MODIFICATION OF MESOPOROUS SILICA NANOPARTICLES FOR IBUPROFEN LOADING AND RELEASE IN DRUG DELIVERY

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Dear Allah,

I am sincerely grateful for everything.

To my parents

Kamarudin Amin & Meme Haryati Abd Hamid

All that I was, I am and all that I wish to be, I owe to both of you.

To my brothers and sisters

Farid, Amira & Firdaus

Thank you for being everything and never failed to be there for me.

To my husband

Khairul Nur Azfar Baharudin & family

Thank you for seeing me through the eyes of love, and overlooking my many flaws.

To the little darling

Nursyifa' Imanina Khairul Nur Azfar

Your smile and laughs are my strength to look forward for tomorrow.

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ABSTRACT

Mesoporous silica nanoparticles (MSN) were synthesized by conventional method and microwave heating as drug delivery platform for the adsorption and release of ibuprofen, an anti-inflammatory drug. MSN was modified by 3-aminopropyltriethoxysilane (APTES) and aluminum (Al) metal. Modification with APTES was conducted via co-condensation (MSN-APT_{co}) and post-grafting method (MSN-APT_{post}) of MSN. The percentages of adsorption of ibuprofen were 100%, 71% and 78%, while the releases were 50%, 100% and 38% for MSN, MSN-APT_{co} and MSN-APT_{post}, respectively, which resulted from the difference in the surface functional group. 1%, 5% and 10% of aluminum (Al) were loaded onto MSN via the impregnation method. The adsorptions of ibuprofen were 35%, 58% and 79%, while the releases were 100%, 86% and 89% for 1%, 5% and 10% Al loaded MSN, respectively. The increase in Bronsted acidity upon loading of Al up to 10% strongly bound the drug, which caused the highest adsorption but the slowest release of ibuprofen. MSN was also synthesized with microwave power of 100W (MSN-MW_{100}), 300W (MSN-MW_{300}) and 450W (MSN-MW_{450}). MSN-MW_{450} exhibited the highest ibuprofen adsorption (100%), followed by MSN-MW_{300} (75%) and MSN-MW_{100} (58%), while the percentages of release were 65%, 81% and 95%, respectively, depicting longer channel of MSN demonstrated higher adsorptivity toward ibuprofen, while simultaneously delayed the release process. From all the studies, the vital factors for ibuprofen delivery were found to be the surface functional group, acidity and also the mesoporous channel length. With these factors, MSN can be designed to fulfill the desired drug delivery system. In conclusion, MSN can be tailored to have suitable features for slow drug release which provide constant release over a defined period to avoid repetitive administration. In parallel, MSN also could be employed as a fast drug release system that provides initial burst of drug release to achieve rapid and maximum relief.

ABSTRAK

Zarah nano silika berliang meso (MSN) telah disintesis dengan kaedah biasa dan gelombang mikro sebagai penyokong untuk penjerapan dan pembebasan ibuprofen, suatu ubat anti-radang. MSN telah diubahsuai dengan 3-aminopropiltrietoksisilana (APTES) dan logam aluminium (Al). Ubahsuai dengan APTES telah dijalankan melalui ko-kondensasi (MSN-APT_{co}) dan kaedah pasca-gabungan (MSN-APT_{post}). Penjerapan ibuprofen adalah 100%, 71% dan 78%, manakala pembebasan adalah 50%, 100% dan 38% masing-masing untuk MSN, MSN-APT_{co} dan MSN-APT_{post}, masing-masing, yang disebabkan oleh perbezaan pada kumpulan berfungsi permukaan. MSN telah ditambah dengan 1%, 5% dan 10% aluminum (Al) telah melalui kaedah pengisitepuan. Peratus penjerapan ibuprofen adalah 35%, 58% dan 79%, manakala pembebasan adalah 100%, 86%, 89% untuk MSN yang masing-masing ditambah 1%, 5% dan 10% Al. Peningkatan pada keasidan *Bronsted* dengan penambahan Al sehingga 10% mengikat ubat dengan lebih kuat, yang menyebabkan penjerapan tinggi tetapi pembebasan yang lambat. MSN telah disintesis menggunakan gelombang mikro berkuasa 100W (MSN-MW_{100}), 300W (MSN-$MW_{300)}$ dan 450 W (MSN-MW_{450}). MSN-MW_{450} mempamerkan penjerapan ibuprofen tertinggi (100%), diikuti dengan MSN-MW_{300} (75%) dan MSN-MW_{100} (58%), manakala peratus pembebasan adalah masing-masing 65%, 81% and 95%, menandakan saluran yang lebih panjang menunjukkan penjerapan yang lebih tinggi terhadap ibuprofen, dalam masa yang sama melambatkan proses pembebasan. Daripada semua kajian, faktor penting untuk penyampaian ibuprofen yang ditemui adalah kumpulan berfungsi permukaan, keasidan dan juga panjang saluran liang meso. Dengan faktor-faktor ini, MSN boleh direkacipta untuk memenuhi sistem penyampaian ubat yang dikehendaki. Kesimpulannya, MSN boleh direka untuk mempunyai ciri-ciri yang sesuai untuk penyampaian ubat secara perlahan di mana menyediakan pembebasan yang berterusan dalam masa yang telah ditetapkan untuk mengelakkan pengambilan berulang. Sejajar dengan itu, MSN juga boleh dijadikan sistem penyampaian ubat yang pantas yang menyediakan permulaan pembebasan ubat yang cepat untuk mencapai kelegaan yang pantas dan maksimum.

