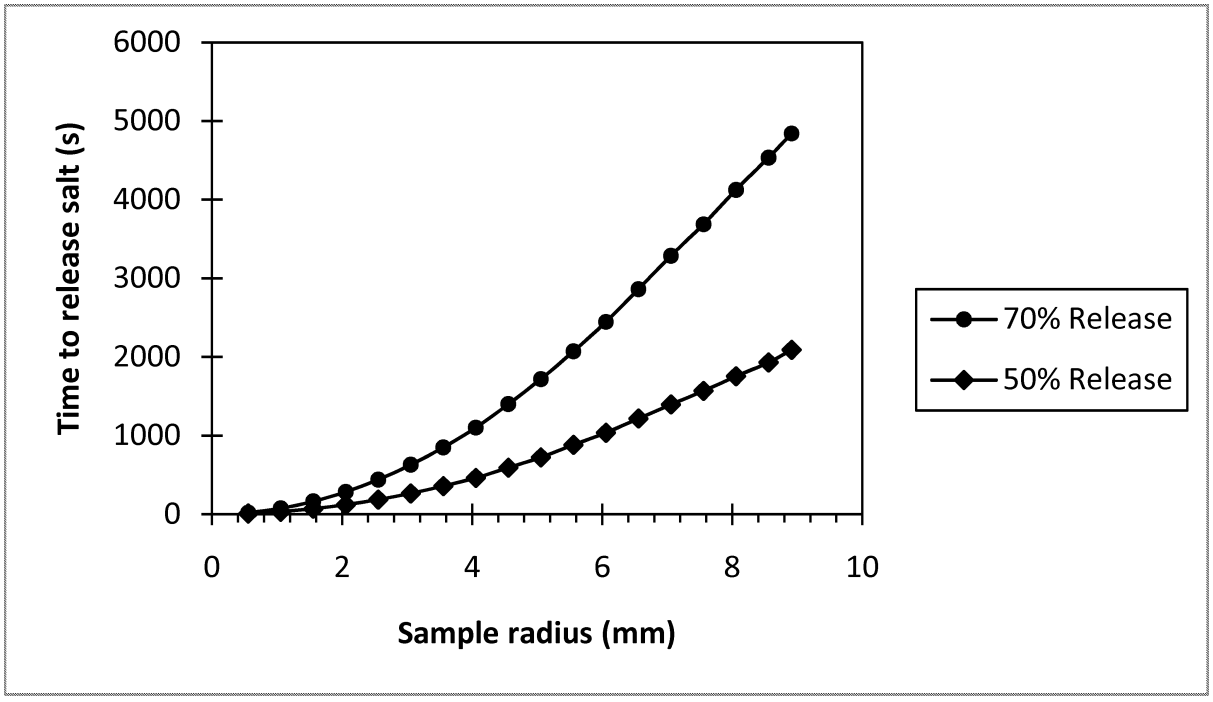
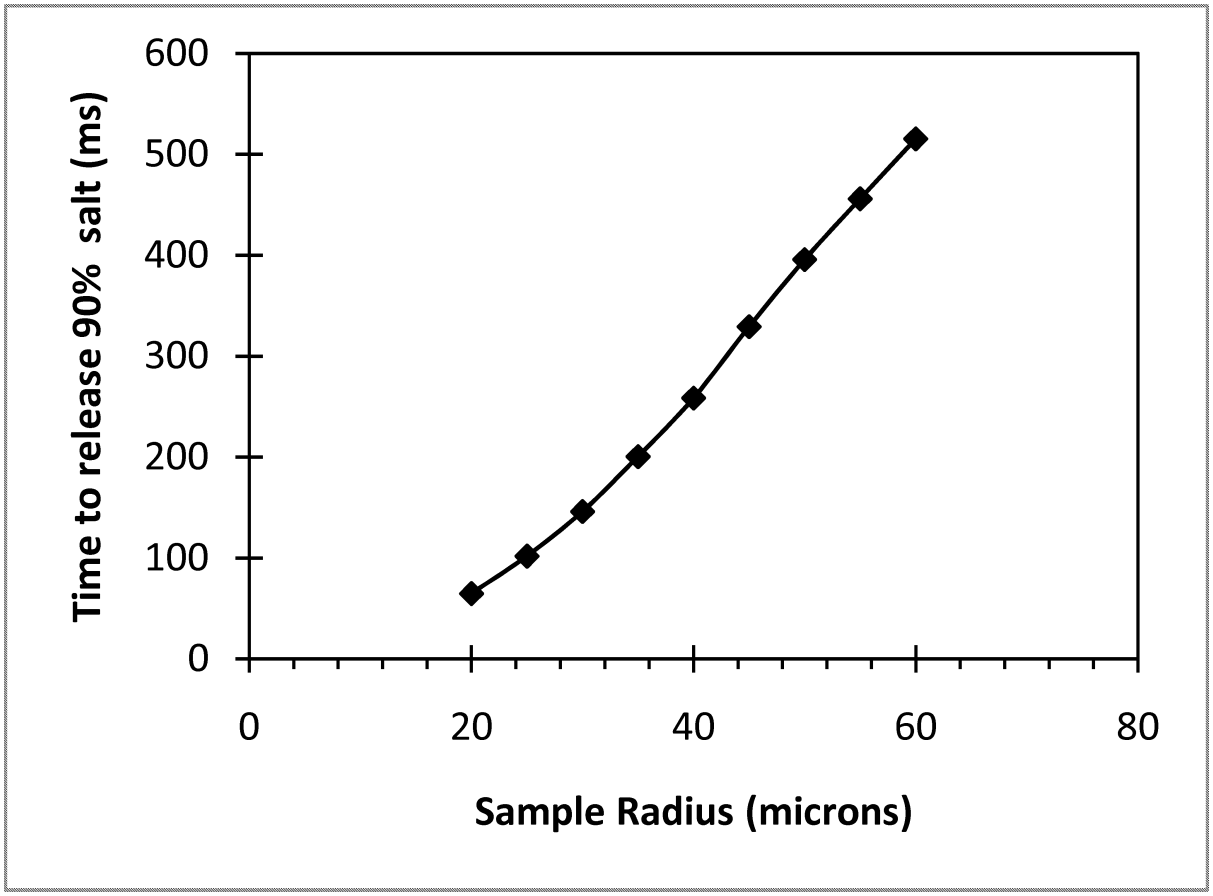


Figure 11



4. Conclusions

It can be concluded that the in-vitro method of testing salt release from hydrocolloids is reliable and repeatable. In the future this will be extended to include different structures which can be compared with the gel experiments carried out in this study. Release curves gathered from the different hydrocolloids (gelatin, gellan and alginate) show there is very little to impede the diffusion of sodium ions out of the structure into the surrounding media. Compression effects on the release rate are evident and are predominately a result of the gel's structure. In the single phase structures tested there is no evidence of increased release rate with compressions that do not cause fracture of the structure. When compressions are significant enough to cause fractures within the samples, the release increases depending on the extent of failure and increased surface area. Gelatin, tending towards many discrete pieces released a greater amount of its salt content than gellan,

which compressed forming fractures, but staying in a single piece. It is agreed that for semisolids at least, deformations during oral processing are important (Finney & Meullenet, 2005), and that the deformations themselves can affect the outcome. We have shown that there is not much control for salt release using these gels, however more complex structures may allow other release profiles to be designed.

Modelling the process has allowed effective diffusivities of each system to be obtained, and while they do not vary significantly to that of water, the values can be used to predict release rate from a variety of different geometries and conditions which match analytical solutions for similar systems in Crank (1975).

5. Acknowledgements

The authors would like to acknowledge EPSRC for funding this work

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Table 1: Materials and sample formulations used in all the experiments

Table 2: Effective diffusivities of sodium ions for tested samples at 25˚C

Values for effective diffusivity at 25˚C used within the diffusion model in COMSOL which best-fitted experimental data were taken.

Table 3: Effective diffusivities of sodium ions for tested samples at 37˚C

Values for effective diffusivity at 37˚C used within the diffusion model in COMSOL which best fitted experimental data were taken.

Figure 1 Vessel diagrams for two experimental setups used in this study

Figure 2 Salt release over time into 200ml of water from 3g cylinders of 3% alginate, 1% gellan and 8% gelatin at 25˚C.

Figure 3 Comparison of salt release into 200ml of water over time from 3g cylinders of 1% gellan at 25 and 37˚C

Figure 4 Salt release into 200ml of water over time of 3g cylinders of 6,8, and 10% gelatin at 37˚C

Figure 5 Comparison of model output and experimental results of salt release over time from 3g cylinders of 0.5% gellan at 25˚C

Figure 6 Stress strain curves for 3 sequential compressions of a 3g cylinder of 8% gelatin. Tests were performed at a constant rate of 1mm/s to a strain of 30%

Figure 7 Stress strain curves for 6 sequential compressions of a 3g cylinder of 8% gelatin. Tests were performed at a constant rate of 1mm/s to a strain of 75%.

Figure 8 Salt release over time (into 200ml of water) from 3g cylinders of 8% gelatin at 25˚C at 0%, 30% and 75% strain. Samples compressed to 75% were not reproducible and are presented as individual experiments.

Figure 9 Salt release over time into 200ml of water from 3g cylinders of 1% gellan and 8% gelatin at 25˚C at 0% and 75% strain. A single experiment is shown for gelatin and gellan at 75% strain for comparison since results were not repeatable.

Figure 10 Time to release 50% and 70% salt from gellan spheres of varying radius, data is generated using COMSOL

Figure 11 Time to release 90% salt from emulsion droplet sized gellan spheres of varying radius, data is generated using COMSOL.