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LIST OF ABBREVIATIONS

^{27}Al NMR	-	Aluminum Nuclear Magnetic Resonance
^{29}Si NMR	-	Silicon Nuclear Magnetic Resonance
APTES	-	3-Aminopropyl Triethoxysilane
BET	-	Brunauer-Emmet-Teller
BJH	-	Barrett-Joyner-Halenda
COX-1	-	Cyclooxigenase
CTAB	-	Cetyl trimethylammonium Bromide
EG	-	Ethylene glycol
FBS	-	Fetal bovine serum
FESEM	-	Field-Emission Scanning Electron Microscopy
FT-IR	-	Fourier Transform Infra-Red
HMS	-	Hollow mesoporous spheres
HSM	-	Hollow Slica Microspheres
MCM-41	-	Mobil composirion Matter 41
MSN	-	Mesoporous Silica Nanoparticle
MSN-Al	-	MSN loaded Al metals
MSN-Al	-	MSN modified by Al
MSN-APT	-	MSN modified by APTES
MSN-MW	-	MSN synthesized by microwave
MSU	-	Michigan State University
MTS	-	Methyl triethoxysilane
MW	-	Microwave
NMR	-	Nuclear magnetic resonance

NSAID	-	Non-steroidal anti inflammatory drig
PEM	-	Polyelectrolyte multilayer
PSS	-	sodium polystyrene sulfonate
Q	-	Degree of condensation
SBA-5	-	Santa Barbara Amorphous 15
TEM	-	Transmission Electron Microscopy
TEOS	-	Tetraethyl orthosilicate
TG	-	Thermogravimetry
UV	-	Ultraviolet
WRL	-	Human Hepatic Cell
XRD	-	X-Ray Diffraction
XRF	-	X-ray Fluorescence

LIST OF SYMBOLS

Å	-	Angstrom
cm	-	Centimeter
g	-	Gram
K	-	Kelvin
kJ	-	Kilojoule
m	-	Meter
µmol	-	Micromole
ml	-	Milliliter
min	-	Minutes
%	-	Percentage
θ	-	Theta
wt %	-	Weight Percentage
W	-	Pore diameter
Vp	-	Pore volume
t	-	Pore wall thickness
S	-	Specific surface area
SA	-	Si/Al molar ratio
a_0	-	Lattice
d_{100}	-	d-value space
h	-	Hour
nm	-	Nanometer

CHAPTER 1

INTRODUCTION

1.1 Research Background

Fortified by the exciting discovery of new kinds of molecular sieves called MS-41 in the early 1990s, exploration on the synthesis of mesoporous silica materials has received growing attention and advanced rapidly (Kresge *et al.,* 1992; Inagaki *et al.,* 1993). Great endeavors have been conducted in the tailoring of particle size, pore diameter, morphology, structure, surface properties and functionalization of mesoporous silica to improve their applications in the fields of catalysis, separation, adsorption, and drug delivery, etc (Ying *et al.,* 1999; Sayari and Hamoudi, 2001; Raja and Thomas, 2002; Liu *et al.,* 2005). As one of the most promising application for human health care, controlled drug-delivery systems represent an ever-evolving field for biomedical materials science.

From a technical perspective, controlled drug delivery implies the ability to control the distribution of therapeutic agents both in space and time. In other words, controlled drug delivery embodies both control of the rate of release of a drug, and the delivery of this drug to a specific organ or location in the body (Barbe *et al.,* 2004).

In recent years, mesoporous silica nanoparticles (MSN) have been well developed as effective drug storage vehicles in drug delivery systems (Manzano *et al.,* 2008, Mortera *et al.,* 2010) owing to their large pore volume, high surface area (Vallet-Regi *et al.,* 2001), ease of functionalization (Lei *et al.,* 2010), low toxicity and biodegradability. However, one of the main and specific problems of drug delivery system by mesoporous materials at current is the pore sizes that could not encounter all types of desired drugs which consist of bulky and different features. For this application, the morphology control of MSNs, especially their particle size, dispersivity and pore size are important issues because particles or aggregates with sizes above 300 nm may lead to thrombosis (Barbe *et al.,* 2004) and the pore diameter determines the dimensions of drug molecules which can be loaded in them. In this sense, synthesis of controllable mesoporous material by an efficient method is crucial and imperative tasks.

Moreover, one of the main targets of current delivery systems in the pharmaceutical industry is to provide a sustained released over time of the active agent in order to maintain its concentration within therapeutic values and below the diligence toxicity threshold (Shi *et al.,* 2011). It is supposed that this delivery rate could be modulated by modifying the interaction between the confined molecule and the mesoporous silica medium. This objective could be achieved by functionalization of the pore wall, with such as 3-aminopropyltriethoxysilane (APTES). Modification of mesoporous silica by APTES has been conducted by Wang *et al,* (2009a) and they reported that the release of drug molecules was found to be dependent on the type of functional groups in the materials (Wang *et al.,* 2009a). Generally, surface functionalization of mesoporous silica materials via covalent bonding of organic groups can be achieved by two methods: post grafting synthesis and co-condensation (Sharma and Asefa, 2007). The resulting functionalized mesoporous materials may help to deliver drugs efficiently and thus, minimize the drugs possible adverse effects. The main advantage of introducing any functionality within the pore walls of MSN is that the non-siliceous group will not partially block the mesopores. This allows better diffusion of the molecules of interest through the pores when using the material (Slowing *et al.,* 2010). The presence of pores of uniform size lined with

silanol groups considers these materials potential interest as host of a variety of guest chemical species, such as amino groups (Moller and Bein, 1998).

In this study, ibuprofen was chosen as a model molecule, as it is currently used in a range of pharmaceutical formulations an analgesic and anti-inflammatory drug. Ibuprofen that was designated as a core medicine in the "WHO Model List of Essential Medicines" is generally derived from propanoic acid (Dutta *et al.*, 2012). Ibuprofen is known to have an antiplatelet effect, though it is relatively mild and short-lived compared to aspirin or other better-known antiplatelet drugs (Esch *et al.*, 1995). Ibuprofen can be impregnated into mesoporous silica materials by reacting with the active groups on the mesoporous framework, for instance, by hydrogen bond with surface silanol groups (Szegedi *et al.*, 2011). Cross-reference to recent studies on ibuprofen delivery by carriers based on both mesoporous silica and metal-organic framework systems, should facilitate extension of this current knowledge in this fields by giving broarder view. Therefore, herein we attempt to synthesis and characterize mesoporous materials with different properties, as well as studying its activity towards ibuprofen immobilization and release profile.

Additionally, mesoporous silicas incorporated inorganic groups such as transition metals and metal oxides are also known as potential materials for the adsorption of drugs. For instance, Cu^{2+} loaded onto SBA-15 was reported to be an effective adsorbent for naproxen via the metal-drug complexion (Rivera-Jimenez *et al.*, 2010), while MnO-loaded SBA-15 performed well as a vehicle for a doxorubicin anti-cancer drug due to the accessibility of its paramagnetic center for encapsulation/sustained release/intracellular delivery of drugs (Chen *et al.*, 2012b). On the other hand, zeolite was also reported as a good candidate drug carrier because the Al allows potential interactions with the drug. In a series of SiO_2/Al_2O_3 ratio studies, extra-framework Al in zeolite Y was found to form a complex with the drug 5-fluorouracil (Datt *et al.*, 2013). The Al content was also reported to generate acid sites that play an important role in ibuprofen adsorption (Das *et al.*, 2009). However, the adsorptivity of such zeolites toward a wide range of drugs is still low due to their small pore sizes. In this sense, the use of larger pore size mesoporous silica with

incorporated Al may offer greater advantages for drug adsorption. Besides, detailed reports on the understanding of acidity in terms of Lewis or Brönsted acid sites with relative to the drug delivery are still rare. Acordingly, in this study we also attempt to introduce Al onto the MSN, to observe its potential towards the adsorption and release of ibuprofen.

The traditional synthesis method of mesoporous materials is the hydrothermal route, which uses a certain amount of surfactants, as well as acid or alkali to compose a mixed aqueous preparation. Next, inorganic sources are added and heated to crystallize, followed by filtration, drying, and calcination or extraction to remove the template. Although finely ordered mesoporous materials are obtained, the process is time and energy consuming (Jiang *et al.,* 2008; Yu *et al.,* 2012). Heating solids in the conventional system leads to an uneven temperature distribution due to poor heat transfer into the bulk of the material. The outer temperature may be substantially higher than the inner one, because the material itself acts as an insulator. In these modern days where scientific findings and technology go hand in hand, any improvement to a synthesis technique that saves time in the synthesis of new materials or improves the properties of materials would be extremely beneficial (Saxena and Chandra, 2011). It is known that hydrothermal synthesis of inorganic materials using microwave heating promotes nucleation and can reduce the synthesis time and particle size significantly in comparison with the conventional convection heating method (Newalkar *et al.,* 2001; Hwang *et al.,* 2004; Yoon *et al*., 2008). Besides, with faster polymerization under microwave irradiation, it was also found that the swelling rate of the material was much higher compared to a material prepared by conventional heating. In fact, scanning electron microscopy revealed that the material produced under microwave irradiation consisted of evenly distributed pores (Xu *et al.,* 2005).

Therefore, within this context, microwave irradiation under different heating power was applied to the synthesis of MSN, with the expectation of a reduction in synthesis time and formation of MSN with enhanced drug adsorption properties. The relationship between material crystal growth and crystallinity, surface area, pore size,

particle size, and morphology are also discussed. We suggest an approach to the formation of MSN from a mixture of cetyl trimetylammonium bromide (CTAB), water, ethylene glycol, ammonia, and tetraethyl orthosilicate (TEOS). Ammonia was chosen as the catalyst and ethylene glycol as the co-solvent because of their polarity, which is higher than that of NaOH and methanol or ethanol which are commonly used to synthesize MSN. The understanding of those parameters provided control of the structural and morphological characteristics of these materials was beneficial for the design of a drug delivery system.

1.2 Problem Statement

Recent studies show that the mesoporous silica nanoparticles appears as one of the best candidate for drug delivery system due to its tuneable pore size, large pore volumes, high specific surface area, good thermal stability, biocompatible and non-toxic nature (Tourne-Peteilh *et al.,* 2003). However, the loading of drug onto the support often faced several problems due to lack of activity due to small pore size that could not encounter all the desired drugs, as well as the deficiency of active sites. In order to overcome these problems, the modification towards the MSN to improve its physicochemical properties and efficiency of drug loading and release are highly required. Other main and more specific problems of drug delivery systems at present is the loss of activity of several drugs before reaching the target tissue as a result of premature degradation of the active agent. The other concern also focused towards the efficiency of the designed system, which is important. Despite its efficiency, conventional heating during MSN synthesis may also be time and energy consuming.

Recently, Szegedi et al (2012) reported that modification by organic groups such as amine had a positive effect on the adsorption capacity of ibuprofen. However, application of much higher amount of organic group than the

stoichiometrically needed results in the development of disadvantageous properties, such as functionalization of outer surface of the silica particles and unfavourable agglomeration. Thus, the study of modified MSN for drug delivery is still a challenge and imperative task.

1.3 Hypothesis

Due to the highly ordered structures, high surface area, large pore sizes, and the silica surface that could be modified and functionalized, the MSN is expected to provide an excellent utilities for drug adsorption and release. In this sense, the synthesis of controllable and tailorable mesoporous material by an efficient method is a crucial and imperative task. Due to the differences of drugs nature, not all kind of drugs suits the surface chemical of MSN, which then the functionalization and modification takes role. Functionalization is conducted accordingly based on the desired drug's characteristics to enhance and assist in the adsorptivity. In fact, mesoporous silica shows high density of silanol groups, which can be used to obtain functionalized surfaces by grafting organic or inorganic groups. Organic functionalization agent, such as 3-aminopropyltriethoxysilane (APTES) could provide binding sites to the desired drugs by the $–NH_2$ groups on the surface. Apart from the organic groups, functionalization of MSN by inorganic groups, such as metals or metal oxides also offers great advantages to the MSN, such as introducing acid sites to interact with the interest drug. Moreover, due to the time consuming of the conventional heating during MSN synthesis, microwave-assisted synthesis offers higher advantages on reducing the synthesis time as is expected to preserve the MSN properties and good activity towards the ibuprofen adsorption and release.

1.4 Objective of the Study

The objectives of this study are as follows:

1. To study the performance of ibuprofen delivery on mesoporous silica nanoparticles (MSN)
2. To study the effect of 3-aminopropyltriethoxysilane modified MSN on the performance of ibuprofen delivery.
3. To study the effect of Al metal modified MSN on the performance of ibuprofen delivery.
4. To study the effect of microwave-synthesized MSN on the performance of ibuprofen delivery.

1.5 Scope of the Study

The scope of this study consists of four parts, which are:

Study the performance of ibuprofen delivery on mesoporous silica nanoparticles (MSN)

1. The MSN was prepared as the standard material, using tetraethyl orthosilicate (TEOS) as the silica source, ethylene glycol as the co-solvent, ammonium hydroxide as the catalyst and the temperature of reaction was kept at 80°C. Adsorption of ibuprofen was carried out under room temperature, while the release process was conducted in the simulated body fluid (SBF) at 37°C. The SBF is a suitable medium for this study as it resembles the environment in the human body.

2. Study the effect of 3-aminopropyltriethoxysilane modified MSN on the performance of ibuprofen delivery.

   3-aminopropyltriethoxysilane was introduced onto MSN surface by co-condensation and post-synthesis method. Adsorption of ibuprofen was carried out under room temperature, while the release process was conducted in the SBF at 37°C.

3. Study the effect of Al metal modified MSN on the performance of ibuprofen delivery.
   1%, 5% and 10% of Al metal was loaded onto the MSN. Adsorption of ibuprofen was carried out under room temperature, while the release process was conducted in the SBF at 37°C.

4. Study the effect of microwave-synthesized MSN on the performance of ibuprofen delivery.
   MSN was synthesized by using microwave power of 100W, 300W and 450W. Adsorption of ibuprofen was carried out under room temperature, while the release process was conducted in the SBF at 37°C.

1.6 Significant of the Study

This research was conducted to synthesize and modify the MSN. The physicochemical characterization with relation to the adsorption and release of ibuprofen was also studied. The keystone in the development of MSN in drug delivery systems along this study is the alteration of surface through organic compound and inorganic compound, as well as different approach to the synthesis method, because this process provides numerous possibilities of enhancements to the MSN properties to control drug adsorption and release.

1.7 Research Flow-Chart

The research flow-chart is summarized in Figure 1.1. The MSN is synthesized by hydrothermal method, and then modified by APTES and Al. Different approach of synthesis method, which is microwave, is also conducted. All synthesized MSN is then subjected to characterization, and tested for ibuprofen adsorption and release.

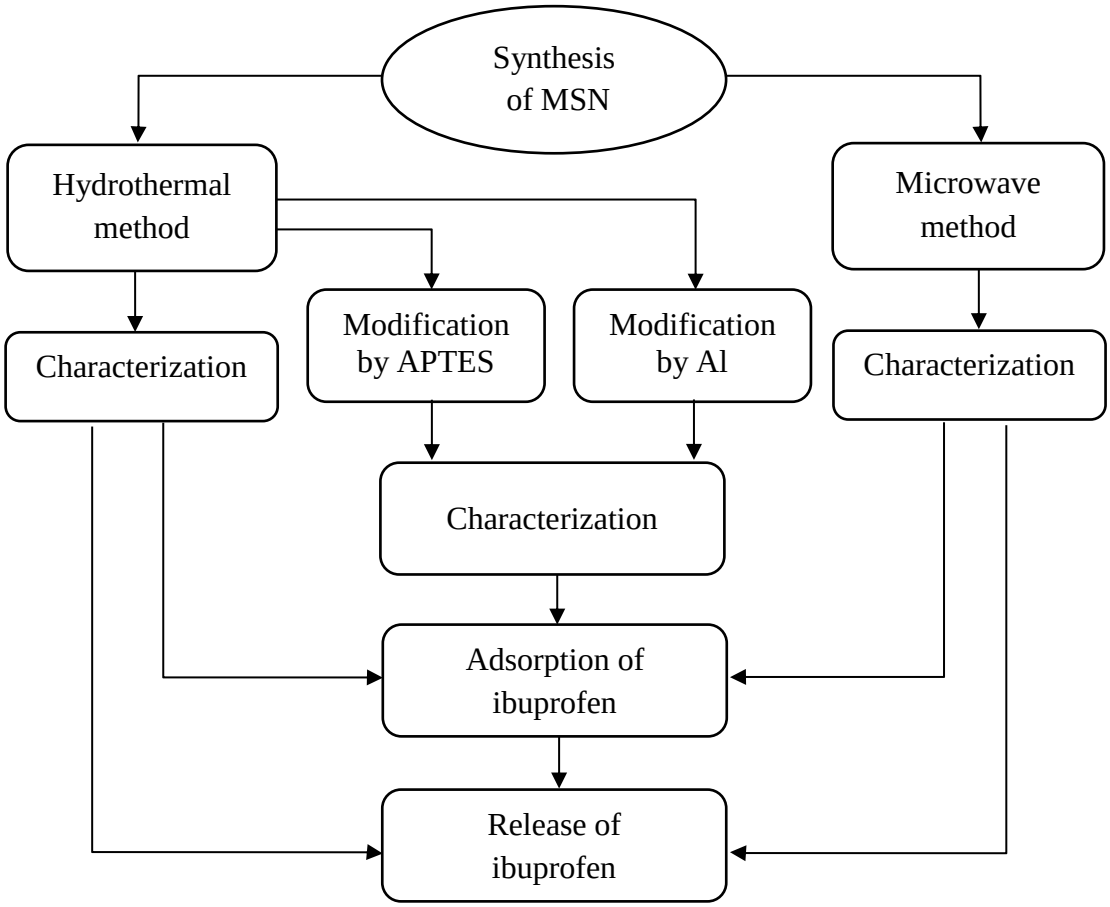

Figure 1.1 Research flow chart.

1.8 Thesis outline

This thesis was divided into five chapters. Chapter 1 described the general introduction of the study, problem statement and hypothesis, research objectives, scope and significant of research. This chapter brief describes the demand of the research for synthesis of mesoporous silica materials for the application towards drug delivery system. The general introduction is about the importance of MSN modification, as it offers room for improvement and enhancement of the adsorption and release of drugs. The conventional preparation methods of catalyst were also emphasized and the potential of microwave as a different synthesis medium was also highlighted. Problem statement of the current research was addressed to provide clear objectives of the present study and the scope of study covers the research work that will be conducted to meet these objectives.

Chapter 2 covers the background of drug delivery, the utilization of nanomaterials, and modification which had been explored previously in drug delivery application. Chapter 3 describes the particulars of the materials and chemical reagents used in the present work, the procedure for catalyst preparation and modification along with all the characterization studies. The next part presents the adsorption and release of ibuprofen study.

In Chapter 4, results and discussion was divided by characterization study and drug delivery performance of MSN, MSN modified by 3-aminopropyltriethoxysilane, MSN modified by Al metals, and MSN synthesized by microwave. All results and proposed mechanism were presented and discussed comprehensively. Finally, Chapter 5 covered the conclusions about the study. The recommendations for future studies were also given in this chapter.

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